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- 1 Accepted version before minor changes;
- 2 Magma fragmentation in highly explosive basaltic eruptions induced by rapid crystallisation

- 4 Fabio Arzilli¹, Giuseppe La Spina¹, Mike R. Burton¹, Margherita Polacci¹, Nolwenn Le Gall²,
- 5 Margaret E. Hartley¹, Danilo Di Genova³, Biao Cai⁴, Nghia T. Vo⁵, Emily C. Bamber¹, Sara
- 6 Nonni⁵, Robert Atwood⁵, Ed W. Llewellin⁶, Richard A. Brooker⁷, Heidy Mader⁷ and Peter D. Lee²

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- 8 ¹School of Earth and Environmental Sciences, University of Manchester, Manchester M139PL, UK
- 9 ²Department of Mechanical Engineering, University College London, London, UK
- ³Institute of Non-Metallic Materials, Clausthal University of Technology, Zehntner Str. 2a, 38678
- 11 Clausthal-Zellerfeld, Germany
- ⁴ School of Metallurgy and Materials, University of Birmingham, Birmingham B15 2TT, UK
- 13 ⁵Diamond Light Source, Harwell Science and Innovation Campus, Didcot OX11 0DE, UK
- ⁶Department of Earth Sciences, Durham University, Durham DH1 3LE, UK
- ⁷School of Earth Sciences, University of Bristol, Bristol BS8 1RJ, UK

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- 18 *Corresponding author: Dr. Fabio Arzilli
- 19 Corresponding author present affiliation: School of Earth and Environmental Sciences, The
- 20 University of Manchester, Oxford Road, Manchester, M13 9PL, UK
- 21 E-mail address: fabio.arzilli@manchester.ac.uk
- 22 Phone: +393298429732; +447904104670

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Basaltic eruptions are the most common form of volcanism on Earth and planetary bodies. The low viscosity of basaltic magmas inhibits fragmentation, favouring effusive and lavafountaining activity, yet highly explosive, hazardous basaltic eruptions do occur. The processes that promote fragmentation of basaltic magma remain unclear, and are subject to debate. Here, we use a numerical conduit model to show that rapid ascent of magma during explosive eruption produces large undercooling. Novel in situ experiments reveal that undercooling drives exceptionally rapid (~minutes) crystallisation, inducing a step-change in viscosity that triggers magma fragmentation. Experimentally-produced textures are consistent with products of basaltic Plinian eruptions. We apply the numerical model to investigate basaltic magma fragmentation over a wide parameter space and find that all basaltic volcanoes have the potential to produce highly explosive eruptions. The critical requirements are initial magma temperatures lower than 1100 °C, in order to reach a syneruptive crystal content of > 30 vol.%, and thus a magma viscosity $\geq 10^5$ Pa s, which our results suggest is the minimum viscosity required for the fragmentation of fast ascending basaltic magmas. Our study provides both a demonstration and explanation of the processes that drive basaltic Plinian eruptions, revealing how typically effusive basaltic volcanoes can produce unexpected highly explosive, and hazardous, eruptions.

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Explosive eruptions are undoubtedly the most violent and catastrophic expression of volcanic activity^{1,2}. Highly explosive Plinian eruptions can inject > 0.1 km³ of pyroclastic material producing volcanic plumes that can reach up to 40 km in height, impacting both regional and global climate, and producing a significant threat to proximal populations^{3,4}. Basaltic volcanoes are usually characterised by effusive and mild-explosive lava fountaining activity⁵, however, basaltic Plinian eruptions can occur⁶⁻¹¹. The main process characterising highly explosive activity is magma fragmentation, which is the transition from ductile to brittle behaviour of molten magma^{1,2}. For silicic magmas, fragmentation is attributed to high strain rates associated with acceleration of the bubbly mixture^{1,12-14} or from the bubble overpressure associated with restricted bubble expansion^{14,15-18}. The low viscosity of basaltic magmas, however, makes high strain rates, or the bubble overpressure necessary to trigger fragmentation difficult to achieve^{1,19}.

Crystallisation during magma ascent may significantly increase magma viscosity, leading to fragmentation²⁰. However, crystallisation in basaltic magmas has previously been thought to occur on timescales significantly longer than the time required for magma to ascend from a crustal storage chamber to the vent²¹, particularly in the case of highly explosive eruptions.

In volcanic conduits, the crystallisation kinetics of an ascending magma are driven by degassing and cooling²²⁻²³. Plagioclase and pyroxene crystallisation are sensitive indicators of magma dynamics in volcanic conduits^{21,24-27} and their kinetically controlled abundance can rapidly change magma rheology²⁸⁻²⁹. Our understanding of crystallisation kinetics in magmas is underpinned by *ex situ* crystallisation quench experiments. Here we perform novel *in situ* experiments in order to visualize and quantify the evolution of rapid crystallisation in basaltic magmas, as with this approach it is possible to ascertain in real time when crystals start to form and how quickly magma crystallinity evolves.

A frequently used model to describe crystallisation as function of time is given by an exponential law^{21,23}, where the rate of crystallisation is controlled by the characteristic time $\tau^{(c)}$. The characteristic time is a measure of how fast a process will approach the equilibrium^{21,30}. The smaller

 $\tau^{(c)}$, the faster crystals reach their equilibrium abundance (see Methods section). La Spina et al. (ref. 21) demonstrated that the time required to reach the equilibrium crystal fraction in basalts is ~4.6 times $\tau^{(c)}$. Furthermore, during mild lava-fountaining basaltic activity²¹, $\tau^{(c)}$ is in the order of thousands seconds, resulting in a crystallisation time of ~2 hours. Crystallisation rate increases with magma ascent rate, as cooling and decompression rates increase³⁰⁻³⁴. Therefore, $\tau^{(c)}$ is smaller and the equilibrium crystal fraction will be reached faster. However, the characteristic times of crystal growth during fast magma ascent have not been quantified.

In order to quantify the characteristic times of plagioclase and pyroxene crystallisation during rapid ascent of basaltic magma, here we present the first *in situ* 4D (3D plus time) crystallisation kinetics experiments under fast cooling rates, using fast synchrotron X-ray microtomography. Our experiments provide the first estimation of the characteristic time for plagioclase and pyroxene crystallisation in trachybasaltic magmas during a rapid and continuous increase of undercooling, where ΔT is defined as the difference between the highest temperature at which plagioclase and pyroxene is expected to crystallise and the temperature of the magma²⁴⁻²⁷.

