

Highlights:

- 4D quantification of growing clinopyroxene and plagioclase crystals
- Temperature and time control the nucleation of pyroxene and plagioclase in basalts
- Investigation of the crystal effect on the rheology and transport of basaltic lava
- Reproduction of the final solidification history of the 2001 Etna lavas
- Application of real-time synchrotron X-ray imaging to forecast lava flow hazards

***In situ* quantification of crystallisation kinetics of plagioclase and
clinopyroxene in basaltic magma: implications for lava flow**

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21 **Abstract**

22 Crystallisation is a complex process that significantly affects the rheology of magma, and
23 thus the flow dynamics during a volcanic eruption. For example, the evolution of crystal
24 fraction, size and shape has a strong impact on the surface crust formation of a lava flow, and
25 accessing such information is essential for accurate modelling of lava flow dynamics. To
26 investigate the role of crystallisation kinetics on lava flow behaviour, we performed real-
27 time, *in situ* synchrotron X-ray microtomography, studying the influence of temperature-time
28 paths on the nucleation and growth of clinopyroxene and plagioclase in an oxidised,
29 nominally anhydrous basaltic magma. Crystallisation experiments were performed at
30 atmospheric pressure in air and temperatures from 1250 °C to 1100 °C, using a bespoke high-
31 temperature resistance furnace. Depending on the cooling regime (single step *versus*
32 continuous), two different crystal phases (either clinopyroxene or plagioclase) were
33 produced, and we quantified their growth from both global and individual 3D texture
34 analyses. The textural evolution of charges suggests that suppression of crystal nucleation is
35 due to changes in the melt composition with increasing undercooling and time. Using
36 existing viscosity models, we inferred the effect of crystals on the viscosity evolution of our
37 crystal-bearing samples to trace changes in rheological behaviour during lava emplacement.
38 We observe that under continuous cooling, both the onsets of the pāhoehoe-‘a‘ā transition
39 and of non-Newtonian behaviour occur within a shorter time frame. With varying both
40 temperature and time, we also either reproduced or approached the clinopyroxene and
41 plagioclase phenocryst abundances and compositions of the Etna lava used as starting
42 material, demonstrating that real-time synchrotron X-ray tomography is an ideal approach to
43 unravel the final solidification history of basaltic lavas. This imaging technology has indeed
44 the potential to provide input into lava flow models and hence our ability to forecast volcanic
45 hazards.

46

47 **Keywords**

48 X-ray microtomography, Basalt, Crystallisation, Lava flow

49

50 **1. Introduction**

51 Nucleation and growth of crystals are complex processes that dramatically affect the
52 evolution of textures in cooling magmas, and therefore the eruptive dynamics. These
53 processes have been extensively studied, but mainly *ex situ* using laboratory cooling and
54 decompression experiments (e.g. Mollo and Hammer, 2017, for a review; Giuliani et al.,
55 2020, and references therein) and through the textural analysis of both experimental and
56 eruptive products from two-dimensional (2D) and three-dimensional (3D) methods (e.g.
57 Baker et al., 2012, and references therein). Until very recently, there was a paucity of *in situ*
58 data (Ni et al., 2014; Schiavi et al., 2009). Thanks to the emerging developments of X-ray
59 synchrotron imaging techniques and *in situ* apparatus (e.g. Azeem et al., 2017; Drakopoulos
60 et al., 2015), we can now directly capture the progressive development of a crystal texture in
61 4D (Arzilli et al., 2019; Polacci et al., 2018; Tripoli et al., 2019), i.e. three spatial dimensions
62 plus time, which provides a deeper understanding of magma crystallisation and solidification
63 during flow emplacement.

64 Pāhoehoe and ‘a‘ā are the two most common surface morphologies in basaltic lava flows,
65 with smooth and rough textures, respectively (e.g. Wentworth and Macdonald, 1953;
66 Peterson and Tilling, 1980). Most lavas initially erupt as pāhoehoe and may evolve to ‘a‘ā
67 during the progress of the flow. This irreversible transition (e.g. Hon et al., 2003) occurs
68 dominantly in response to the increase in viscosity that, in turn, is controlled by the chemical

evolution of the residual melt, oxygen fugacity conditions and the increase in crystallinity with distance from the vent (e.g. Cashman et al., 1999; Peterson and Tilling, 1980; Vona et al., 2011). Thus, pāhoehoe lavas are typically less crystalline (<25–30 vol% vs >35 vol%) and less viscous than ‘a‘ā lavas.

Here, we studied and quantified *in situ* the nucleation and growth kinetics of pyroxene and plagioclase, the two most common phenocrysts in basaltic magmas. These phenocrysts were produced during single-step and continuous cooling experiments, to evaluate the effect of varying both temperature and time on crystal texture. Specifically, we monitored the evolution of the volume fraction, size and shape of clinopyroxene and plagioclase phenocrysts. We also determined the rheology of the crystal-bearing samples for an accurate modelling of lava flow behaviour. Polacci et al. (2018) and Arzilli et al. (2019) partially presented the single-step cooling experiments. Polacci et al. (2018) quantified the disequilibrium crystallisation of clinopyroxene and titanomagnetite using an empirical model accounting for the volume fraction of crystals, while Arzilli et al. (2019) show that rapid (in minutes) crystallisation can trigger magma fragmentation in highly explosive basaltic eruptions. In this work, we present new data that enables us to (1) quantify the 3D growth of individual clinopyroxenes and plagioclases, (2) infer the evolving rheology of the crystal-bearing samples, and (3) compare the crystallisation kinetics induced under different cooling pathways. As such, this work contributes to a better understanding of the final solidification history of lava flows at basaltic volcanoes.

2. Material and Methods

2.1. Starting material

A sample suite from the Lower Vents (LV, 2500 m and 2100 m vents) of the 2001 eruption of Mt. Etna, Italy (ET01, Table 1), was used as the starting material for the *in situ* cooling experiments. The erupted trachybasaltic lava (47.7 wt% SiO₂, 5.3 wt% Na₂O+K₂O, Corsaro et al., 2007) contains phenocrysts of clinopyroxene (8–14 vol%), plagioclase (4–7 vol%), olivine (1–3 vol%) and titanomagnetite (less than 1 vol%). Most of the clinopyroxene crystals are subhedral and sub-millimeter in size; others range from 3 to 10 mm. Plagioclase crystals are euhedral and sub-millimeter in size; millimeter crystals are rare (Corsaro et al., 2007).

The rock sample was crushed, placed in a platinum (Pt) crucible and melted in a 1 atm Nabertherm MoSi₂ box furnace at 1400 °C for 4 h in air. The process was repeated twice. The melt was then quenched by pouring it onto a steel plate, before being cored into glass cylinders measuring 2.8 mm in diameter × 4 mm in length. The composition of the anhydrous starting glass was measured by Electron probe microanalysis (EPMA) (Table 1).

2.2. Experimental approach

Two types of cooling-driven crystallisation experiments were carried out, defined by either (1) a large drop in temperature followed by a dwell time of 4 h at the target temperature (*single-step cooling* SSC) or (2) a continuous temperature decrease at a constant rate (*continuous cooling* CC). These experiments were performed at the I12 beamline of the Diamond Light Source (DLS, Harwell campus, United Kingdom) using the Alice high temperature (max 1460 °C) resistance furnace with controlled heating/cooling rate (max 24 °C/min; Azeem et al., 2017). The cylindrical samples were contained in alumina (Al₂O₃) holders (inner diameter 3 mm, wall thickness 0.5 mm) that were fitted on top of an Al₂O₃ rod attached to the rotation stage, as shown in the sketch in Fig. 1. During the experiments, the

temperature was monitored to ± 5 °C by two type R thermocouples placed close to the sample holder.

All experiments were performed in air, at ambient pressure and a temperature ranging between 1100 °C and 1250 °C to simulate lava flows at near-vent conditions. The experimental procedure (Fig. 2, Table 2) consisted of an initial constant heating (at 24 °C/min) of the sample up to 1250 °C, which is above the liquidus temperature, and a 30 min isothermal hold to ensure the sample was completely melted and homogeneous. The sample was then either cooled as rapidly as possible (24 °C/min) to 1170 °C (experiment SSC1170) or 1150 °C (experiment SSC1150) and held at constant temperature for 4 h, or cooled at a controlled rate of 0.5 °C/min to 1100 °C (experiment CC0.5), before cooling down to room temperature (at 24 °C/min). The choice of a cooling rate of 0.5 °C/min is within the range of estimated rates at which basaltic lava flows cool (0.01–15 °C/min, Cashman et al., 1999; Witter and Harris, 2007).

During cooling-induced crystallisation, synchrotron X-ray tomography was conducted in phase-contrast mode with a detector-sample distance of 2300 mm, in order to visualise and analyse in real-time the nucleation and growth of crystals. X-ray tomographic images were acquired using a 53 keV monochromatic X-ray beam and a high-speed camera, providing a field of view of 8×7 mm² and a pixel size of 3.2 µm. In each scan dataset, a series of 1800 projections were acquired over a 180° rotation with an exposure time of 0.05 s per projection. For each crystallisation experiment, up to 80 datasets were captured at a temporal resolution of 3 min per scan, from before the end of sample melting/homogenisation at 1250 °C to the final cooling down to room temperature.

2.3. Analytical methods

2.3.1. Textural analysis

Diamond Light Source in-house scripts derived from the Python-based framework TomoPy (Gursoy et al., 2014) were used to reconstruct the tomographic datasets, including flat-field correction, centre determination (Vo et al., 2014) and the removal of ring artefacts (Titarenko et al., 2010) and blobs (Vo et al., 2018).

The reconstructed images were then processed using ImageJ (Abramoff et al., 2004, Fiji Version 1.0) and Avizo (ThermoFisher Scientific, Version 9.4) software. ImageJ was first used to reduce the size of the images to 8-bit, adjust brightness and contrast to highlight the phenocrysts, and crop a Volume of Interest (VoI, $692 \times 726 \times 606$ – 814 voxels) that contains either clinopyroxene or plagioclase crystals. The VoI was chosen to exclude the top air/melt interface meniscus and the trapped air bubbles at the bottom and edges of the sample container (Fig. A.1a). All subsequent analysis was performed in Avizo (Fig. A.2). Noise reduction was achieved by successively applying bilateral and median filters in 3D, in order to efficiently segment crystals.

Crystals were segmented with interactive thresholding, and successively dilating, eroding and/or opening the segmented volumes, as well as removing small particles (mainly oxides). A large part of the oxides had to be removed manually due to the difficulty of automatically identifying and separating them from the clinopyroxene and plagioclase phenocrysts. Three scan datasets per experiment (9 in total) were selected, representing three different steps of the crystallisation process at $t_1 = 60$ min, $t_2 = 160$ min and $t_3 = 200$ min after the start of the dwell/cooling time (t_0).

