

Supplementary File 1

Dynamics of plagioclase textural evolution and the role of thermal history on the nucleation process

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BSE Images

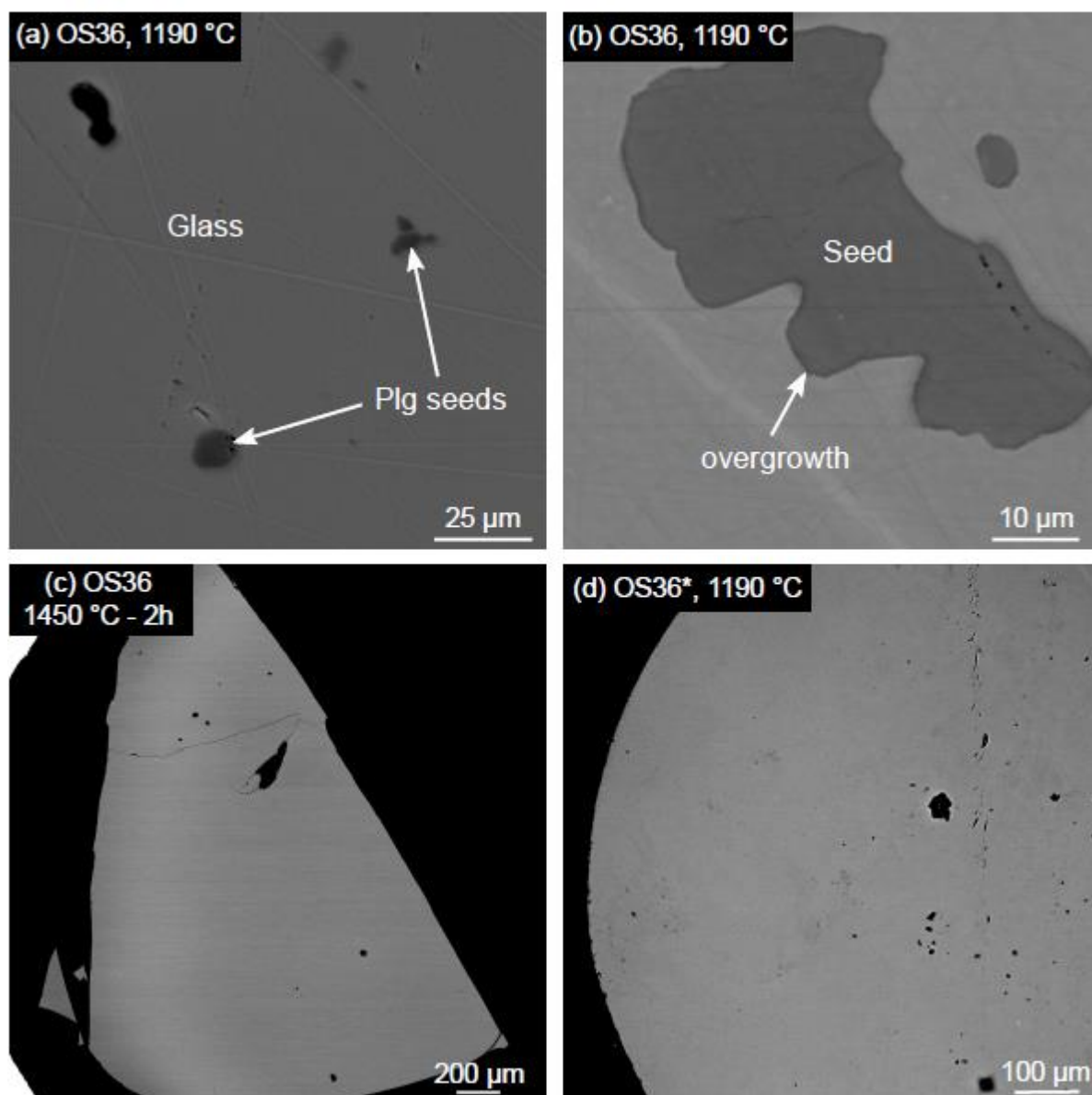


Fig. S1: **a, b** Plagioclase seeds observed after heating the OS36 starting material to 1190 °C for 24h. A possible early overgrowth around the seed is visible in panel (**b**). **c** Glassy material obtained after heating the OS36 starting material to 1450 °C for 2 h to produce the OS36* starting material, with no crystal fragments visible. **d** Glassy material obtained after heating the OS36* material to 1190 °C for 2 h.