Crystallisation experiments at high undercooling

Crystallisation experiments were performed *in situ* at beamline I12-JEEP, Diamond Light Source, Harwell, UK, using a trachybasaltic glass (Supplementary Table 1) from the 2001 Etna eruption as the starting material (see Methods). We combined a bespoke high-temperature environmental cell³⁵ with fast synchrotron X-ray microtomography to image the evolution of crystallisation in real time³⁶ in two experiments. In the first part of the experiment, crystallisation was induced by decreasing temperature from 1250 °C to either 1170 °C or 1150 °C at 0.4 °C s⁻¹ at ambient pressure with a dwell time at the final temperature of 4 h³⁶ (Supplementary Fig. 1). During these 4 h, blocky and prismatic clinopyroxene and oxide crystals were able to grow³⁶ (Fig. 1 and Supplementary Figs. 2 and 3), and the residual melt evolved to a basaltic trachyandesitic composition (Supplementary Table 1; Supplementary Figs. 2 and 3). After 4 h at sub-liquidus

conditions (1170 °C and 1150 °C), the system was perturbed through a rapid cooling rate of 0.4 °C s⁻¹, inducing a sudden and continuous increase of undercooling (ΔT). This produced a rapid crystallisation event from the residual melt (Fig. 1; Supplementary Figs. 2 and 3). Our results show that skeletal plagioclase crystals grew during this final stage of rapid cooling, specifically between 1112 and 1073 °C after a dwell time of 4 hours at 1150 °C (Fig. 1) and between 1131 and 1053 °C after a dwell time of 4 hours at 1170 °C (Supplementary Fig. 3a,b,c). Plagioclase crystals grew to equilibrium abundance in ~100 seconds (Fig.1), i.e. between two 3D scans. Following this initial burst of rapid plagioclase growth, dendritic clinopyroxene crystals began to nucleate heterogeneously on plagioclase and grew to their final size in the following 180 seconds (Fig. 1b,c; Supplementary Figs. 2b,c, 3d,e, 4 and 5).

The large ΔT reached in a relatively short time during our *in situ* 4D crystallisation experiments generated distinctive skeletal plagioclase crystals with swallow-tail morphology and dendritic pyroxene (Fig. 2a-c), similar to the skeletal plagioclase and dendritic pyroxene crystals observed in the products of explosive basaltic Plinian eruptions^{7-11,37,38}, such as Etna 122 B.C.^{7,8,37,38} (Fig. 2d).

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Skeletal plagioclase crystallisation occurred at ΔT between 60 and 140 °C with a growth rate of $3x10^{-5}$ cm s⁻¹ (mm h⁻¹) (Supplementary Table 2), whilst dendritic pyroxene crystallised at ΔT between 60 and 175 °C with a growth rate of $2x10^{-5}$ cm s⁻¹ (mm h⁻¹) (Supplementary Table 2). This indicates that a rapid increase of ΔT (>60 °C) induces fast crystallisation. As the equilibrium pyroxene crystal content is achieved within ~180 s, we can infer that the pyroxene characteristic time under large ΔT is < 40 s. For plagioclase, where the equilibrium crystal content is achieved

within 90 s the characteristic time is < 20 s. These are about two orders of magnitude less than the characteristic time found by La Spina et al. (ref. 21) for effusive and mild lava fountaining activity at Etna (Italy), Stromboli (Italy) and Kilauea (Hawaii), which involved much smaller ΔT (30-60 °C)²¹ and magma ascent rates of ~3 m s⁻¹.

Ex situ experiments were also performed using a TZM cold seal pressure vessel apparatus, in order to investigate whether fast crystallisation also occurs in a hydrous trachybasaltic melt during rapid and continuous cooling and decompression (see Methods), simulating fast magma ascent in the conduit. The experimental approach consisted of holding the sample for 30 minutes at 75 MPa and 1070 °C before the sample was decompressed and the temperature decreased. After 30 minutes pressure and temperature were decreased continuously for 300 s at 0.2 MPa s⁻¹ and 0.2 °Cs⁻¹ respectively, reaching 15 MPa and 1010 °C (Supplementary Table 3; Supplementary Fig. 6). The rapid decompression and cooling enabled large Δ Ts to be the achieved (>100 °C; see Supplementary Fig. 6) in 300 s, favouring predominantly clinopyroxene crystallisation, with minor plagioclase and oxide (Supplementary Fig. 7). This indicates that τ ^(c) is < 60 s, in agreement with the order of magnitude estimated from *in situ* experiments.

Large undercooling can produce significant syn-eruptive microlite crystallisation during rapid magma ascent²⁴⁻²⁷. This increase in crystallinity dramatically increases the viscosity of the magma¹⁹. This process has been proposed to explain explosive basaltic Plinian eruptions, supported by evidence of high microlite contents^{7-11,37,38}. However, a mechanism for this exceptionally fast crystallisation during rapid magma ascent in basaltic Plinian eruptions, favouring fragmentation, has not been demonstrated so far.

Numerical simulation of the 122 B.C. Etna basaltic Plinian eruption

Magma fragmentation in basaltic Plinian eruptions has been investigated with conduit models, where crystallisation has been assumed either to be constant²⁰ or at equilibrium⁴⁰. Recent results demonstrate that disequilibrium crystallisation plays a fundamental role in magma dynamics within

the conduit²¹, but syn-eruptive disequilibrium crystallisation has not yet been considered for basaltic explosive volcanism^{20,40}.

We used the conduit model described by La Spina et al. (ref. 21, 23) to investigate the effect of the new experimentally constrained characteristic times for crystallisation with large undercooling derived here on the ductile-brittle transition of basaltic magma during highly explosive eruptions. Since we are focussing our attention on explosive eruptions with high mass eruption rates (>10⁶ kg/s), it is reasonable to assume that outgassing is negligible for this kind of activity. Indeed, outgassing will be inhibited by fast magma ascent and fragmentation will be achieved faster than outgassing⁴¹. As a test case, we consider the 122 B.C. Etna basaltic Plinian eruption⁶⁻⁸. To model fragmentation we adopt the strain-rate criterion introduced by Papale (ref. 1):

$$\dot{\gamma} = k \frac{G_{\infty}}{\mu} \tag{1}$$

where $\dot{\gamma}$ is the elongational strain rate, k=0.01 is a constant, μ is the magmatic viscosity and G_{∞} is the elastic modulus at infinite frequency. The other constitutive equations are reported in the Methods section. In Figure 3 we report the calculated plagioclase undercooling, crystal content and viscosity as function of depth for $\tau^{(c)}$ = 10 and 1000 s. Large undercooling is produced by adiabatic expansion of exsolved volatiles, and mitigated by the latent heat of crystallisation for $\tau^{(c)}$ = 10 s (Fig. 3a). In this case, numerical results show a rapid increase in crystallinity and in viscosity at depths below 2 km (Fig. 3c), leading to fragmentation and explosive Plinian eruption. Importantly, numerical results indicate that strain-induced fragmentation is favoured by a combination of rapid viscosity increase (about one order of magnitude in 5 seconds approaching the fragmentation depth) and fast decompression/ascent rates (~0.2 MPa s⁻¹ and ~15 m s⁻¹) that generate high strain rates. For $\tau^{(c)}$ = 1000 s, instead, the crystallisation rate is slow, and viscosity stays within the fragmentation threshold all along the conduit.

Sensitivity analyses on basaltic magma fragmentation driven by rapid crystallisation

Having established that fast crystallisation plays an important role on basaltic Plinian eruptions, we performed a sensitivity study with our numerical model to investigate the parameter space whereby basaltic fragmentation driven by rapid crystallisation may occur. We use the Etna 122 B.C. eruption as a test case. We focus on the behaviour of ΔT as a function of characteristic time of crystallisation, pressure, temperature and magmatic H₂O content at the conduit inlet, conduit radius, and initial phenocryst content. A detailed description of the initial condition of the sensitivity study can be found in the Methods section. We performed several sensitivity studies assuming a characteristic time of $\tau^{(c)} = 10$ s, as observed in our experiments, and $\tau^{(c)} = 1000$ s, as observed for Etna 2001 in mild explosive activity. We also examined $\tau^{(c)} = 1$ and 100 s for completeness.