In addition to quantify the mineral abundances (Table 2 and Fig. 3), a total of 10 clinopyroxenes in SSC1150 and 10 plagioclases in CC0.5 (Fig. A.3), located in various places in the VoI, were isolated to measure their size (3D length L_{3D}) and volume (V), in

order to assess their growth rate in non-volumetric (Y_{L3D}) and volumetric (Y_V) terms (Figs. 4–6, Tables 2, A.2 and A.3). Changes in crystal length/width aspect ratios (r_p) were also quantified over time, with both the crystal length (Length3D) and width (Width3D) automatically calculated as the longest and shortest Feret diameters, respectively. Most of the segmented crystals were connected, corresponding to aggregates of single crystals (Polacci et al., 2018), so no crystal number density is given.

2.3.2. Chemical analysis

Run products were analysed for their chemistry. Glass, titanomagnetite, clinopyroxene and plagioclase crystals were analysed by EPMA using a Jeol JXA 8530F electron microprobe at the University of Manchester, UK. Analyses were performed using a 15 kV accelerating voltage, 10 nA beam current and beam size of 10 μ m. See Polacci et al. (2018) for more details on the procedure.

3. Results

3.1. Mineral assemblages and crystal abundances

All experimental charges contain crystals of titanomagnetite (Table 2), confirming their oxidised nature. They appeared visible in the melt after 12 and 6 min of the dwell time in the SSC1170 and SSC1150 experiments (Polacci et al., 2018), respectively, and after 60 min of continuous cooling in the CC0.5 experiment (although up to t_2 and reaching 1170 °C the oxide volume fraction is near-zero). The two single-step cooled samples (SSC1170, SSC1150) also contain clinopyroxene phenocrysts of augite composition (Table A.1), while the continuously cooled sample (CC0.5) is characterised by the presence of plagioclase and

the absence of clinopyroxene. In both SSC1170 and SSC1150, plagioclase was not able to form after 4 h at constant temperature. It started to grow only in the final cooling to room temperature, at $T = 1131\text{--}1053\text{ }^{\circ}\text{C}$ for SSC1170 and $T = 1112\text{--}1073\text{ }^{\circ}\text{C}$ for SSC1150 (Arzilli et al., 2019).

In SSC11170, the volume fraction of clinopyroxene is systematically very low ($<0.002\text{ vol\%}$), with only three distinguishable crystals in the segmented VoI (Fig. 3a–c). They are part of a small number of clinopyroxenes present at the bottom of the sample holder, which appeared after 30 min of dwell time (Polacci et al., 2018) on the surface of an entrapped air bubble and in close contact with alumina (Fig. A.1c). Since no other clinopyroxene crystals nucleated in the rest of the charge within 4 h, the temperature of $1170\text{ }^{\circ}\text{C}$ seems to be very close to the liquidus temperature. Consequently, we defined the liquidus temperature of clinopyroxene at $1170\text{ }^{\circ}\text{C}$.

In SSC1150, the nucleation of clinopyroxene crystals started within the first 12 min of the dwell period (Polacci et al., 2018), preferentially at the air-melt interface and on alumina walls (Fig. A.1b), i.e. heterogeneously. The clinopyroxene volume fraction increases with time, from $1\text{ vol\%}$ at $t_1 = 60\text{ min}$ up to $9\text{ vol\%}$ at $t_3 = 200\text{ min}$ (Fig. 3d–f), as well as with reducing temperature by $20\text{ }^{\circ}\text{C}$, from $1170\text{ }^{\circ}\text{C}$ to $1150\text{ }^{\circ}\text{C}$ (Fig. 3a–f).

In CC0.5, we did not observe any phenocryst crystallisation within the first hour of the experiment. Plagioclase crystals started to nucleate and grow at $\sim 1205\text{ }^{\circ}\text{C}$, i.e. after 90 min from the beginning of cooling. They formed on tiny air bubbles attached to the walls of the container. Afterwards, the volume fraction of plagioclase increased with increasing time and decreasing temperature, reaching $22\text{ vol\%}$ at $t_3 = 200\text{ min}$ and $T = 1150\text{ }^{\circ}\text{C}$ (Fig. 3g–i). Clinopyroxene crystallisation did not occur during the continuous cooling at $0.5\text{ }^{\circ}\text{C/min}$. A

few tiny crystals were observed in the final unquenched product, suggesting that they formed somewhere during final cooling at 24 °C/min, at temperatures below 1100 °C.

In all three experiments, crystals nucleate mostly between t_1 and t_2 (Fig. 3) and then grow with time and/or cooling, i.e. growth is dominant from t_2 onwards.

3.2. Crystal sizes and growth rates

Compared to previous studies, here we provide individual *in situ* 3D measurements of crystal growth kinetics, contributing to a better understanding of crystallisation processes and, as a consequence, of the solidification of basaltic lava flows.

Clinopyroxene size (L_{3D}) nearly triples with increasing the duration of the SSC1150 experiment, ranging from 90–320 μm at t_1 (60 min dwell) to 150–890 μm at t_2 (160 min dwell) to 250–1090 μm at t_3 (200 min dwell, Fig. 4a, Tables 2 and A.2). Plagioclase L_{3D} doubles with mutually increasing time and decreasing temperature in CC0.5, ranging from 270–520 μm at t_2 to 445–1160 μm at t_3 (Fig. 4b, Tables 2 and A.3). Both the clinopyroxene and plagioclase volumes, averaged over 10 single crystals, increased fivefold from 0.3 to 1.6 $\times 10^{-6} \text{ cm}^3$ and from 0.8 to 3.8 $\times 10^{-6} \text{ cm}^3$, respectively, during the 140 min hold/cooling from t_1 to t_3 (Fig. 4, Tables 2, A.2 and A.3). The average clinopyroxene growth rate (Y_{L3D}) is nearly constant with time, ranging between $6.9 \times 10^{-6} \text{ cm/s}$ (t_1) and $5.8 \times 10^{-6} \text{ cm/s}$ (t_3), while the average clinopyroxene volumetric growth rate (Y_V) almost triples between $4.6 \times 10^{-11} \text{ cm}^3/\text{s}$ (t_1) and $12 \times 10^{-11} \text{ cm}^3/\text{s}$ (t_3) (Fig. 5, Tables 2 and A.2). Both the average Y_{L3D} and Y_V of plagioclase crystals are more impacted by time and temperature, with Y_{L3D} almost quadrupling (3.7 to 16 $\times 10^{-6} \text{ cm/s}$) during 40 min of cooling from t_2 to t_3 , and Y_V increasing by a factor of 10 (4.1 to 55 $\times 10^{-11} \text{ cm}^3/\text{s}$, Fig. 5, Tables 2 and A.3).

233

234 3.3. *Crystal morphologies*

235 In SSC1150, clinopyroxene crystals formed with a gradual transition from elongated
236 prismatic shapes to multi-branched crystals as growth proceeds (Fig. 4a; Polacci et al., 2018;
237 Arzilli et al., 2019). Spherulitic morphologies were also observed, corresponding to
238 heterogeneously nucleated crystal aggregates similar to that reported by Arzilli et al. (2015)
239 for alkali feldspars. Plagioclase crystals from the CC0.5 run product exhibit an elongate
240 bladed habit (Fig. 4b). No change in morphology was observed with increasing time and
241 decreasing temperature, although crystals become more elongated and branched. We
242 observed a morphological difference in the iron oxides that exhibit an equant shape under
243 single-step cooling (in SSC1150) and a skeletal form under continuous cooling (in CC0.5).

244

245 4. Discussion

246 4.1. *Single-step versus continuous cooling: differences in mineralogical assemblages and* 247 *kinetics*

248 4.1.1. *Mineralogical differences*

249 Depending on the type of cooling (single-step vs continuous), we noticed the presence or
250 absence of clinopyroxene and plagioclase. Clinopyroxene is present in both single-step
251 experiments, although in a very small amount in SSC1170, from near the beginning of the
252 dwell period (after 12 min dwell in SSC1150, Polacci et al., 2018), while plagioclase remains
253 absent until the final cooling stage (Arzilli et al., 2019), occurring as skeletal crystals. On the
254 contrary, only plagioclase crystallised from the melt on continuous cooling (CC0.5), with the
255 occurrence of minor clinopyroxenes (too small for analysis by EPMA) during the final

cooling to room temperature. For comparison, the equilibrium assemblage calculated by
MELTS software (Ghiorso and Sack, 1995; Asimow and Ghiorso, 1998), at ambient pressure
and fugacity (magnetite-hematite MH redox buffer), comprises both plagioclase ($T_{\text{liquidus}} =$
1208 °C) and clinopyroxene ($T_{\text{liquidus}} = 1188$ °C). As discussed above (section 3.1), the
texture of the SS1170 charge suggests that the liquidus temperature of clinopyroxene is closer
to 1170 °C. This enabled us to determine the nucleation undercooling ($\Delta T = T_{\text{liquidus}} -$
 $T_{\text{experimental}}$) as a function of temperature: for clinopyroxene $\Delta T = 0$ °C at 1170 °C and 20 °C
at 1150 °C, for plagioclase $\Delta T = 0$ °C at 1220 °C, 38 °C at 1170 °C and 58 °C at 1150 °C.
Both the SSC1170 and SSC1150 experiments were thus run below the predicted plagioclase
liquidus, meaning that plagioclase should have been present within the 4-h dwell. Similarly,
we performed part (80 min) of the CC0.5 experiment below the clinopyroxene liquidus
without nucleating clinopyroxene crystals. In each cooling regime, the nucleation of one
crystal phase is suppressed, attesting to disequilibrium crystallisation.

The absence of either plagioclase or clinopyroxene in cooling-induced crystallisation
experiments, while it should be thermodynamically stable, was reported previously by some
authors (e.g. Corrigan, 1982; Gibb, 1974; Lofgren, 1983; Vetere et al., 2015). They mainly
attribute the failure of these phases to nucleate to the degree of undercooling (a large ΔT , or
high cooling rate, inhibiting the nucleation of plagioclase), a limited experimental duration
and a low number of heterogeneous nucleation sites. In our SSC1150 single-step experiment,
the nucleation of clinopyroxene followed a rapid (24 °C/min) temperature drop that drove a
rapid increase of undercooling to 20 °C for clinopyroxene and to 58 °C for plagioclase, and
brought the melt to sub-liquidus conditions where both phases can grow. However, although
the nucleation of clinopyroxene was activated after 12 min at constant temperature,
plagioclase was not able to crystallise within the 4 h dwell. This indicates first that time plays

a major role in the evolution of crystallisation, and second that clinopyroxene needs lower activation energy to nucleate, associated with its lower number of tetrahedral units (Al–O and Si–O bonds) and less polymerised structure in comparison to plagioclase, which in turn translates into a shorter delay in nucleation (Iezzi et al., 2008). In addition, following these relatively fast nucleation and growth, the residual melt became depleted in clinopyroxene components (Ca, Mg, Fe, Table A.1), possibly delaying the stabilization of plagioclase nuclei. In contrast, and despite its more complex structure, the slow cooling rate of 0.5 °C/min enabled the nucleation of plagioclase. This suggests that suppression of crystal nucleation is not simply related to structural control alone, as previously suggested by Corrigan (1982). Following the nucleation and growth of plagioclase crystals near the liquidus temperature, the residual melt became depleted in plagioclase components (anorthite: Ca and Na, Table A.1), creating a local disequilibrium that may have delayed the formation of clinopyroxene. Nucleation sites for clinopyroxene may also be missing. They may have been suppressed because of a longer time spent above their liquidus temperature (122 min for CC0.5 instead of 3 min for SSC1150, from the end of melting/homogenisation) or occupied by early-formed plagioclase crystals.