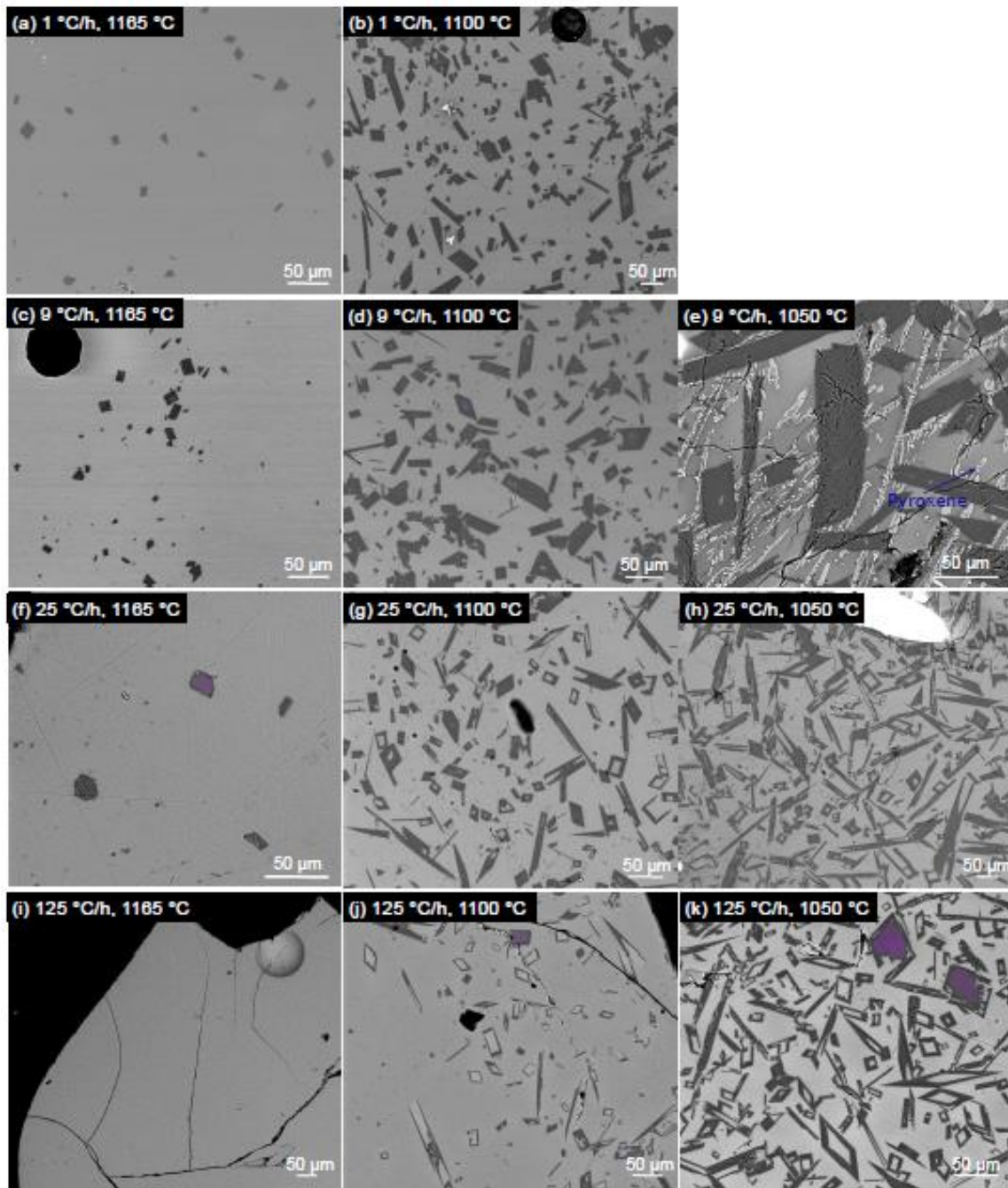


Fig. S2: BSE images of cooling runs conducted at **a, b** 1 °C/h, **c–e** 9 °C/h, **f–h** 25 °C/h, and **i–k** 125 °C/h, using the seed-bearing OS36 starting material initially heated to 1190 °C. The increase in cooling rate is marked by a global evolution from euhedral tabular/prismatic shapes at 1 °C/h (**b**) and 9 °C/h (**d–e**), to skeletal crystals with hopper cavities at 25 °C/h (**g–h**) and 125 °C/h (**j–k**). We also observed the depression of the liquidus temperature with increasing cooling rate; at 125 °C/h, no crystals are observed at 1165 °C (**i**). Purple areas in some panels correspond to the initial plagioclase fragment with an overgrowth part around.