Numerical results show that undercooling is principally controlled by $\tau^{(c)}$ (Fig. 4a), because the release of latent heat during rapid crystallisation (i.e. small $\tau^{(c)}$) affects the temperature of the system, and, consequently, the undercooling. Therefore, undercooling and characteristic time are linked, affecting one each other. However, our numerical results indicate that, within the parameters space investigated, ΔT is always maintained between 60 and 190 °C (Fig. 4b), which is enough to enable rapid crystallisation in any case. Furthermore, the sensitivity analyses indicate that, after the characteristic time, initial temperature and water content (exsolved + dissolved) play an important role on controlling undercooling (Fig. 4a). ΔT at the point of fragmentation increases as initial temperature decreases (Fig. 4b), meaning that cooler magma in the chamber is more likely to produce microlite crystallisation during ascent, as ΔT will already be large under the pre-eruptive condition. An increase in the initial total H₂O (exsolved + dissolved) also produces an increase of ΔT (Fig. 4c), caused by greater cooling of the system due to enhanced adiabatic gas expansion. Furthermore, higher water content favours rapid crystallisation of microlites, due to the increase in diffusivity within the melt.

Our sensitivity studies show that a lower initial temperature and a higher pre-eruptive crystal content results in a greater likelihood of explosive eruptions (Fig. 4d,e). This is consistent with estimates of pre-eruptive temperatures obtained for the Etna and Fontana Plinian eruptions, which

range between 1000 and 1060 °C³⁷. Regarding the pre-eruptive crystal content, however, products erupted from basaltic Plinian eruptions are characterized by a small phenocryst content (<10 vol.%)^{7,8,10,11,37,38,40}. Therefore, in this context of low initial temperature and low phenocryst content, our results highlight that the characteristic time of crystallisation has a fundamental role on the likelihood of producing an explosive eruption. Indeed, a small $\tau^{(c)}$ is the primary cause for a significant increase of the syn-eruptive crystal fraction over short timescales during magma ascent, which consequently increases magma viscosity and, thus, the probability of magma fragmentation (Fig. 4f). Moreover, our sensitivity analysis demonstrates that when the syn-eruptive crystal content exceeds 30 vol.%, all numerical solutions reach the fragmentation threshold (Fig. 4f). Indeed, in a fast ascending magma (from our sensitivity analyses we have an average ascent rate between ~5-50 m/s), this increase in syn-eruptive crystal content produces a rapid and dramatic increase of viscosity (about one order of magnitude in less than 10 s close to the fragmentation depth), increasing the likelihood of magma fragmentation. Numerical results also show that, as soon as viscosity exceeds 10⁵ Pa s, all the numerical solutions reach the fragmentation threshold, generating an explosive eruption (Fig 4g). Indeed, for a fast ascending magma where outgassing can be neglected and strain-rates are high, the key parameter controlling the triggering of fragmentation is viscosity. Therefore, 10⁵ Pa s represents a minimum viscosity determining a drastic change in eruptive style for activities characterised by high mass eruption rates. This minimum viscosity is one order of magnitude lower than previously reported for low-viscosity magmas ^{1,42}.

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Experimental and natural observations combined with a numerical model allow us to conclude that pre-eruptive temperatures <1100 °C favour the formation of highly explosive basaltic eruptions, such as Plinian volcanism, driven by fast syn-eruptive crystal growth under high undercooling. This implies that all basaltic systems on Earth have the potential to produce powerful explosive eruptions.

METHODS

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Starting material. The starting material, used for our crystallisation experiments, is a trachybasalt from the lower vents of the 2001 Mt. Etna eruption^{36,43}. The anhydrous, glassy starting material was obtained by melting crushed rock samples in a Pt crucible. Melting was performed in a Nabertherm® MoSi₂ box furnace at 1400 °C and at atmospheric pressure. The melt was left in the furnace for four hours to allow the melt to fully degas and to dissolve the crystals present. The melt was then quenched in air to glass. This procedure was repeated two times to homogenise the melt. Finally, anhydrous glassy cylinders 3 mm in diameter and 4 mm in length were drilled from the synthesized glass for ex situ and in situ 4D crystallisation experiments. The chemical composition of the anhydrous glassy starting material has been analysed with a Jeol JXA 8530 F microprobe at the Photon Science Institute, University of Manchester, Manchester, UK, and are reported in Supplementary Table 1. Analyses were performed using a 15 kV accelerating voltage, 10 nA beam current and beam size of 10 µm. Standards used for calibration were albite for Na, periclase for Mg, corundum for Al, fayalite for Fe, tephroite for Mn, apatite for P, sanidine for K, wollastonite for Ca and Si and rutile for Ti. Sodium and potassium were measured first to minimize loss owing to volatilisation. Hydrous trachybasaltic glass with ~3 wt. % of H₂O was obtained melting the starting material and homogenizing H₂O in a Pt capsule at 100 MPa and 1200 °C. The hydrous starting glasses were produced using a TZM cold seal pressure vessel apparatus at the School of Earth Sciences, University of Bristol, Bristol, UK. Before to perform crystallisation experiments Raman spectra were collected from the hydrous starting glass, using a Thermo ScientificTM DXRTMxi Raman Imaging Microscope at the School of Earth Sciences, University of Bristol (Bristol, UK) in order to

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Ex situ decompression and cooling experiments.

check that the amount of H₂O was ~3 wt. %.

Ex situ experiments were performed in order to investigate the process of fast crystallisation in a hydrous trachybasaltic melt during a continuous decompression (0.2 MPa s⁻¹) and cooling (0.2 °Cs⁻¹), simulating magma ascent in the conduit. Two experiments were performed using a TZM cold seal pressure vessel apparatus at the School of Earth Sciences, University of Bristol, Bristol, UK. The redox condition of the apparatus was NNO, adding ~1 vol.% of hydrogen to the pressuring argon. Capsules (Au₈₀-Pd₂₀) were loaded with hydrous glass with a cylindrical shape. The experimental approach consisted of holding the sample for 30 minutes at 75 MPa and 1070 °C before being decompressed and cooled. The first experiment was quenched after 30 minutes at 75 MPa and 1070 °C, in order to texturally characterise the initial conditions before that fast decompression and cooling were applied. In the second experiment, after 30 minutes pressure and temperature were released continuously in 300 s at 0.2 MPa s⁻¹ and 0.2 °Cs⁻¹ respectively, reaching 15 MPa and 1010 °C (Supplementary Table 3; Supplementary Fig. 6). The sample was rapidly quenched at the final conditions. Final pressure was chosen as approximate fragmentation pressures calculated by modeling. Decompression and cooling rates were predicted from the conduit model for magma ascent during basaltic Plinian eruptions.

In situ synchrotron X-ray microtomography experiments. The experiments were performed at the beamline I12-JEEP⁴⁴, Diamond Light Source, Harwell, UK. For these *in situ* crystallisation experiments, we used the high-temperature resistance Alice furnace⁴⁵, which allows us to control cooling at 0.05 °Cs⁻¹ to 0.4 °Cs⁻¹, and the P2R in situ rig³⁵ for high speed rotation. Temperature was measured with a R type thermocouple, which was close to the sample, positioned in the middle of the hot spot. The dimension of the Alice furnace's hot spot with homogeneous temperature is ~5x5x5 mm; our samples were positioned within this area. The R type thermocouple provided us the temperature of the sample with an uncertainty of ± 5 °C. The sample holder was an alumina crucible (Supplementary Fig. 2), which is suitable for the temperature range investigated and it has low X-ray attenuation coefficient. The glassy cylinders were heated *in situ* in the Alice furnace up

to 1250 °C for 30 minutes (Supplementary Figure 1). After the initial annealing period, crystallisation was induced by decreasing temperature from 1250 °C to 1170 °C or 1150 °C at ambient pressure, holding at the final temperature for 4 h³⁶ (Supplementary Figure 1). After this step, the system was perturbed by a rapid cooling at rate of 0.4 °Cs⁻¹ in order to investigate the rapid crystallisation in real time (Supplementary Figure 1), reaching high undercooling (up to ~180 °C) in a short time.