Our observations show that the cooling rate and the resulting increase of undercooling through time are important factors controlling crystallisation. In this sense, Vetere et al. (2015) documented that plagioclase is only able to crystallise at low to moderate cooling rates (< 3 °C/min). However, Arzilli et al. (2019) observed the crystallisation of plagioclase at a high cooling rate of 24 °C/min. The difference may be related to initial conditions, since superheating, as applied by Vetere et al. (2015), can promote an efficient dissolution of pre-existing nuclei (e.g. Arzilli et al., 2015), while a time-step at sub-liquidus conditions can promote the formation of stable nuclei, which once subjected to a rapid thermal perturbation of the system are able to grow rapidly. The effects of thermal history on the mineralogical

assemblage of cooling basalts appear to be not yet well understood, stressing that more systematic real-time investigations should be performed.

4.1.2. Crystal nucleation kinetics

The experimental charges have higher crystal abundances at the gas-melt and container-melt interfaces because of the lower surface free energy required for nucleation. At the container walls, the heterogeneous nucleation of plagioclase and clinopyroxene crystals is associated with the presence of tiny air bubbles and the early formation of Al–Mg oxides (Fig. A.4), both acting as nucleation sites. The presence of pre-existing nuclei is an important mechanism that contributes to the solidification of basaltic lava flows (e.g. Mollo et al., 2015), emphasising that *in situ* observations can unravel the dynamics of heterogeneous crystal nucleation and its important implications on the development of basaltic textures, and so on conduit processes and eruptive dynamics.

In addition to being reduced by heterogeneous nucleation, the nucleation delay, i.e. the time between when the melt is brought below its liquidus and the onset of nucleation (Rusiecka et al., 2020), is reduced as a function of undercooling. As expected with increasing undercooling and/or experimental duration (e.g. Rusiecka et al., 2020 and references therein; Vona and Romano, 2013), the nucleation of clinopyroxene occurs faster and the volume fraction of crystals increases (Fig. 3). The plagioclase-free texture of the SSC1150 sample, during the 4-h dwell, is also consistent with the findings of Rusiecka et al. (2020).

In contrast to previous *in situ* research (Ni et al., 2014) that reports the nucleation of olivine or clinopyroxene crystals only in a short event of a few minutes at the beginning of the experiment, we observed multiple nucleation events up to t_2 (160 min dwell/cooling) and the

continuous growth of both clinopyroxene (SSC1150) and plagioclase crystals (CC0.5). These multiple nucleation events, reported as two main nucleation/growth pulses in SSC1150 (at 60 and 150 min dwell) in Polacci et al. (2018), may be induced by changes in melt composition with further effects on the liquidus temperature, following the nucleation and growth of first crystals. Such multiple nucleation events have been recently reported by Giuliani et al. (2020) to account for the formation of clinopyroxene in basaltic melt.

4.1.3. Crystal growth kinetics

In the time interval of our experiments, clinopyroxene and plagioclase crystals grew at rates of 10^{-6} cm/s and 10^{-5} – 10^{-6} cm/s, respectively. A clinopyroxene growth rate of 10^{-6} cm/s is similar to, or higher than, the one reported in previous works on anhydrous basaltic and trachybasaltic melts (10^{-6} – 10^{-9} cm/s, Baker, 2008; Burkhard, 2005; Giuliani et al., 2020; Ni et al., 2014; Orlando et al., 2008; Pontesilli et al., 2019). Lower growth rates of 10^{-7} – 10^{-9} cm/s have been reported for plagioclase crystals (Arzilli et al., 2015; Giuliani et al., 2020; Orlando et al., 2008). These differences are associated with the use of *in situ* data, since we measured dynamic growth rates that are known to decrease with increasing dwell time (e.g. Pontesilli et al., 2019), with impingement by surrounding crystals. *Ex situ*, growth rates are determined for entire experimental periods and are thus averaged and possibly underestimated. In addition, the use of 2D data provides less accurate shape information.

At constant undercooling ($\Delta T = 20$ °C), the clinopyroxene growth rate by size (Y_{L3D}) is nearly constant through time, and the volumetric growth rate (Y_v) slightly increases (Fig. 5). With increasing undercooling by 20 °C, from 38 °C to 58 °C, both the Y_{L3D} and Y_v plagioclase growth rates quadruple (Fig. 5). Kirkpatrick (1975) also showed an increase of the rate of crystal growth with increasing undercooling for a large temperature range.

Following Tamman (1899) and as predicted by the theory (Kirkpatrick, 1975), we can expect the rate of crystal growth to be near-zero at the liquidus, increase to a maximum and then decrease with increasing undercooling.

The initially individual clinopyroxene and plagioclase crystals grew along thermodynamically favourable directions, i.e. perpendicular to the surface of heterogeneities (Al-walls of the sample holder, air bubbles and first-nucleated crystals, including many oxides), forming networks of agglomerated crystals (Fig. 3). Clinopyroxene crystals grew radially from the top of the VoI and the edge walls of the alumina container, mainly by branching and aggregation (Fig. 3d–f). Plagioclases also align along preferential directions, perpendicular to the sample holder (Fig. 3g–i). The euhedral textures developed by both the clinopyroxenes and titanomagnetites in SSC1150 result from interface-controlled growth mechanisms, which are favoured by a small undercooling of 20 °C and a high temperature of 1150 °C that is close to the liquidus temperature of 1170 °C. In contrast, the plagioclase blades observed in CC0.5 indicate some disequilibrium between the melt and crystals, linked to larger undercoolings and departure from their liquidus temperature (1208 °C). The shape of these rapidly growing crystals is controlled by diffusion, with a slow diffusivity of cations in the melt (Mollo and Hammer, 2017). Attainment of equilibrium did not occur during the course of the continuous cooling experiment (~5 h) for plagioclase. In that experiment, the oxide crystals are skeletal, which also attests to their development under conditions of rapid growth and high degrees of undercooling.

Aggregation of smaller and mutually touching crystals was reported to occur in Etnean pāhoehoe and ‘a‘ā lavas at syn-eruptive conditions over the effect of undercooling (e.g. Lanzafame et al., 2013). This underlines the close reproducibility of the near-surface crystallisation of basaltic melts by our innovative *in situ* approach and thus its utility in lava flow studies.

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378 4.1.3. Rheology of the crystal-bearing samples

379 Growing crystals increase the viscosity of the suspending basaltic lava and introduce non-
380 Newtonian rheology (e.g. Giordano et al., 2008; Mueller et al., 2010; Vona et al., 2011),
381 which in turn affects its transport (e.g. Soldati et al., 2016). Assessing the rheological
382 evolution of crystallising basaltic melts is thus required for lava flow modelling, to predict
383 how the lava flow will evolve in time and space. Here, we infer the evolving rheology of our
384 SSC1150 and CC0.5 crystal-bearing samples from a Hershel-Bulkley model (Herschel and
385 Bulkley, 1926; Mader et al., 2013):

$$\tau = \tau_0 + K\dot{\gamma}^n, \quad \text{Eq. 1}$$

386 where τ is the shear stress, τ_0 is a yield stress, K is the consistency, $\dot{\gamma}$ is the strain-rate and n
387 is the flow index. The rheological impact of the crystal phase depends primarily on the ratio
388 of the crystal volume fraction ϕ to the maximum crystal packing fraction ϕ_m (ϕ/ϕ_m)
389 because τ_0 , K and n are all functions of ϕ/ϕ_m (Mader et al., 2013). ϕ_m can be calculated
390 from the aspect ratio of the crystals r_p (Mader et al., 2013; Mueller et al., 2010):

$$\phi_m = 0.55 \exp \left[-\frac{(\log_{10} r_p)^2}{2} \right]. \quad \text{Eq. 2}$$

391 Both ϕ and r_p are measured directly from our *in situ* data (Section 2.3.1). SSC1150 contains
392 clinopyroxene crystals with average aspect ratios of 4.07 (60 min dwell), 7.41 (160 min
393 dwell) and 7.35 (200 min dwell, Tables 2 and A.2), and CC0.5 contains plagioclase crystals
394 with average r_p of 4.24 (160 min cooling) and 5.76 (200 min cooling, Tables 2 and A.3).
395 Consistency and flow index are calculated following Mader et al. (2013):

$$K = \mu_0 \left(1 - \frac{\phi}{\phi_m}\right)^{-2}, \quad \text{Eq. 3}$$

$$n = 1 - 0.2r_p \left(\frac{\phi}{\phi_m}\right)^4, \quad \text{Eq. 4}$$

396 where μ_0 is the viscosity of the melt phase, which can be calculated as a function of
 397 composition and temperature (Giordano et al., 2008). At 1170 °C and 1150 °C, the viscosities
 398 of melt samples are respectively of 2.01 and 2.18 log Pa s. Computed values of ϕ_m , the ratio
 399 ϕ/ϕ_m , K and n are tabulated in Table 2.

400 Mader et al. (2013) show that τ_0 is negligible for $\phi/\phi_m < 0.8$, which is the case for all of our
 401 tomographic reconstructions. In this case, Equation 1 reduces to a power law in which
 402 suspension viscosity η depends on strain rate:

$$\eta = K\dot{\gamma}^{n-1}. \quad \text{Eq. 5}$$

403 They further argue that shear thinning effects can be neglected for most strain rates of
 404 practical interest if $n \geq 0.9$, which is the case for most of our tomographic reconstructions. In
 405 this case, $\eta \equiv K$ and the Newtonian suspension viscosity can be calculated directly using
 406 Equation 3.