The experiments were performed in phase-contrast mode, setting the sample-to-detector distance at 2300 mm in order to work in the edge-detection regime⁴⁶ (Supplementary Table 4). The projections were acquired using a monochromatic X-ray beam with energy of 53 keV. In each scan, 1800 tomographic projections were acquired by the detector with equiangular steps over a full rotation angle of 180° (Supplementary Table 4). The exposure time for the acquisition of each projection was 0.05 s (Supplementary Table 4), therefore, the temporal resolution of each scan was of 90 seconds. The isotropic pixel size is 3.2 μ m. The detector was a high-resolution imaging PCO edge camera with optical module 3, corresponding to a field of view of 8.0 mm × 7.0 mm. Scan acquisition started before the end of the annealing, covering the cooling period between 1250 °C and the dwell temperatures (1170 and 1150 °C), the entire duration of the dwell time and the final rapid cooling rate of 0.4 °Cs⁻¹.

Image reconstruction and processing. Tomographic projections were reconstructed into 32-bit slices by using Diamond I12 in-house python codes, using the *gridrec* algorithm^{47,48} (http://confluence.diamond.ac.uk/display/I12Tech/Reconstruction+ scripts+for+time+series+tomography)^{49,50}. The pre-processing pipeline includes centre of rotation calculation⁴⁹, zinger removal, blob removal⁵⁰, and regularisation-based ring removal⁵¹.

The reconstructed slices were converted to 8-bit raw format and stacked using ImageJ software⁵² to obtain volumes in which the isotropic voxel has an edge size of 3.2 µm. Reconstructed volumes of experiments ET1150 and ET1170 were then cropped using Avizo® software v.8.0 (FEI

Visualization Sciences Group) in order to select the volume of interest (VOI) (Supplementary Table 4). In the experiment ET1150 plagioclase and pyroxene crystals nucleated and grew in a relatively large pocket of melt (Figs 1 and 2). Therefore, the VOI selected consists of a volume of melt where the rapid crystallisation of plagioclase and pyroxene occurred during the final rapid cooling rate of 0.4 °Cs⁻¹. In the experiment ET1170 plagioclase and pyroxene crystals formed in narrow layers of melt (Supplementary Fig. 3), during rapid continuous cooling at a rate of 0.4 °Cs⁻¹.

Three-dimensional visualization (volume rendering) of the reconstructed volumes was obtained using the commercial software VGStudio 3.0 (Volume Graphics), which allowed us to make 3D textural observations of the plagioclase and pyroxene crystal morphologies (Fig. 2). Therefore, the reconstructed volume of each scan allowed us to quantify when and at which range of temperature plagioclase and pyroxene crystals were able to form.

Image segmentation and analysis of plagioclase. Segmentation is the process that allows separation of objects from the background to obtain binary volumes containing only the feature of interest. Segmentation of plagioclase crystals from the glassy matrix was performed using the semi-automatic volume segmentation^{53,54} in Avizo software v. 8.0 (Supplementary Table 4). This segmentation requires manual drawing of the outlines of crystals on the 2D slices. This is repeated every 5–10 slices, depending on the size of the crystal and the complexity of their shape, along the crystal length. The crystal shape is reconstructed automatically by the software through an interpolation procedure. The advantage of this technique is that the operator can obtain the real morphology of the object of interest by visual inspection^{53,54}.

The reconstructed 3D images were processed and analysed with the Pore3D software library, custom-developed at Elettra⁵⁵. The Pore3D software allowed us to quantify the number of plagioclase crystals, the volume and the maximum length of each crystal, operating directly in the 3D domain^{55,56}. As we were able to obtain the 3D shape of plagioclase crystals and the real maximum axis length (L_{3D}) we could calculate the growth rate (Y_{L3D}) of plagioclase crystals

(Supplementary Table 2), using the experimental duration of growth (experimental duration). The growth rate was estimated using the following equation⁵³:

 $Y_{L3D} = (L_{3D}*0.5)/t_{growth}$

where t_{growth} is the time required for crystal growth. The microtomography images give us the opportunity to measure the volume of crystals. The volumetric growth rate (Yv) was calculated (Supplementary Table 2) using the following relationship⁵³:

$$Y_{V} = (V*0.5)/t_{exp}$$

where V is the volume of the crystal.

Image analysis of pyroxene growth kinetics. Back-scattered electron (BSE) images were collected, using a JEOL JSM-6390LA FE-SEM at the School of Earth and Environmental Sciences, University of Manchester, Manchester, UK, in order to analyse the pyroxene morphologies and kinetics. We used an acceleration voltage of 15 kV and beam current of 10 nA. The sizes of dendritic pyroxene crystals were measured in the 2D domain, using BSE images and ImageJ software⁵², as pyroxene morphologies formed during continuous cooling are difficult to resolve and analyse in the 3D domain. The pyroxene growth rate is calculated by dividing the entire length of the dendritic crystal over the duration of the pyroxene growth (Supplementary Table 2), as dendritic crystals grow in one direction.

Electron microprobe analysis. Samples obtained with *ex situ* and *in situ* crystallisation experiments were analysed with a JEOL JXA-8530F field emission electron microprobe at the Photon Science Institute, University of Manchester, Manchester, UK. For both instruments, the operating conditions were as follows: 15 kV accelerating voltage, 10 nA beam current, and beam diameter of 10 or 5 μm (the latter for microlites). Sodium and potassium were measured first to minimise loss by volatilisation. Calibration standards were albite for Na, periclase for Mg, corundum for Al, fayalite for Fe, tephroite for Mn, apatite for P, sanidine for K, wollastonite for Ca

and Si and rutile for Ti. Several compositional maps, with a nominal resolution of 1 µm2, have been also collected using the EPMA (EPMA map, are reported in Supplementary Figs. 4, 5 and 7).

Constitutive equations for the conduit model. In this work we use the 1D steady-state model for magma ascent described by ref. (21, 23, 57). The governing equations used in this work are reported in ref. (57). The application to a specific volcano is achieved by providing constitutive equations to describe the specific rheological, solubility, crystallisation, outgassing, and fragmentation behaviour of the system.

Following ref. (58), the viscosity of the liquid phase is modelled as:

$$\mu_l = \mu_{melt} \cdot \theta(x_c^l),$$

where μ_{melt} is the viscosity of the bubble-free, crystal-free liquid phase and θ is a factor which increases viscosity attributed to the presence of crystals⁵⁹.

We use an empirical relationship to estimate μ_{melt} as a function of water concentration and temperature, as in ref. (60) (based on the Vogel-Fulcher-Tammann equation):

$$\log(\mu_{melt}) = A + \frac{B(y, x_{d_{H_2O}}^{md})}{T - C(y, x_{d_{H_2O}}^{md})},$$

where the viscosity μ_{melt} is in Pa s and T is the temperature in Kelvin. The parameter A is the logarithmic value of the viscosity at infinite temperature and it is assumed to be constant for all melts. The parameters B and C, instead, are functions of the melt composition y and of the dissolved water content $x_{d_{H_2O}}^{md}$. In this work, we used the composition of the average melt inclusion composition (Etna 122 B.C.) from ref. (61). Furthermore, as crystallisation proceeds, viscosity is increased according to the empirical model described in ref. (62):

$$\theta = \frac{1 + \varphi^{\delta}}{[1 - F(\varphi, \xi, \gamma)]^{B\phi^*}},$$

where

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$$F = (1 - \xi)erf\left[\frac{\sqrt{\pi}}{2(1 - \xi)}\varphi(1 + \varphi^{\gamma})\right], \quad \varphi = \frac{\left(\sum_{j=1}^{n_c} x_{c_j}^l\right)}{\phi^*}.$$

- The fitting parameters B, δ , ξ , γ and ϕ^* chosen for this work are the same used in ref. (63).
- 376 The model proposed in this work takes into account two different gas components: water and
- 377 carbon dioxide. The equilibrium profile of the dissolved gas content $x_{d_i}^{md,eq}$ of component i follows
- 378 the Henry's Law, i.e.

$$x_{d_i}^{md,eq} = \sigma_i \left(\frac{P_{g,i}}{\overline{P}}\right)^{\varepsilon_i},$$

- where $P_{g,i} = \alpha_{g_i} P_g / \alpha_g$ is the partial pressure of the *i*-th gas component expressed in Pa, $\bar{P} = 1$
- Pa is used to make the expression in the brackets adimensional, σ_i is the solubility coefficient and ε_i
- is the solubility exponent. We assume that the solubility parameter σ_i and ε_i are constant during
- ascent. For this work we adopted the following parameters $\sigma_{H_2O} = 1.8911 \times 10^{-6}$; $\varepsilon_{H_2O} = 0.5257$;
- 384 $\sigma_{CO_2} = 2.2154 \times 10^{-12}$; $\varepsilon_{CO_2} = 1.075$. In this work, we assume also equilibrium exsolution, which
- means that the dissolved volatile contents always follow the equilibrium profile.
- The crystallisation model adopted here has been proposed in ref. (23). We consider the three
- different major crystal components erupted by Etna volcano: plagioclase, pyroxene and olivine. We
- assume that crystals stay coupled with the melt (i.e. no fractional crystallisation). For a better
- modelling of crystal nucleation and growth, we also assume that the equilibrium crystal contents are
- 390 functions of temperature, pressure and dissolved water content. With these assumptions, the
- equilibrium mass fraction $x_{c_i}^{l,eq}$ of crystal phase j is computed using the polynomial function

$$x_{c_j}^{l,eq}(P^*, T^*, x_d^*) = \zeta_{j,1}(P^*)^2 + \zeta_{j,2}(T^*)^2 + \zeta_{j,3}(x_d^*)^2 + \zeta_{j,4}(P^*)(T^*) + 392$$

$$+\zeta_{j,5}(T^*)(x_d^*)+\zeta_{j,6}(x_d^*)(P^*)+\zeta_{j,7}(P^*)+\zeta_{j,8}(T^*)+\zeta_{j,9}(x_d^*)+\zeta_{j,10},$$

- where P^* is the liquid pressure expressed in bars, T^* is the temperature expressed in Celsius
- degrees and x_d^* is the dissolved water concentration in weight percent. From $x_{c_j}^{l,eq}$, the equilibrium
- 395 crystal volume fraction β_j^{eq} can be computed using the relation

$$\beta_j^{eq} = \frac{\rho_l x_{c_j}^{l,eq}}{\rho_{c_j}}.$$

The parameters $\zeta_{j,i}$ are calculated fitting the polynomial function over a large range of data obtained at different pressures, temperatures and water contents with alphaMELTS⁶⁴, a command line version of MELTS⁶⁵. As previously, we used the average melt inclusion composition (Etna 122 B.C.) from ref. (61).

Disequilibrium crystallisation is considered in our model and characteristic times controls the time needed to reach equilibrium crystal content. The differential equation in the conduit model which describes the crystal volume fraction within the conduit is the following²¹:

$$\frac{\partial \alpha_l \rho_c \beta u_l}{\partial z} = -\frac{1}{\tau^{(c)}} \alpha_l \rho_c (\beta - \beta^{eq}).$$

Here, α_l is the volume fraction of the liquid phase, ρ_c is the density of the crystals, u_l is the velocity of the liquid phase, z is the vertical axis, β is the actual total crystal volume fraction, whereas β^{eq} is the equilibrium total crystal volume fraction. If we multiply all the terms by the characteristic time $\tau^{(c)}$, we notice that the smaller is $\tau^{(c)}$, the smaller has to be $(\beta - \beta^{eq})$. In other terms, the smaller is the characteristic time, the faster the crystals will reach the equilibrium value.

Formally, as described in La Spina et al. (ref. 21), the characteristic time reflects the time required to reduce the difference between the actual and the equilibrium value to e^{-1} (~37%) of the initial difference. This means that, if β_0 is the initial value of a physical parameter β (which, in our case, is the crystal volume fraction), and β_{eq} is the equilibrium value in response to a perturbation of the system, at the characteristic time τ , we have

415
$$\beta(\tau) = \beta^{eq} + e^{-1}(\beta_0 - \beta^{eq}).$$

The experimental phase diagram for Etna basalt erupted during the 122 B.C. eruption³⁷ provides the plagioclase and pyroxene liquidus at different pressures and temperatures, whilst the conduit model is able to track temperature evolution within the conduit. Combining both of these data, we can estimate ΔT with respect to the plagioclase and pyroxene liquidus during magma ascent.

For this work, as we are interested in the highly explosive activity, we assumed no relative velocity between gas and melt. Furthermore, as we indicated in the main text, we used as fragmentation model the strain-rate criterion introduced by ref. (1).

Initial condition for the sensitivity analysis.

The range of input parameters adopted for the sensitivity analysis are the following: 140–160 MPa for the inlet pressure at 6000 m depth, 1050–1100 °C for the magma inlet temperature, 5–30 m for the radius of the conduit, 2.0–4.0 wt.% for the total water content, 0.1–2.0 wt.% for the total CO₂ content, 0–20 vol.% for the initial phenocrysts, and 1–1000 s for the characteristic time of crystallisation. As we do not know the probability distribution of the uncertain input parameters, we have assumed a uniform distribution within the aforementioned ranges. The sensitivity analysis was performed on using the DAKOTA toolkit (Design Analysis Kit for Optimization and Terascale Applications)⁶⁶, an open-source software developed at Sandia National Laboratories that provides a flexible and extensible interface between analysis codes and iterative systems analysis methods such as uncertainty quantification, sensitivity analysis, optimization, and parameter estimation.