407 Results (Tables 2, A2 and A3) show that our samples exhibit a Newtonian behaviour during
 408 both the dwell and continuous cooling, with small ϕ/ϕ_m of 0.02 at t_1 to 0.24 at t_3 in SSC1150
 409 and of 0.20 at t_1 to 0.53 at t_3 in CC0.5, respectively. However, higher values of this ratio
 410 (0.64 in SSC1150 and 0.58 in CC0.5) are expected after more than the 200 min
 411 dwell/cooling, by considering a maximum ϕ of 0.24 as found in the LV magmas (Corsaro et
 412 al., 2007). With more time, our two samples produced under different temperature-time
 413 conditions, would thus evolve towards a non-Newtonian behaviour, with consequences for
 414 magma transport (see Section 4.2).

415 Magma viscosity is also dependent on shear rate, which facilitates the nucleation and growth
416 of crystals (Vona and Romano, 2013). During transport and emplacement, magmas and lavas
417 undergo a range of shear rates ($2.5\text{--}0.001\text{ s}^{-1}$, Kolzenburg et al., 2018 and references therein).
418 Although we did not investigate the effect of varying the deformation rate on the rheological
419 evolution of crystallising basaltic melts, previous experimental studies (Kolzenburg et al.,
420 2018; Tripoli et al., 2019; Vona et al., 2011; Vona and Romano, 2013) showed that shear
421 deformation accelerates crystallisation kinetics, resulting in shorter nucleation delays and
422 higher crystal fractions.

423 In more detail, Vona et al. (2011) performed *ex situ* isothermal crystallisation experiments
424 while capturing the temporal evolution of the viscosity of anhydrous Etna basalt of the 1991–
425 1993 eruption deformed under a low strain rate of 0.53 s^{-1} . In air, at $1157\text{ }^{\circ}\text{C}$ and 1 atm,
426 crystallisation takes place within 29–45 h, associated with an increase in apparent viscosity to
427 a stable value of 3.07 log Pa s ($\phi = 0.16$). The final mineralogical assemblage is composed of
428 spinel (minor) and plagioclase, which differs from our SSC1150 experiment, where
429 clinopyroxene nucleated after a short 12-min delay. Their starting melt composition slightly
430 differs from ours, but not enough to justify such a difference in crystallisation time, as well as
431 in the order of nucleation of crystal phases. We suggest, instead, that these differences can be
432 explained by a longer time spent above the liquidus temperature (5h at $1400\text{ }^{\circ}\text{C}$ in Vona et
433 al., 2011 vs 30 min at $1250\text{ }^{\circ}\text{C}$ in our study), which allows a more complete dissolution of
434 pre-existing nuclei. These differences can also be explained by the use of a different
435 apparatus (concentric cylinder viscometer) equipped with a spindle that causes chemical
436 heterogeneities and mechanical perturbations in the melt (Vona et al., 2011). Recent findings
437 (e.g. Tripoli et al., 2019; Kolzenburg et al., 2020) demonstrated that in the absence of thermal
438 and/or mechanical fluctuations the nucleation of plagioclase is not favoured, as observed in
439 our single-step experiments.

440 Using the model of Vona et al. (2011), which considers the magma strain rate and both the
 441 crystal fraction and crystal shape factors (Vona and Romano, 2013), we calculated the
 442 relative viscosity of our SSC1150 suspensions (liquid + crystal, considering both
 443 clinopyroxene and oxide phases). As expected (Vona et al., 2011 and references therein), the
 444 relative viscosity increases with increasing crystal fraction ϕ , from 1.21 log Pa s at $\phi = 0.05$
 445 (t_1 , Table 2) to 2.39 log Pa s at $\phi = 0.14$ (t_2 , Table 2) to 3.37 log Pa s at $\phi = 0.17$ (t_3 , Table 2).
 446 According to Polacci et al. (2018), a stable clinopyroxene crystal content (~14 vol%) can be
 447 reached after ~500–1500 min dwell (~8–25 h) in the SSC1150 sample, which translates to a
 448 higher relative viscosity of 6.93 log Pa s at $\phi = 0.22$ (including a stable oxide content of 8
 449 vol%, Arzilli et al., 2019).

450 Kolzenburg et al. (2018) were among the first to study the effect of shear rate at non-
 451 isothermal conditions, which can be related to our CC0.5 continuous cooling experiment.
 452 They observed that the onset temperature of crystallisation increases with increasing shear
 453 rate, from 1101 °C (no shear) to 1164 °C and 1178 °C for shear rates of 1.16 and 4.64 s⁻¹,
 454 respectively, at a cooling rate of 0.5 °C/min and using a trachybasalt from the 2013 eruption
 455 of Etna. However, in CC0.5 crystallisation occurs at high temperatures (~1205 °C) without
 456 introducing shear, suggesting that results can hardly be compared with each other. Here
 457 again, differences can be due to the use of a concentric cylinder apparatus with a longer time
 458 spent above the liquidus temperature (several hours at 1300 °C) and more reduced conditions,
 459 and a slightly different starting melt composition. Kolzenburg et al. (2018) also showed that
 460 increasing shear rate increases the cutoff temperature (T_{cutoff} , i.e. the point at which the
 461 sample rheologically solidifies and flow stops), which implies shorter flow distances.
 462 Although it is not relevant to our data, if deformation is taken into account, transitions
 463 between different eruptive styles or flow mechanisms can occur in a shorter amount of time
 464 (Vona and Romano, 2013).

465

466 4.2. Implications for basaltic lava flows

467 4.2.1. Time evolution of magma crystallinity: the pāhoehoe-‘a‘ā transition

468 As described above, our results can be used to investigate the effect of cooling and
469 crystallisation on the rheological properties of lava, and more specifically to retrieve the
470 conditions under which pāhoehoe lavas transform to ‘a‘ā. Such a transition is greatly
471 influenced by the viscosity of the lava, which is dependent on temperature and crystallinity,
472 and associated with the development of yield strength (Cashman et al., 1999). A critical
473 crystallinity range at which pāhoehoe transforms to ‘a‘ā has been identified between 0.18 and
474 0.35 (Hon et al., 2003; Soule et al., 2004), although some Etnean pāhoehoe lavas (termed
475 “cicirara”) can contain up to 40 vol% crystals (e.g. Lanzafame et al., 2013). We inferred such
476 parallel changes of crystallinity and rheological behaviour (onset of non-Newtonian
477 behaviour) after more than 200 min dwell/cooling at a comparable crystallinity of 0.24, as
478 found in the 2001 trachybasalts. In addition, a critical viscosity range for the pāhoehoe-‘a‘ā
479 transition has been calibrated from a combination of field observations and laboratory
480 experiments, from 4 log Pa s (no shear rate) to ~2.7 log Pa s with increasing shear rate (e.g.
481 Cashman and Mangan, 2014, and references therein; Sehlke et al., 2014; Soldati et al., 2017).
482 In line with this finding, such viscosities are approached in our single-step cooling conditions
483 (SSC1150) after 200 min and largely exceeded within 8 h. Under our given P - T - fO_2 -
484 composition conditions, the transition of basaltic lava from pāhoehoe to ‘a‘ā can thus be
485 crossed in ca. 4 h from emission under continuous cooling, or between 3 and 8 h under
486 single-step cooling.

487

4.2.2. Crystallisation timescales in basaltic lava flows and the case study of the 2001 eruption of Mt. Etna

Under single-step cooling (near-isothermal) conditions, our data coupled with the empirical model of Polacci et al. (2018) indicate that 14 vol% of crystals (single-phase) can nucleate and grow over a day. This amount alone is not enough to bring a major shift in rheological behaviour, implying that lava can travel great distances with minimal cooling. Under continuous cooling (non-isothermal) conditions, our results demonstrate that the growth of 22 vol% plagioclase crystals in just over 3 h would soon have a significant impact on flow behaviour. According to Kolzenburg et al. (2020), the lava could solidify and stop flowing after reaching 1128 °C, i.e. after 244 min (~4 h) of continuous cooling at 0.5 °C/min. During flowage onto the surface, the behaviour of basaltic lavas appears to be greatly influenced by both temperature and time, with crystal phases possibly nucleating and growing under different cooling regimes, as explored below.

As a trachybasalt from the 2001 Etna eruption was used as the starting material for our experimental simulations of lava flow emplacement, results can aid in the interpretation of the final crystallisation history of the erupted LV lavas. According to Lanzafame et al. (2013) who unravelled the solidification path of a pāhoehoe lava from Mt. Etna through petrographic analysis, crystals with a size <1 mm (~75 vol%) crystallised during lava emplacement. Our results agree well with this finding, since the SSC1150 single-step cooled sample crystallised the amount of clinopyroxene phenocrysts found in the 2001 lavas (~9–14 vol%) within 200 to 500–1500 min of dwell (Polacci et al., 2018) at 1150 °C. After that time, a critical crystallinity (of maximum 24 vol%) could be reached with the appearance of a second phenocryst phase (6–7 vol% plagioclase) in the same proportions as in the 2001 LV products. For comparison, in the CC0.5 sample, continuously cooled at a rate of 0.5 °C/min, 4 to 7 vol% plagioclase crystals appeared in less than 160 min at $T \geq 1170$ °C. However, 40 min

later ($T = 1150\text{ }^{\circ}\text{C}$) CC0.5 already contained 22 vol% plagioclase and no clinopyroxene, which differs strongly from the LV products. In addition to reproduce the abundance of clinopyroxene crystals in the SSC1150 experiment, we reproduced their composition with an average Mg number (Mg#) of 72 vs of 73 in the LV products (Fig. 6). According to Mollo et al. (2015), the Mg_{54-74} clinopyroxene population in Etna products formed within the range of our experimental conditions, i.e. at 0.1–500 MPa and 1050–1175 $^{\circ}\text{C}$. LV textures are thus very well reproduced by our single-step experiment at 1150 $^{\circ}\text{C}$, meaning first that the interior of lava flows in the 2001 eruption may have cooled almost isothermally, and second that the nucleation of clinopyroxene may have occurred in the shallowest portion of the Etnean plumbing system and during lava transport at the surface. However, this excludes the few larger (3–10 mm) clinopyroxene crystals present in the LV lavas, which cannot be reproduced by our experiments, and that result from crystallisation at depth during storage (Mollo et al., 2015 and references therein), as well as olivine crystals that nucleated and grew under water-saturated conditions at high pressures and temperatures (Mollo et al., 2015). The nucleation of plagioclase seems to have happened differently. According to La Spina et al. (2016), the low plagioclase content in the 2001 LV products indicates a fast (<1 h) vertical ascent from depth (9 km) to the surface. Plagioclase crystals might have nucleated and grew during ascent under non-equilibrium conditions, and not during flow advance, driven by magma degassing (Mollo et al., 2015). In accordance with it, both the lower range of plagioclase core compositions and rim compositions (An_{64} , Corsaro et al., 2007) were reproduced in the SSC1150 experiment (An_{67} , Fig. 6 and Table A.1), implying that plagioclases in the LV trachybasalts did not nucleate outside of the conduit, although their growth was affected by subaerial conditions. The interior of the 2001 lava flows could thus result from a combination of pre-, syn- and post-eruptive crystallisation.