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610 611 612 **Supplementary information** is available in the online version of the paper. 613 614 Acknowledgements 615 The research leading to these results has received funding from the RCUK NERC DisEqm 616 project (NE/N018575/1) and (NE/M013561/1). The beamtime on I12 was provided by Diamond 617 Light Source (EE16188-1) and laboratory space by the Research Complex at Harwell. 618 619 **Author Contributions** 620 M.P., F.A., M.R.B., and P.D.L. conceived the research project. F.A., M.P., G.L.S., N.L.G., B.C., M.E.H., D.D.G., N.T.V., S.N., R.C.A., E.W.L., P.D.L. and M.R.B. contributed to the beamline 621 622 experiments. F.A. collected the volcanic rocks for the starting material. D.D.G., H.M and R.A.B. prepared the starting material. F.A., M.P. and G.L.S performed image reconstruction. F.A. and M.P. 623 624 performed image processing. F.A. performed image segmentation and analysis. G.L.S. performed 625 simulations using the conduit model. R.A.B. and F.A. performed ex situ decompression 626 experiments. E.C.B., F.A. and G.L.S. collected samples of the Etna 122 BC Plinian eruption. E.C.B. 627 and F.A. acquired and analysed back-scattered electron images of Etna Plinian eruption's samples. 628 F.A., G.L.S., M.R.B., M.P. and E.C.B. wrote the manuscript, with contributions from all other 629 authors. 630 **Author Information** 631 632 Reprints and permissions information is available at www.nature.com/reprints. The authors 633 declare no competing financial interests. Readers are welcome to comment on the online version of

paper. Correspondence and requests for materials should be addressed to F.A.

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(fabio.arzilli@manchester.ac.uk)

FIGURES

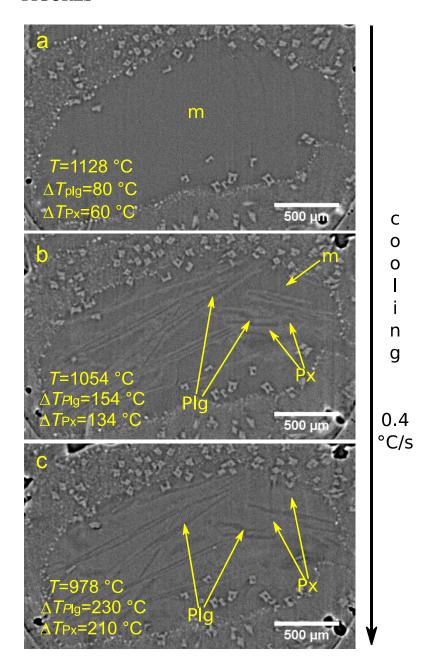
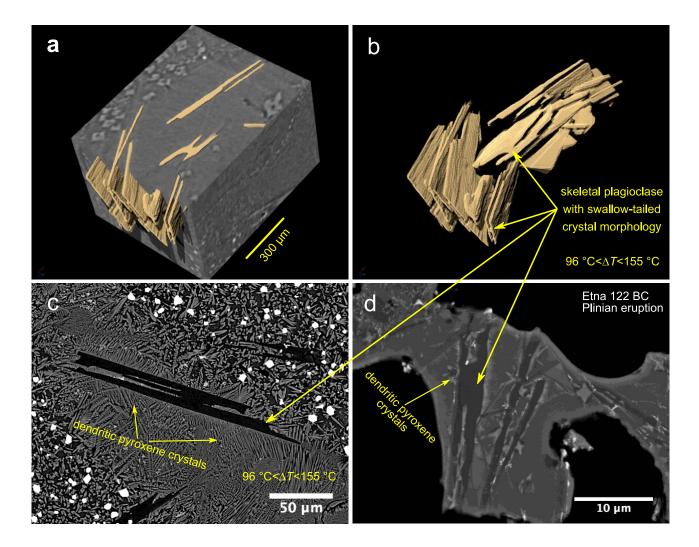


Figure 1. Crystallisation through time. Reconstructed axial slices during continuous cooling at 0.4 °Cs⁻¹: (a) frame after 24 s from the onset of the cooling, in which the temperature ranges between 1144 and 1112 °C (average 1128 °C); (b) frame after 208 s, in which the temperature ranges between 1073 and 1034 °C (average 1054 °C); (c) frame after 392 s, in which the temperature ranges between 997 and 959 °C (average 978 °C). m = melt; plg = plagioclase; px =

pyroxene.



the morphology and the spatial distribution of plagioclase crystals that formed during the rapid cooling at 96<ΔT<155 °C. (b) 3D view of the plagioclase with swallow-tailed crystal morphology. (c) Back scattered electron image of plagioclase with swallow-tailed crystal morphology. (d) Back scattered electron image of plagioclase with swallow-tailed crystal morphology produced during the Etna 122 B.C Plinian eruption. Note heterogeneous nucleation of pyroxene around plagioclase, seen

as a light-coloured halo, and similar to that seen in figure 1b.

Figure 2. Plagioclase crystal morphology. (a) The 3D volume rendering of sample SS1150 shows

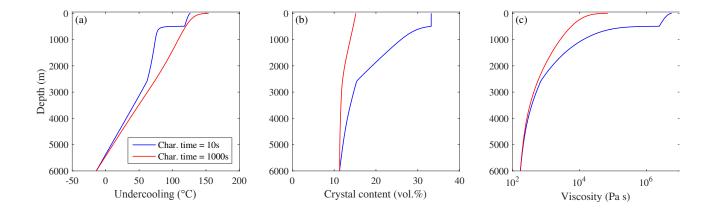


Figure 3. Model results during magma ascent. (a) Undercooling as a function of depth, calculated for $\tau(c) = 10$ (blue) and 1000 s (red). Cooling is driven by adiabatic expansion of gas, mitigated by latent heat of crystallisation particularly in the fast crystallising case. (b) Crystal content in vol%, demonstrating the rapid increase in crystal load when $\tau(c) = 10$ s. (c) Magma viscosity, demonstrating that the higher crystal load produces 3-4 order of magnitude increase in viscosity, leading to fragmentation.

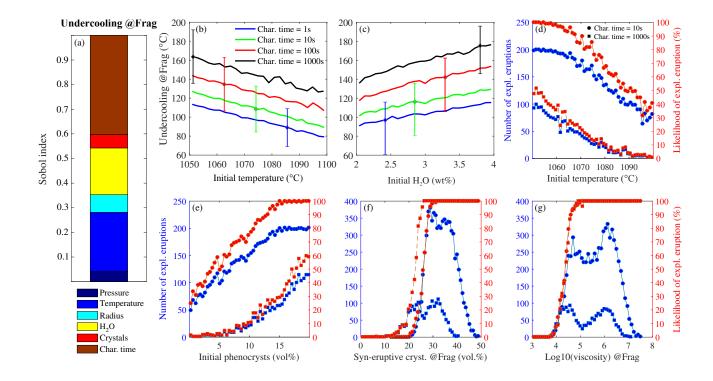
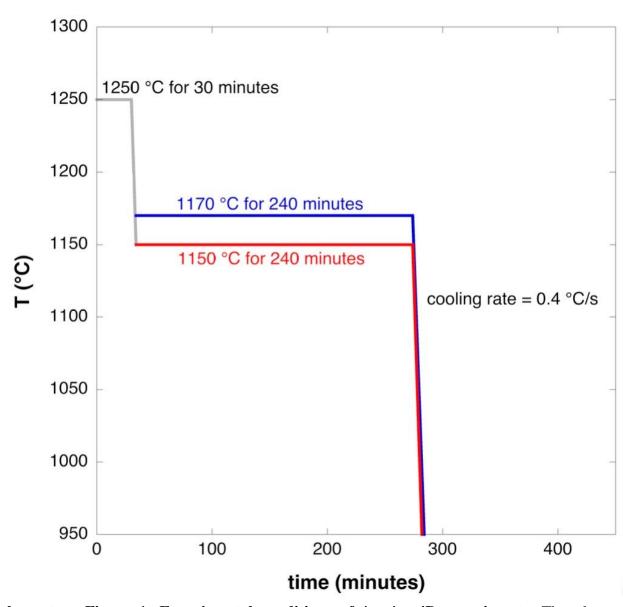
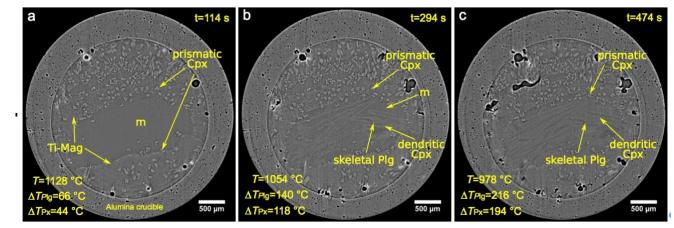