As demonstrated with our study, working *in situ* enables the solidification of a basaltic lava flow to be directly observed, and so reproduced with varying both time and temperature. This opens new perspectives to unravel the solidification path of basaltic lavas, especially as only a small amount of phenocrysts could equilibrate at depth, with most of the crystallisation happening during magma ascent in the conduit and emplacement to the surface (Lanzafame et al., 2013; Giuliani et al., 2020), which are conditions that we can simulate with our *in situ* equipment.

5. Conclusions

Our results from single-step *versus* continuous cooling experiments show that cooling rate and undercooling have a strong influence on the nucleation of clinopyroxene and plagioclase, with implications for the behaviour of basaltic lavas. According to our 3D time-dependent observations combined with published rheological data, the interior of the 2001 lava flows of Etna may have crystallised up to 14 vol% clinopyroxene phenocrysts almost isothermally over a day. Such a nucleation and growth of crystals during lava flow emplacement accompanies changes in rheological behaviour, which affect lava flow length and hazard severity.

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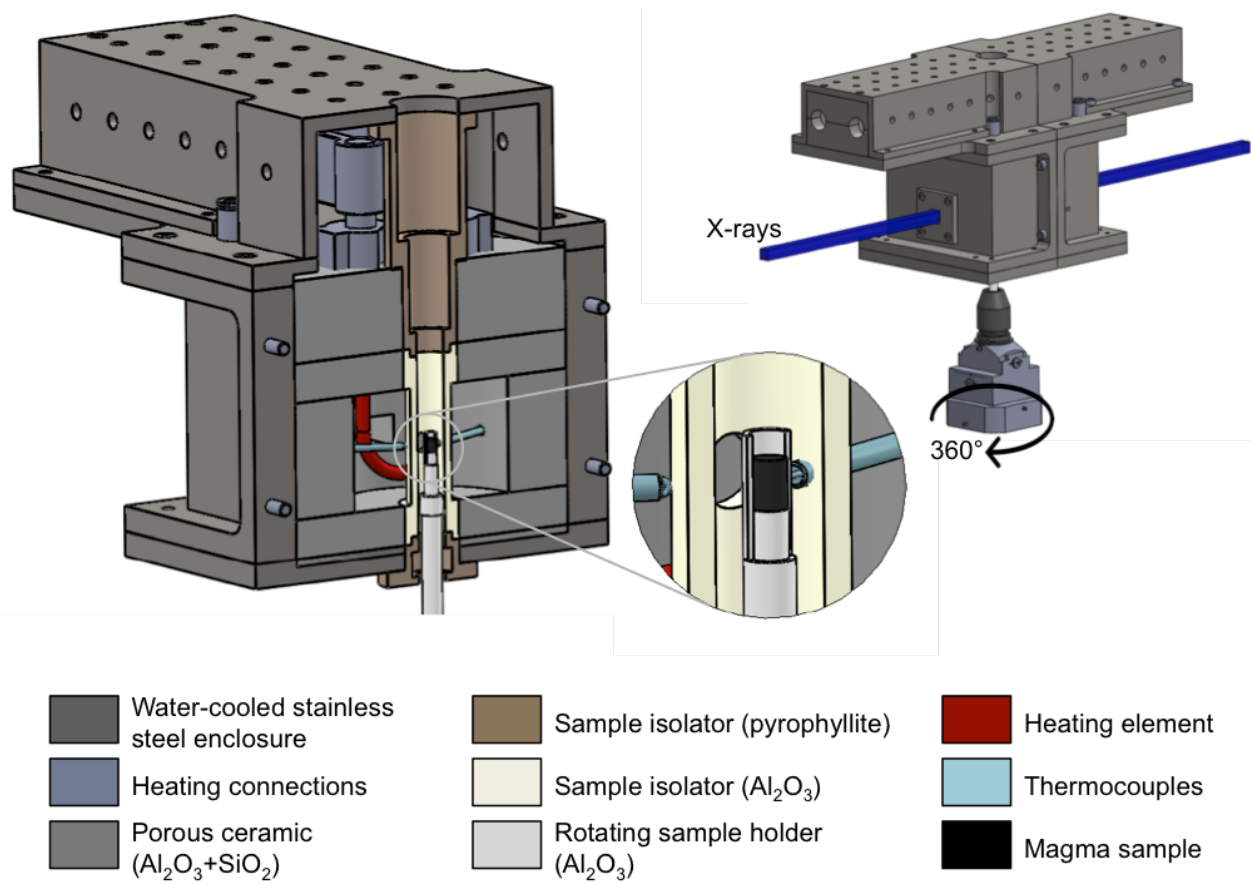
Figure 1. Sketch of the experimental setup and *in situ* resistance furnace

Figure 2. Temperature-time paths followed by the three charges (SSC1170, SSC1150, CC0.5) of this study. t_0 : start of the dwell time/cooling, $t_1 = 60$ min dwell/cooling, $t_2 = 160$ min dwell/cooling and $t_3 = 200$ min dwell/cooling. The small (± 5 – 10 °C) temperature fluctuations are due to sample rotation during image acquisition.

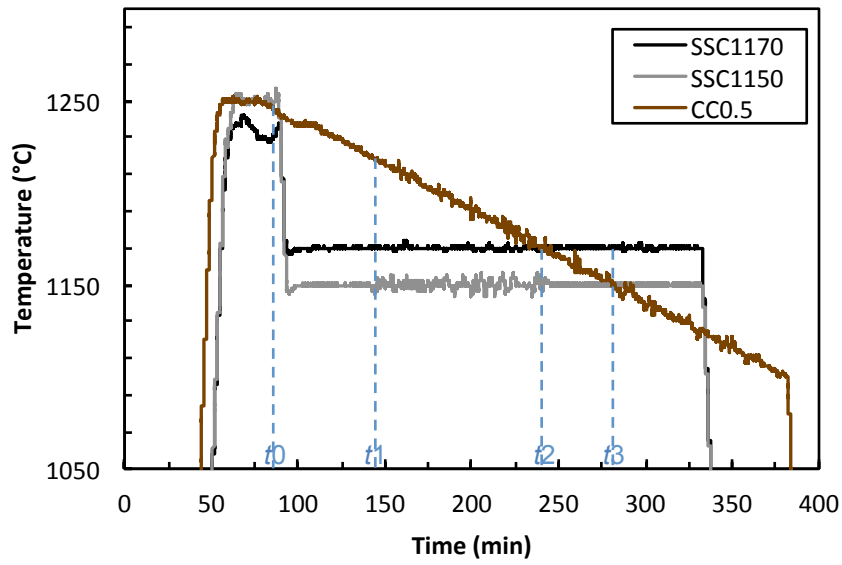


Figure 3. 3D volume renderings of charges SSC1170 (a–c), SSC1150 (d–f) and CC0.5 (g–i) through time, at $t_1 = 60$ min (a, d, g), $t_2 = 160$ min (b, e, h) and $t_3 = 200$ min (c, f, i) after the start of the dwell/cooling time (t_0). In each volume rendering, an orthoslice of the unsegmented dataset is shown in addition to segmented clinopyroxene (Cpx, in light blue) and plagioclase (Plg, in yellow) crystals. The volume fraction of crystals is given in %. Ox: Fe–Ti oxides.

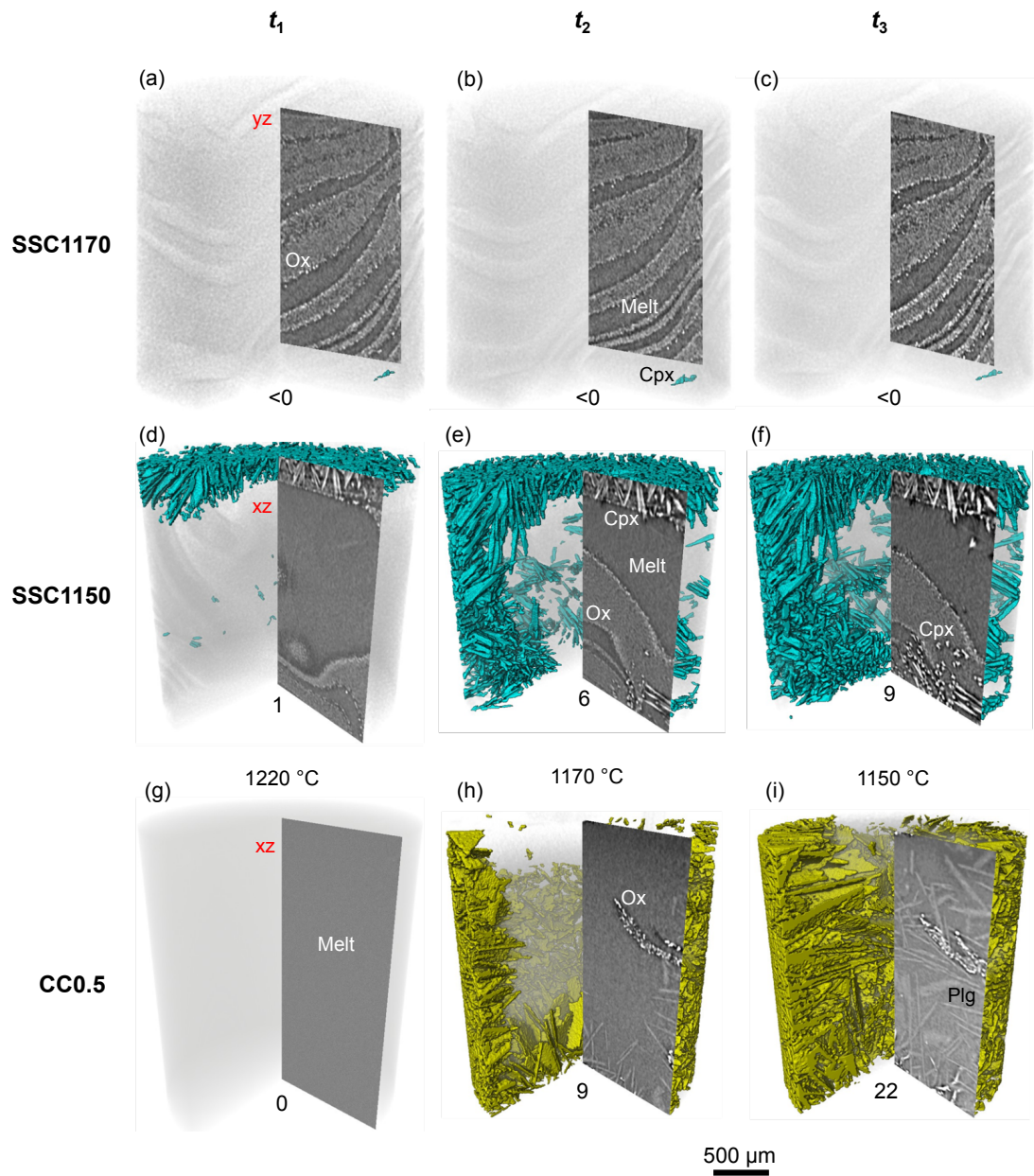


Figure 4. Evolution of (a) clinopyroxene and (b) plagioclase sizes (3D crystal length L_{3D} and volume V) as a function of time and temperature in charges SSC1150 and CC0.5, respectively; t_1 , t_2 , t_3 = 60, 160, and 200 min dwell/cooling time, respectively. Error bars correspond to the standard deviation of the 3D crystal length/volume of the 10 crystals measured in each sample.