Figure 4. Relationships between characteristic time, initial temperature, initial H₂O content of the magma, syn-eruptive crystal content and magma viscosity and the undercooling of the system at the fragmentation level. These figures were calculated using repeated runs of the model while changing individual parameters to reveal the sensitivity of the system to each parameter. Likelihood of explosive eruption as a function of a specific parameter arises from the ratio between the number of model runs producing explosive eruptions divided by the total number of model runs used to test that parameter. Therefore, this is not a probabilistic assessment of eruption risk, but instead depends on the critical model parameters, which control when fragmentation occurs, and the calculated probabilities depend on the choice of upper and lower limits chosen for each investigated parameter. (a) Sobol index. (b) Undercooling vs magma temperature before ascent. (c) Undercooling vs the initial H₂O content of the magma (dissolved and exsolved). For a given input value, the solid lines are the mean of a given output parameter obtained from all the simulations assuming that input value. The error bars, instead, represent the spread of that output values with respect to the corresponding mean. (d) Frequency of explosive eruptions vs magma temperature before ascent. (e) Explosion frequency vs initial phenocryst content. (f) Frequency of explosive

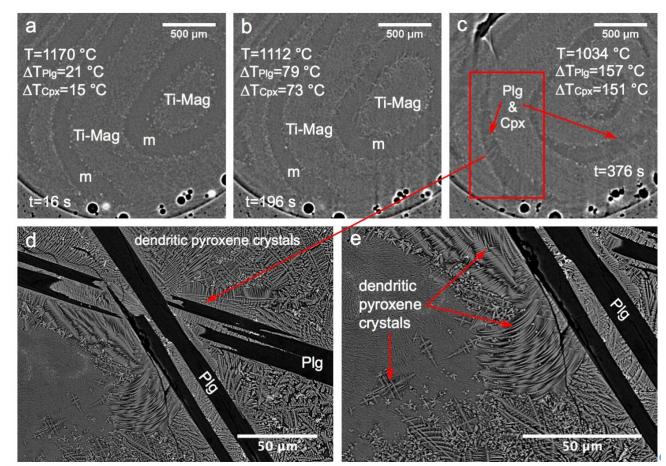
- eruptions vs syn-eruptive crystal content at the fragmentation level. (g) Frequency of explosive
- 683 eruptions vs magma viscosity at the fragmentation level.



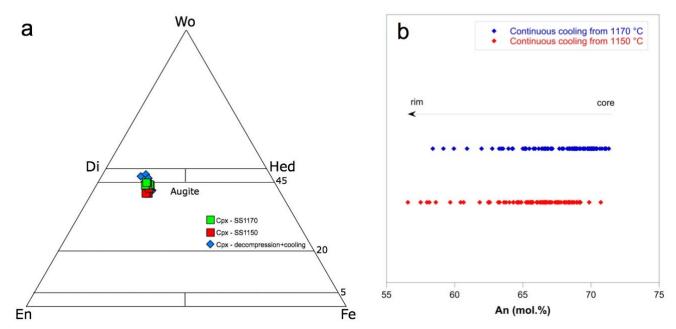
Supplementary Figure 1. Experimental conditions of *in situ* **4D experiments**. The glassy cylinders were heated in situ in the Alice furnace up to 1250 °C for 30 minutes. After the initial annealing period, crystallisation was induced by decreasing temperature from 1250 °C to 1170 °C (SS1170) or 1150 °C (SS1150) at ambient pressure, holding at the final temperature for 240 minutes. After this step, the temperature was perturbed by a rapid cooling at rate of 0.4 °C/s, reaching ~950 °C in ~8 minutes.



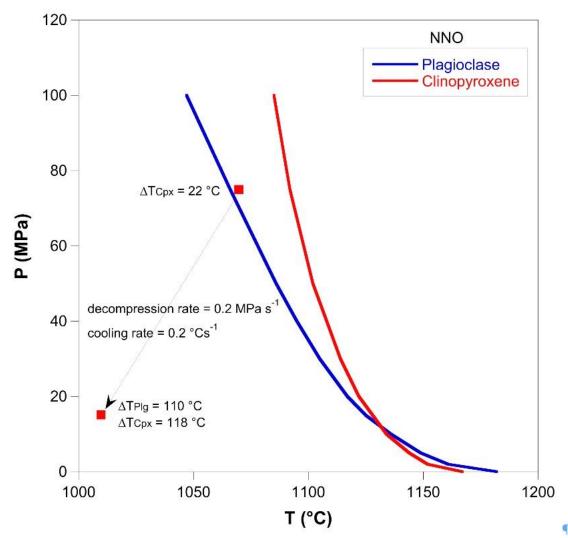
Supplementary Figure 2. Crystallisation through time during experiment SS1150. Reconstructed axial slices during continuous cooling at 0.4 °C s⁻¹: **a,** frame represents the texture after 114 s from the onset of the cooling in which the temperature ranges between 1144 and 1112 °C (average 1128 °C); **b,** frame after 294 s, in which the temperature ranges between 1073 and 1034 °C (average 1054 °C); **c,** frame after 474 s, in which the temperature ranges between 997 and 959 °C (average 978 °C). m = melt; Plg = plagioclase; Cpx = clinopyroxene; Ti-Mag: titanomagnetite. The acquisition time of each frame is 90 s. t = time from the onset of the cooling to the end of the frame acquisition.



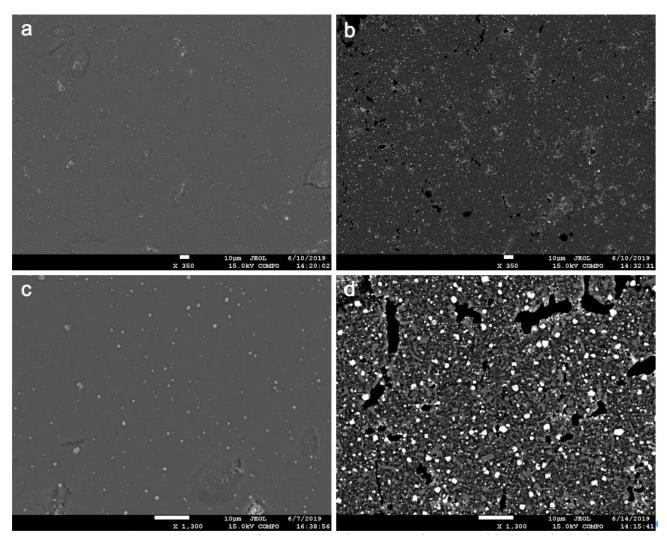
Supplementary Figure 3. Crystallisation through time during experiment SS1170. Reconstructed axial slices during continuous cooling at $0.4 \,^{\circ}\text{C s}^{-1}$: **a,** this frame shows the first 16 s from the onset of the cooling, in which the temperature ranges between 1170 and 1165 $^{\circ}\text{C}$; **b,** frame after 196 s, in which the temperature ranges between 1131 and 1092 $^{\circ}\text{C}$ (average 1112 $^{\circ}\text{C}$); **c,** frame after 376 s, in which the temperature ranges between 1053 and 1015 $^{\circ}\text{C}$ (average 1034 $^{\circ}\text{C}$). m = melt; Plg = plagioclase; Cpx = pyroxene; Ti-Mag: titanomagnetite. The acquisition time of each frame is 90 s. t = time from the onset of the cooling to the end of the frame acquisition.