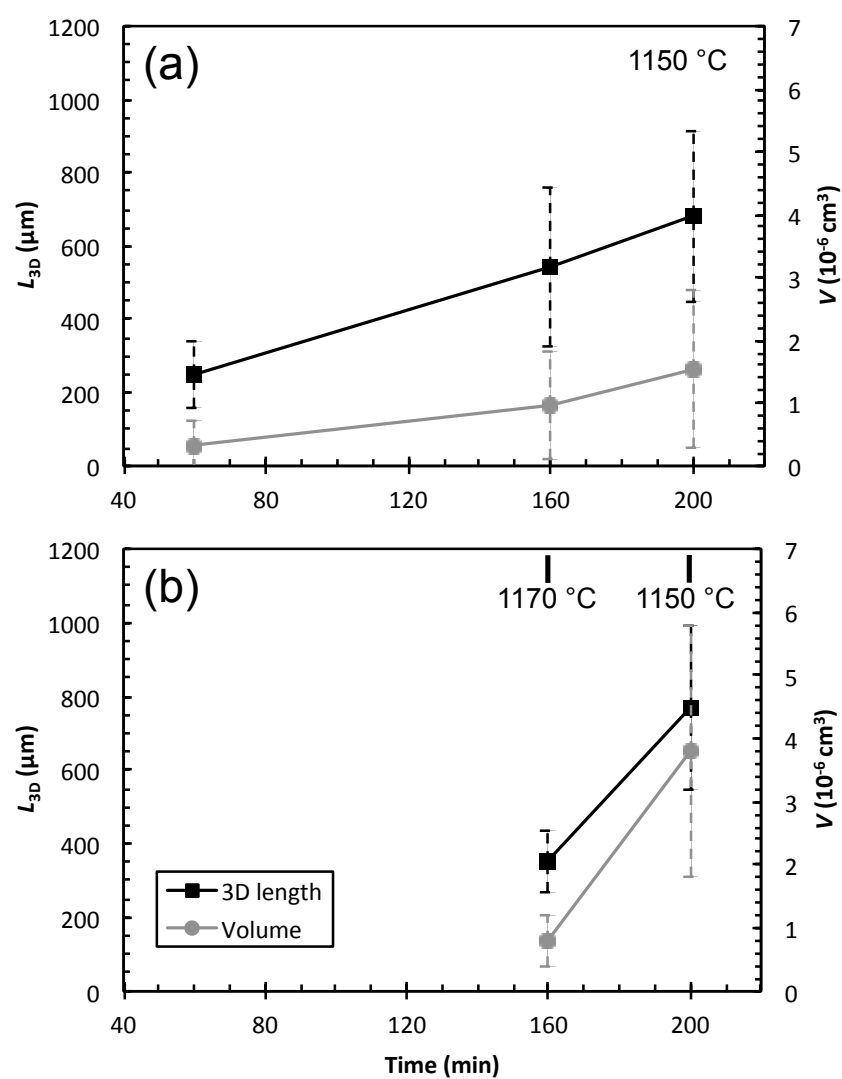


Figure 5. Evolution of crystal growth rates (Y_{L3D} and Y_V) as a function of time and undercooling (ΔT); t_1 , t_2 , t_3 = 60, 160, and 200 min dwell/cooling time, respectively. Cpx: clinopyroxene crystals in SSC1150; Plg: plagioclase crystals in CC0.5. Error bars represent the standard deviation of the 10 crystals measured in each charge at t_1 , t_2 and t_3 .

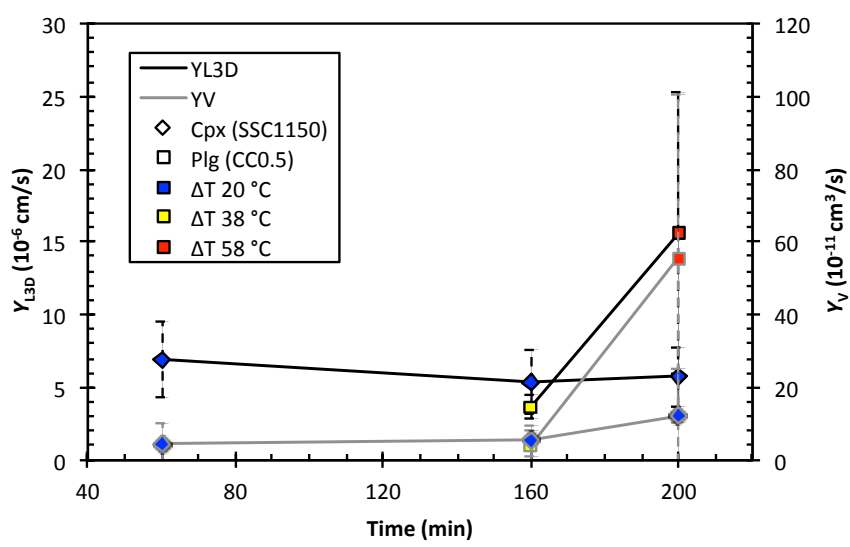


Figure 6. Experimental vs. natural crystal compositions. Data from experiments SSC1170, SSC1150 and CC0.5. Natural data from Corsaro et al. (2007). Mg# (atomic Mg / (Mg + Fe²⁺)) in clinopyroxene crystals; Anorthite content (100 × atomic Ca / (Ca + Na)) in plagioclase crystals.

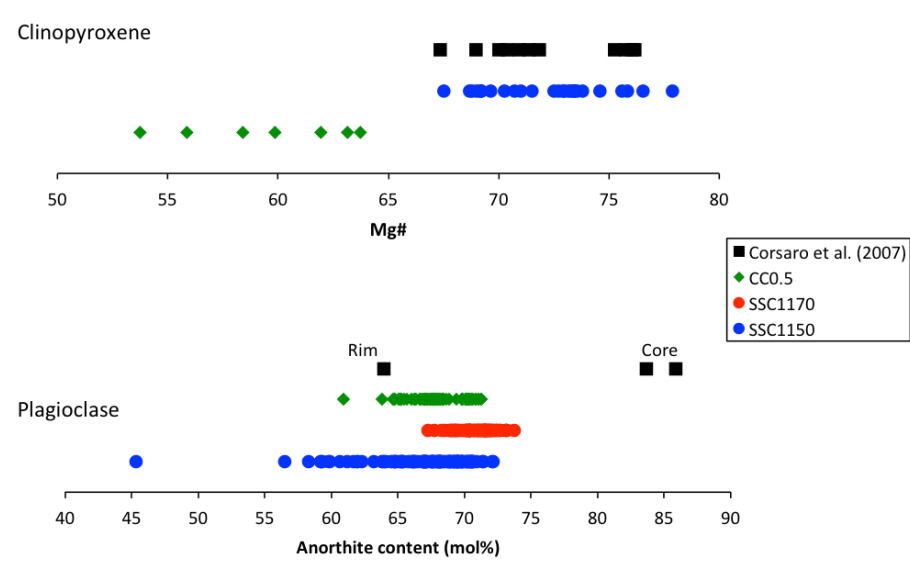


Table 1. Composition of ET01 Lower Vents (LV) volcanics and starting glass, and of phenocrysts

Label	ET01 ^a (<i>n</i> = 6)	Glass ^b (<i>n</i> = 23)	Cpx ^c (<i>n</i> = 4)	Plg ^c (<i>n</i> = 3)
SiO ₂	47.7 (3) ^d	48.4 (2)	48.7 (17)	48.5 (34)
TiO ₂	1.64 (2)	1.79 (7)	1.36 (48)	0.09
Al ₂ O ₃	16.7 (1)	16.2 (5)	4.87 (119)	33.3 (22)
Fe ₂ O ₃	1.77 (2)	nd	nd	nd
FeO	8.84 (11)	10.7 (3)	8.57 (35)	0.62 (15)
MnO	0.17 (0)	0.20 (3)	0.24 (5)	0.02
MgO	6.35 (16)	6.20 (32)	13.2 (11)	0.06 (1)
CaO	11.0 (1)	10.7 (2)	22.0 (7)	15.6 (26)
Na ₂ O	3.37 (6)	3.41 (20)	0.48 (9)	2.45 (1.30)
K ₂ O	1.89 (5)	1.90 (7)	nd	0.15 (19)
P ₂ O ₅	0.48 (1)	0.54 (4)	nd	nd
Total	99.2	99.85	99.2	100.8

^aWhole-rock composition (averaged and normalized to 100% anhydrous) of 2001 Lower Vents (LV) lava flow products, from Corsaro et al. (2007).

^bElectron microprobe analysis (normalized to 100% anhydrous) of ET01 starting glass, from Arzilli et al. (2019). All Fe reported as FeO.

^cElectron microprobe analyses of clinopyroxene (Cpx) and plagioclase (Plg) phenocrysts in 2001 LV volcanics, from Corsaro et al. (2007). All Fe reported as FeO.

^dOne standard deviation in terms of last digit unit cited.

n: number of analyses.

nd: not determined.

Oxides are in wt%.

Table 2. Experimental conditions and results

Charge	T_{init} (°C)	T_{exp} (°C)	R (°C/min)	dwelt time (min)	ΔT (°C)	Phase assemblage ^a	L_{3D} (cm)	V (cm ³)	Y_{L3D} (cm/s)	Y_V (cm ³ /s)
<i>Single-step cooling (SSC)</i>										
SSC1170	1250	1170	24	240	0	gl(92)+ox(8)+cpx(<0.1)	–	–	–	–
SSC1150	1250	1150	24	240	20	gl(83)+ox(8)+cpx(9)	6.8E–02 (2)	1.6E–06 (1) ^b	5.8E–06 (2)	1.2E–10 (1)
<i>Continuous cooling (CC)</i>										
CC0.5	1250	1100	0.5	–	≤108	gl(77)+plg(22)+ox(0.8)	7.7E–02 (2)	3.8E–06 (2)	1.6E–05 (1)	5.5E–10 (5)

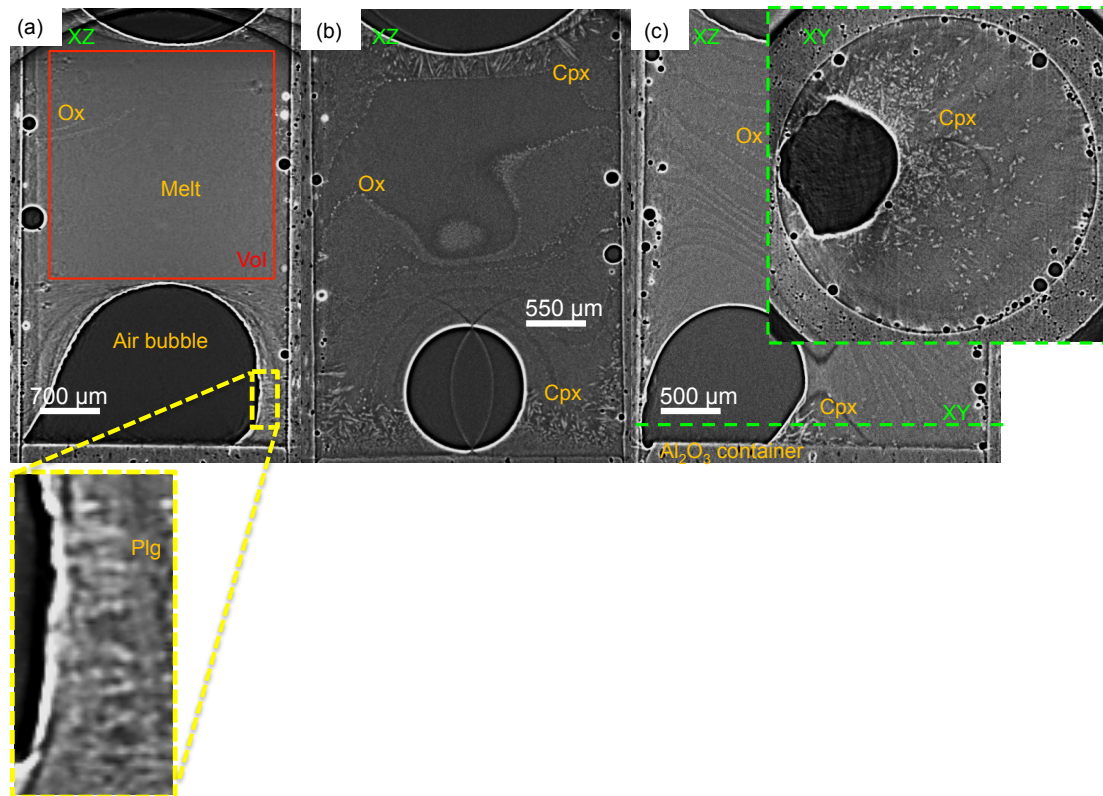
Note: T_{init} = initial (or melting) temperature; T_{exp} = experimental (or final) temperature; R = cooling rate; ΔT = undercooling degree, $T_{\text{liquidus}} - T_{\text{experimental}}$ with $T_{\text{liquidus}} = 1208$ °C for plagioclase and $T_{\text{liquidus}} = 1170$ °C for clinopyroxene; L_{3D} = crystal length; V = crystal volume; Y_{L3D} = crystal growth rate = L_{3D}/t_{exp} with t_{exp} the experimental duration (in s, [Arzilli et al., 2015](#)); Y_V = volumetric growth rate = $(V \times 0.5)/t_{\text{exp}}$ ([Arzilli et al., 2015](#)); r_p = crystal length/width aspect ratio; ϕ_m = maximum crystal packing fraction; ϕ/ϕ_m = ratio of the crystal volume fraction to the maximum crystal packing fraction, κ = consistency; n = flow index. All textural parameters are averaged over 10 selected crystals at t_3 (after 200 min of dwelt/cooling).

^aPhase proportions calculated by 3D image analysis at t_3 ; gl, glass; cpx, clinopyroxene; plg, plagioclase; ox, Fe–Ti oxide. In SSC1150, the oxide contents were calculated to be 4 vol% and 8 vol% at t_1 and t_2 , respectively.

^bStandard deviation of the mean value.

Charge	r_p	ϕ_m	ϕ/ϕ_m	κ	n
<i>Single-step cooling (SSC)</i>					
SSC1170	–	–	–	–	–
SSC1150	7.35 (3)	0.38	0.24	1.72	1
<i>Continuous cooling (CC)</i>					
CC0.5	5.76 (2)	0.41	0.53	4.60	0.91

Fig. A.1. Longitudinal (XZ) slices through the 3D volume renderings of charges (a) CC0.5, (b) SSC1150 and (c) SSC1170 showing the heterogeneous nucleation of crystals (Plg: plagioclase, Cpx: clinopyroxene) after 60 min of cooling/dwell (t_1). Ox: oxide. The volume of interest (VoI, red inset) selected for quantitative image analysis is depicted in (a). In (c) a transverse (XY) slice of the SSC1170 charge is also depicted.



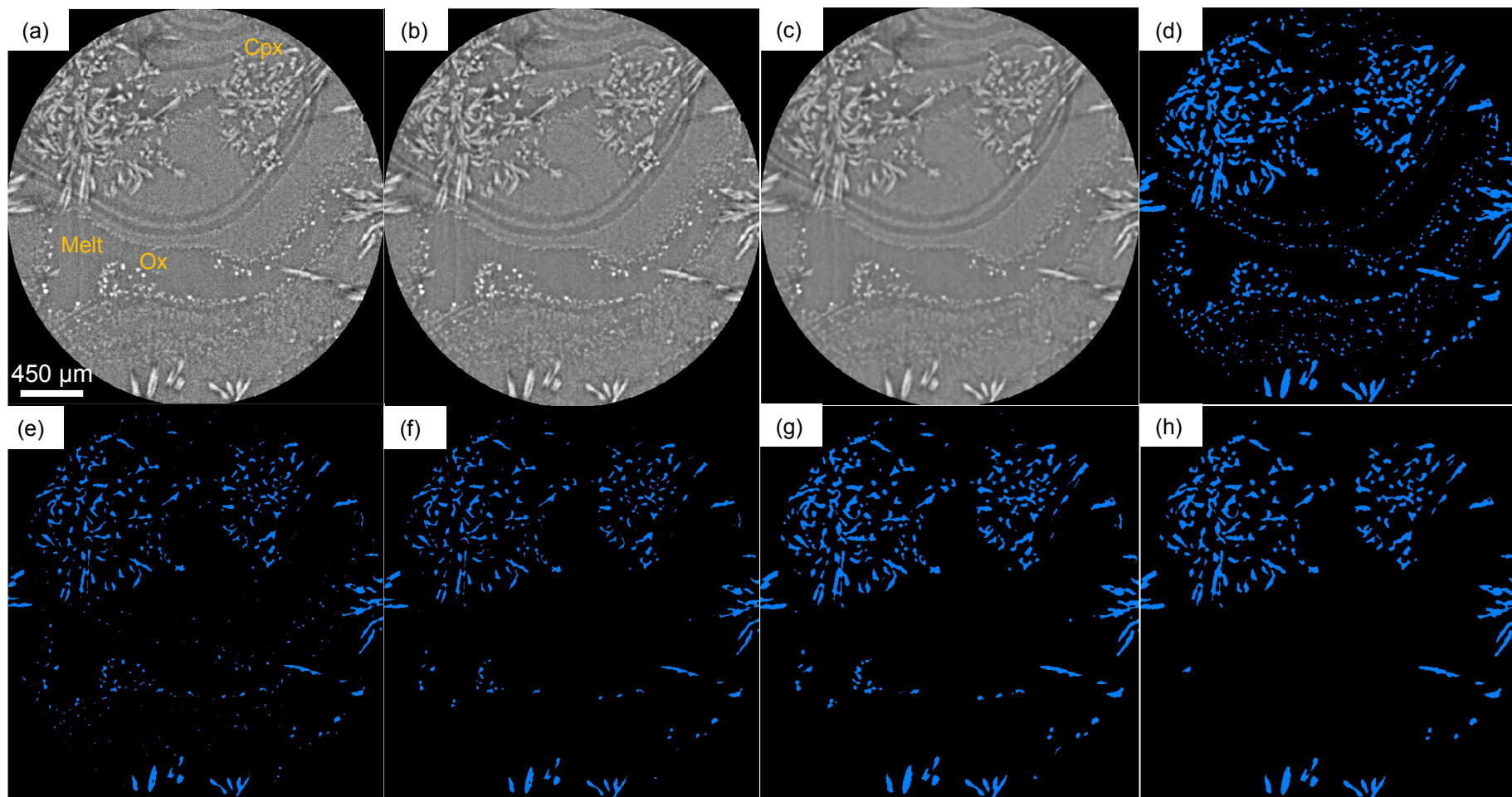


Fig. A.2. Overview of the image analysis procedure using Avizo. A transverse (XY) slice through the 3D volume rendering of the SSC1150 charge after 160 min of dwell (t_2) is shown for illustration. (a) Initial stage, after pre-processing with ImageJ; (b) After bilateral filtering; (c) After median filtering; (d) After interactive thresholding; (e) After erosion; (f) After removing small spots; (g) After dilation; (h) Final stage, after manual correction. Cpx: clinopyroxene; Ox: oxide.

Fig. A.3. Selected clinopyroxene (a) and plagioclase (b) crystals in samples SC1150 and CC0.5, respectively. Scale bars: 200 μm .

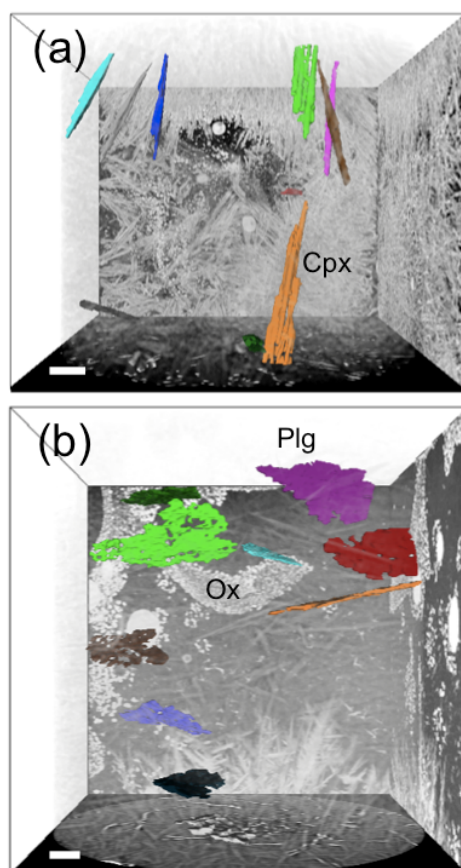


Fig. A.4. (a) SEM image of a test sample showing the formation of an oxide layer on the edge of an alumina container. The sample is a basalt from the Mt. Etna eruption of 2001 that has been left 5 hours in air, at 1 atm and 1190 °C before being air-quenched. (b) ESD analyses demonstrating that the oxides are made of Al and Mg. Ox: oxide; plg: plagioclase; cpx: clinopyroxene.