Supplemetary Figure 4. Chemical compositions of pyroxene and plagioclase formed during experiments. a, Compositions of pyroxene crystals during rapid cooling at ambient pressure (SS1170 and SS1150) and rapid decompressionand cooling (Etna_7_C): green square=SS1170; red square=SS1150; blue diamond=Etna_7_C. En: enstatite; Fs: ferrosilite; Di: diopside; Hd: hedenbergite; Wo: Wollastonite. b, Anorthite contents of plagioclase crystals formed during *in situ* experiments (SS1170 and SS1150).



Supplementary Figure 5. *Ex situ* **experimental conditions of rapid cooling and decompression experiment.** The experimental approach consisted of holding the sample for 30 minutes at 75 MPa and 1070 °C before being decompressed and cooled. The first experiment was quenched after 30 minutes at 75 MPa and 1070 °C, in order to texturally characterise the initial conditions before that fast decompression and cooling were applied. In the second experiment, after 30 minutes pressure and temperature were decreased continuously in 300 s at 0.2 MPa s⁻¹ and 0.2 °Cs⁻¹ respectively, reaching 15 MPa and 1010 °C Final pressure was chosen as approximate fragmentation pressures calculated by modeling. Decompression and cooling rates were predicted from the conduit model used in this study. Plagioclase and clinopyroxene liquidi were obtained with Rhyolite-MELTS software (version 1.2), using the composition of our starting material.



Supplementary Figure 6. Textures of *ex situ* experiments. Back scattered images show the textures of: **a**, **c**, an hydrous basalt quenched at 75 Mpa and 1070 °C after 30 minutes (sample Etna7_C) and **b**, **d**, an hydrous basalt quenched after a rapid decompression and cooling at 15 MPa and 1010 °C (sample Etna7_B). **a**, **c**, The texture of sample Etna7_C is characterised by oxide crystals and glassy matrix. **b**, **d**, The mineral assemblage of sample Etna7_B consists of oxide, clinopyroxene and plagioclase. Oxide formed mostly at initial condtions, after 30 minutes at 1070 °C and 75 MPa (see Etna7_C, panels **a** and **c**). The rapid decompression and cooling allowed the achievement of large Δ Ts (>100 °C; see Supplementary Fig. 6) in 300 s, favouring predominantly clinopyroxene crystallisation, plagioclase and oxide. This indicates that τ ^(c) is 60 s, in agreement with the order of magnitude estimated from *in situ* experiments.

Supplementary Table 1. Chemical composition of the starting material.

Oxide (wt.%)	Bulk composition st. d		Residual melt after 4h at 1170 °C	Residual melt after 4h at 1150 °C		
SiO ₂	48.41	0.16	52.18	52.99		
TiO_2	1.79	0.07	1.7	1.69		
Al ₂ O ₃	16.22	0.50	16.86	17.5		
FeO*	10.66	0.30	6.05	5.4		
MnO	0.20	0.03	0.22	0.24		
MgO	6.20	0.32	5.34	4.77		
CaO	10.66	0.19	11.23	10.54		
Na ₂ O	3.41	0.20	3.74	3.99		
K ₂ O	1.90	0.07	2.09	2.24		
P_2O_5	0.54	0.04	0.59	0.64		
Total	99.85		100	100		

note: FeO* = total iron as FeO. The residual melt compositions after the 240 minutes have been obtained with a mass balance calculation, which indicates that they are basaltic trachyandesite. We considered that the main difference between experiment SS1150 and SS1170 is that the abundance of pyroxene after 4 hours at 1150 °C is higher than that at 1170 °C. Experiment SS1170 (after 4 hour at 1170 °C) indicates that prismatic pyroxene crystals are \sim 1 vol.% and oxide crystals are \sim 8 vol. %, whereas in the experiment SS1150 (after 4 hour at 1150 °C) prismatic pyroxene crystals are more abundant (\sim 8 vol.%).

Supplementary Table 2. Results of plagioclase and pyroxene growth rates during continuous cooling at 0.4 °C/s.

3D analysis							
Plagioclase d	crystal growth rate						
Sample	Growth duration (s)	L3Dmax (cm)	YL3Dmax (cm s ⁻¹)	L3Dmin (cm)	YL3Dmin (cm s ⁻¹)	L3Dave (cm)	YL3Dave (cm s ⁻¹)
SS1150	90	2.77E-02	1.54E-04	6.35E-03	3.53E-05	1.39E-02	7.74E-05
2D analysis							
Plagioclase o	crystal growth rate						
Sample	Growth duration (s)	Lmax (cm)	YLmax (cm s ⁻¹)	Lmin (cm)	YLmin (cm s ⁻¹)	Lave (cm)	YLave (cm s ⁻¹)
SS1170	180	2.73E-02	7.59E-05	1.15E-02	3.20E-05	1.78E-02	4.96E-05
Pyroxene cry	estal growth rate						
Sample	Growth duration (s)	Lmax (cm)	YLmax (cm s ⁻¹)				
SS1170	180	3.6E-03	2.0E-05				
SS1150	180	3.24E-03	1.80E-05				

note: L3Dmax = average length of the 15 largest crystals measured in the 3D domain; YL3Dmax = growth rate calculated using the 15 largest crystals; L3Dmin = average length of the 15 smallest crystals measured in the 3D domain; YL3Dmin = growth rate calculated using the 15 smallest crystals; YL3Dave = the average value of growth rate obtained taking into account the length all the crystals analysed; Lmax = average length of the 15 largest crystals measured in the 2D domain; YLmax = growth rate calculated using the 15 largest crystals.

Supplementary Table 3. Experimental conditions for ex situ experiments performed with TZM cold seal pressure vessel apparatus.

_	Sample	Ti (°C)	Pi (Mpa)	ti (s)	decompression rate (Mpa s-1)	cooling rate (°Cs-1)	Tf	Pf	
-	Etna7_C	1070	75	1800	-	-	1070	75	
	Etna7_B	1070	75	1800	0.2	0.2	1010	15	

Supplementary Table 4. Summary of X-ray microtomography acquisition conditions and image processing details of the volumes of interest (VOI) for experiment ET1150.

Energy	Distance sample to detector	Projection	Exposure time	Voxel size	Original imaged volume	Analysed VOI	Segmentation
keV	mm	number	s	μm^3	pixels (mm³)	mm^3	
53	2300	1800	0.05	3.2x3.2x3.2	1683x1683x1383 (128.4)	8.2037	semi-automatic volume segmentation

Avizo software v. 8.0