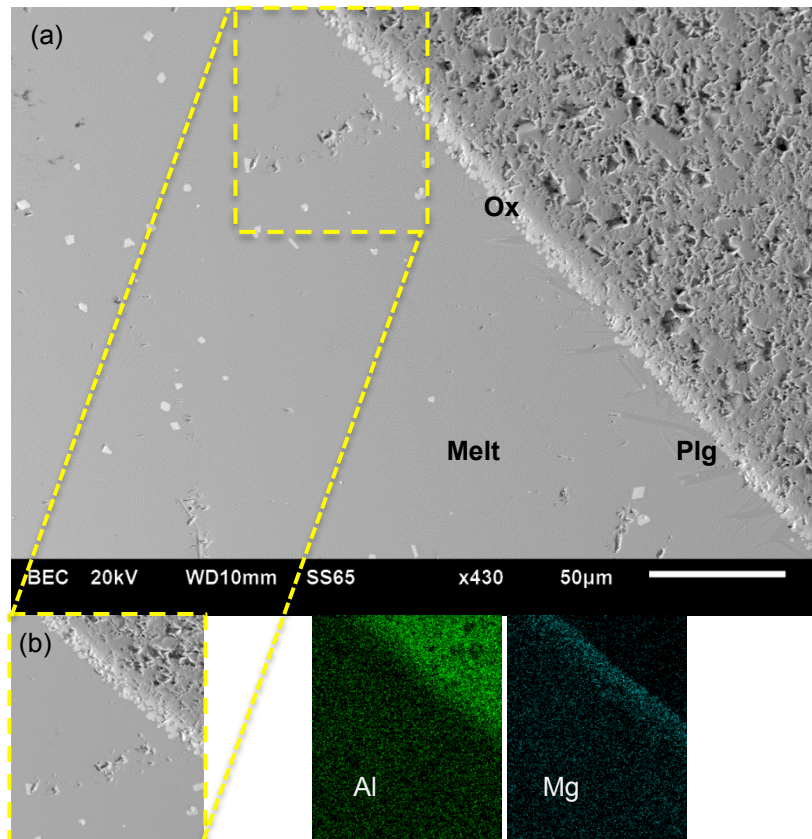


Table A.1. Experimental compositions

Charge	Phase	SiO ₂	TiO ₂	Al ₂ O ₃	FeO	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	Cr ₂ O ₃	NiO	Total	Mol%
SSC1170	gl ^a (3) ^b	50.6(4) ^c	1.85(6)	15.8(1)	8.52(11)	0.22(4)	6.25(1)	11.3(2)	3.41(9)	1.50(16)	0.52(7)	0.03(1)	0.03(1)	98.0	
SSC1150	gl(10)	53.4(4)	1.73(3)	18.1(2)	5.90(16)	0.18(1)	4.77(16)	7.97(24)	4.91(7)	2.84(10)	nd	0.01(0)	0.01(0)	96.2	
	cpx(26)	42.7(12)	1.88(29)	7.97(86)	9.21(69)	0.17(2)	13.3(8)	21.0(3)	0.47(3)	0.01(1)	nd	0.01(1)	0.01(1)	96.8	En ₄₅ Wo ₄₀
CC0.5	gl(8)	53.8(12)	2.11(20)	14.9(7)	7.13(88)	0.24(8)	5.37(56)	7.57(67)	4.49(58)	3.53(40)	0.85(17)	0.02(1)	0.02(0)	98.2	
	plg(69)	51.1(8)	0.14(4)	27.6(6)	2.99(28)	0.03(2)	0.22(10)	13.4(4)	3.49(24)	0.41(7)	0.05(5)	0.02(1)	0.02(1)	99.3	An ₆₈ Or ₇

^aGlass analyses normalized to 100% anhydrous, with all Fe as FeO.

^bNumber of microprobe analyses.

^cOne standard deviation in terms of last digit unit cited.

n: number of analyses.

nd: not determined.

gl, glass; cpx, clinopyroxene; plg, plagioclase. Enstatite En = 100 × at. Mg / (Mg + Fe + Ca); Wollastonite Wo = 100 × at. Ca / (Mg + Fe + Ca) in pyroxene, calculated with Fe = FeO; Anorthite An = 100 × at. Ca / (Ca + Na); Orthoclase Or = 100 × at. K / (K + Na) in plagioclase.

Table A.2. 3D clinopyroxene crystal measurements

Crystal	Time	L_{3D} (cm)	Y_{L3D} (cm/s)	V (cm ³)	Y_V (cm ³ /s)	r_p
cpx-01	t_1	0.032	9.0E-06	1.0E-06	1.4E-10	2.22
	t_2	0.063	5.1E-06	2.7E-06	1.4E-10	3.58
	t_3	0.069	2.4E-06	2.9E-06	3.0E-11	3.61
cpx-02	t_1	0.032	8.8E-06	2.4E-07	3.4E-11	5.31
	t_2	0.074	7.0E-06	9.5E-07	5.9E-11	12.6
	t_3	0.087	5.4E-06	1.5E-06	1.1E-10	11.1
cpx-03	t_1	0.026	7.2E-06	2.5E-07	3.5E-11	5.69
	t_2	0.049	3.8E-06	6.3E-07	3.1E-11	7.08
	t_3	0.071	9.2E-06	9.7E-07	7.2E-11	7.89
cpx-04	t_1	0.026	7.1E-06	1.2E-07	1.6E-11	—
	t_2	0.068	7.1E-06	6.4E-07	4.4E-11	14
	t_3	0.079	4.4E-06	9.7E-07	6.9E-11	12.4
cpx-05	t_1	0.009	2.5E-06	4.1E-08	5.8E-12	3.05
	t_2	0.036	4.4E-06	5.2E-07	4.0E-11	4.26
	t_3	0.05	6.1E-06	1.0E-06	1.1E-10	4.99
cpx-06	t_1	—	—	—	—	—
	t_2	0.089	9.2E-06	2.3E-06	1.2E-10	5.54
	t_3	0.109	8.4E-06	4.6E-06	4.9E-10	6.03
cpx-07	t_1	—	—	—	—	—
	t_2	0.062	6.5E-06	6.4E-07	3.3E-11	10.1
	t_3	0.079	6.9E-06	1.1E-06	1.1E-10	11
cpx-08	t_1	—	—	—	—	—
	t_2	0.055	5.7E-06	8.9E-07	4.7E-11	7.19
	t_3	0.064	4.0E-06	1.2E-06	7.3E-11	7.36
cpx-09	t_1	—	—	—	—	—
	t_2	0.032	3.3E-06	2.7E-07	1.4E-11	5.75
	t_3	0.047	6.3E-06	7.7E-07	1.0E-10	5.88
cpx-10	t_1	—	—	—	—	—
	t_2	0.015	1.5E-06	9.5E-08	5.0E-12	3.97
	t_3	0.025	4.4E-06	4.3E-07	7.0E-11	3.24

cpx, clinopyroxene; L_{3D} = 3D crystal length; Y_{L3D} = crystal growth rate; V = crystal volume; Y_V = volumetric crystal growth rate; r_p = crystal aspect ratio (= length/width ratio). t_1 , t_2 , t_3 = 60, 160, and 200 min dwell, respectively.

Note that at t_2 growth rates are calculated for the time interval between t_1 and t_2 and at t_3 the rates are calculated for the t_2 – t_3 time interval.

These single measurements are assumed to contain an error of 10% because of variation in the segmentation threshold.

Table A.3. 3D plagioclase crystal measurements

Crystal	Time	L_{3D} (cm)	Y_{L3D} (cm/s)	V (cm ³)	Y_V (cm ³ /s)	r_p
plg-01	t_2	0.052	5.4E-06	1.3E-06	6.7E-11	4.40
	t_3	0.101	2.0E-05	6.2E-06	1.0E-09	4.36
plg-02	t_2	0.045	4.7E-06	1.6E-06	8.2E-11	4.90
	t_3	0.058	5.5E-06	2.8E-06	2.6E-10	6.21
plg-03	t_2	0.038	4.0E-06	1.0E-06	5.2E-11	3.71
	t_3	0.070	1.3E-05	2.9E-06	4.0E-10	4.20
plg-04	t_2	0.035	3.7E-06	6.0E-07	3.1E-11	3.98
	t_3	0.065	1.2E-05	2.5E-06	3.9E-10	4.64
plg-05	t_2	0.034	3.6E-06	9.6E-07	5.0E-11	4.17
	t_3	0.059	1.0E-05	2.3E-06	2.8E-10	5.10
plg-06	t_2	0.031	3.2E-06	3.4E-07	1.8E-11	6.08
	t_3	0.090	2.5E-05	3.9E-06	7.5E-10	9.63
plg-07	t_2	0.028	2.9E-06	5.8E-07	3.0E-11	3.90
	t_3	0.045	6.9E-06	1.5E-06	1.9E-10	3.87
plg-08	t_2	0.028	2.9E-06	4.1E-07	2.1E-11	3.49
	t_3	0.116	3.7E-05	8.1E-06	1.6E-09	6.45
plg-09	t_2	0.027	2.8E-06	4.0E-07	2.1E-11	3.49
	t_3	0.072	1.9E-05	2.6E-06	4.5E-10	9.11
plg-10	t_2	—	—	—	—	—
	t_3	0.095	8.0E-06	4.9E-06	2.0E-10	3.98

plg, plagioclase; L_{3D} = 3D crystal length; Y_{L3D} = crystal growth rate; V = crystal volume; Y_V = volumetric crystal growth rate; r_p = crystal aspect ratio (= length/width ratio). t_1 , t_2 , t_3 = 60, 160, and 200 min cooling time, respectively.

Note that at t_2 growth rates are calculated for the time interval between t_1 and t_2 and at t_3 the rates are calculated for the t_2 – t_3 time interval.

These single measurements are assumed to contain an error of 10% because of variation in the segmentation threshold.

Credit Author Statement

M.P., M.R.B., F.A. and P.D.L. conceived the research project. All authors, N.L.G., F.A., G.L.S., M.P., B.C., M.E.H., N.T.V., R.C.A., D.D.G., S.N., E.W.L., M.R.B. and P.D.L. contributed to the beamline experiments. F.A. collected the volcanic rocks for the starting material. D.D.G. prepared the starting material. F.A., M.P., G.L.S. and N.T.V. performed image reconstruction. N.L.G. performed image processing with help from F.A. and M.P. N.L.G. performed the image segmentation and analysis. F.A. and M.E.H. performed chemical analysis. N.L.G. wrote the manuscript, with contributions from all other authors.