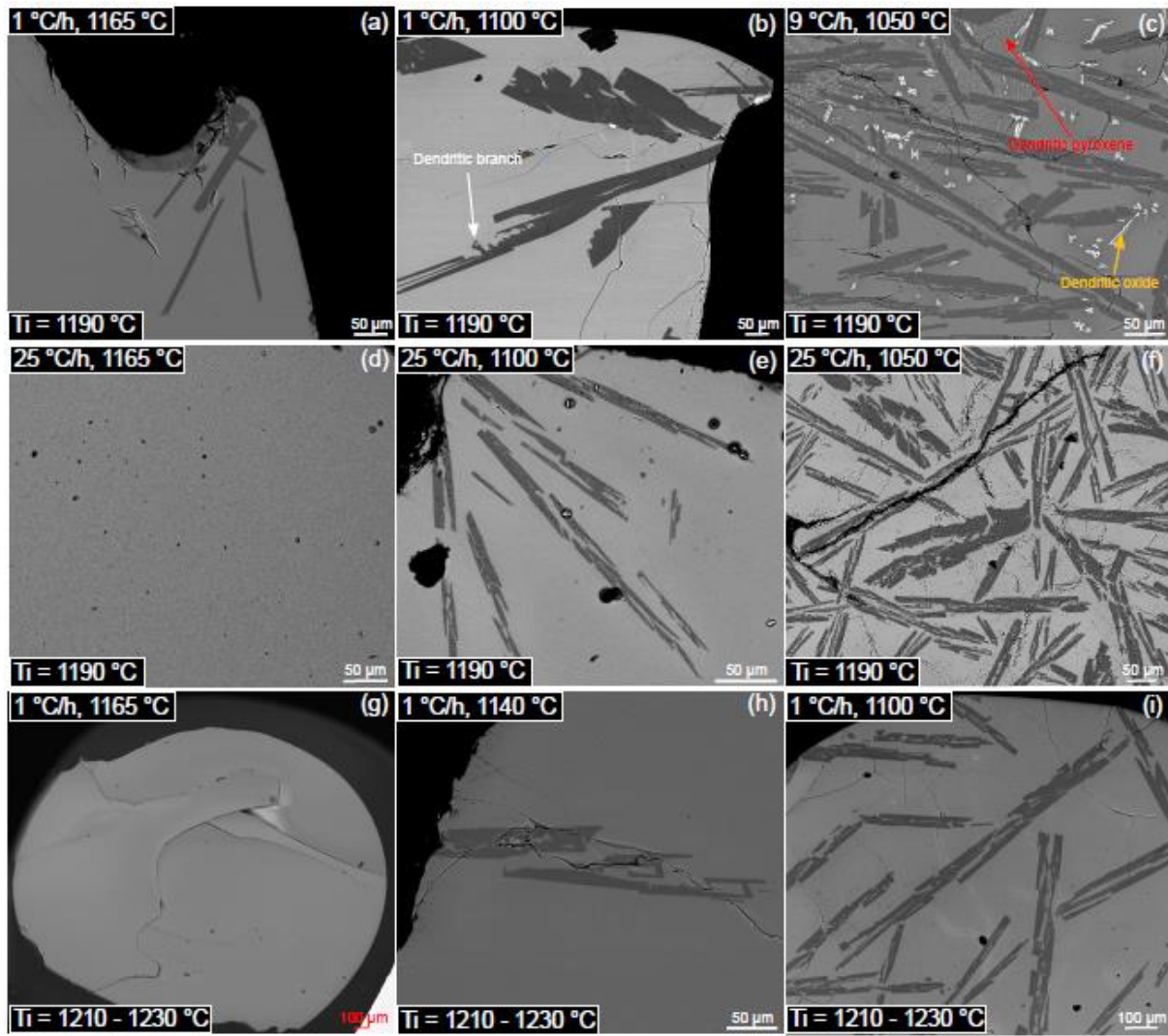


Fig. S3: BSE images of cooling runs conducted at **a, b** 1 °C/h, **c** 9 °C/h, and **d–f** 25 °C/h using the seed-free OS36* starting material initially heated to 1190 °C. Except for the run cooled at 1 °C/h and quenched at 1165 °C (**a**), which exhibits elongated plagioclase crystals, all other samples are characterized by dendritic plagioclase (**b–f**). Similar textures are observed in experiments involving a stronger initial superheating step to 35–55 °C above the liquidus temperature (**g–i**). Increasing the cooling rate (**d**) and the degree of initial superheating (**g**) leads to a decrease in the temperature at which nucleation begins.

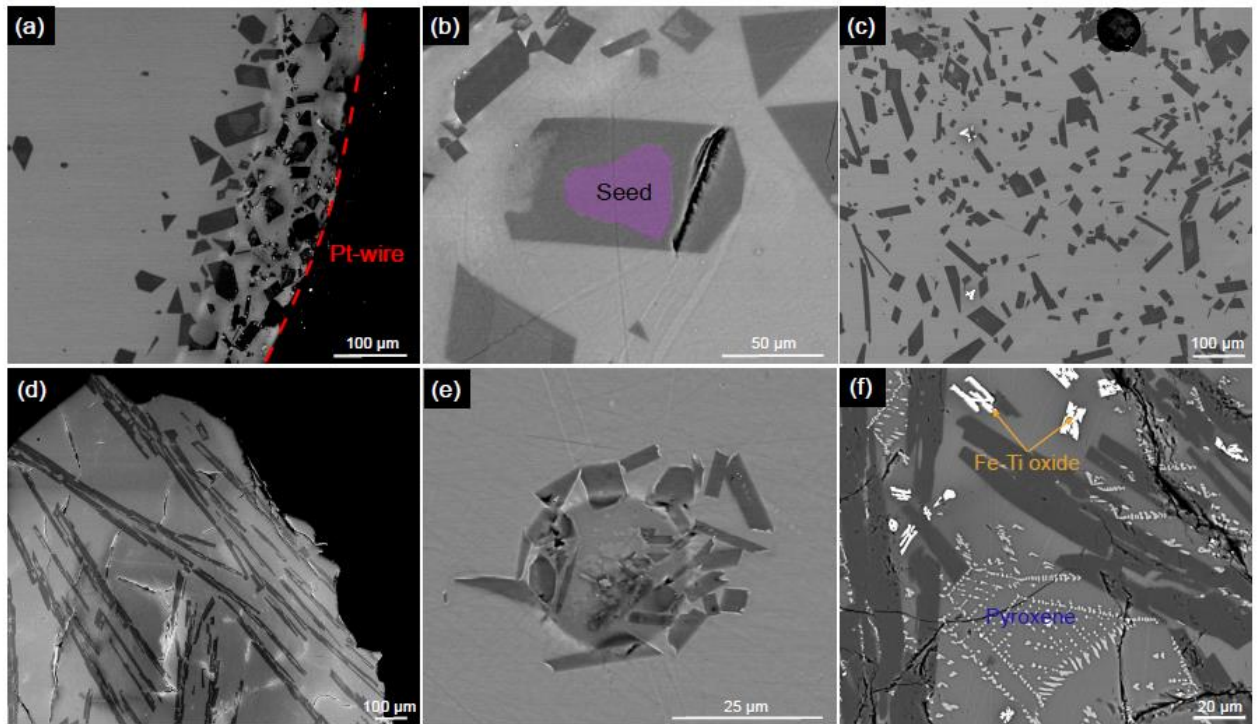


Fig. S4: BSE images illustrating the various types of nucleation observed in our experiments. **a, d** Plagioclase nucleation around the Pt wire and along the sample edges, respectively. The strong influence of the Pt-wire on nucleation is particularly evident at the onset of crystallization. **e, f** Heterogeneous nucleation on foreign substrates (e.g., bubbles and crystals of a different mineralogy). In contrast, we also observed **b** "self-nucleation," corresponding to overgrowth around a seed of the same crystallographic nature. Note the presence of a bright boundary layer surrounding the long axis of plagioclases nucleated from bubbles (**e**). **c** A typical texture from seed-bearing runs, where nucleation may result from either homogeneous or heterogeneous mechanisms, or a mixture of both.

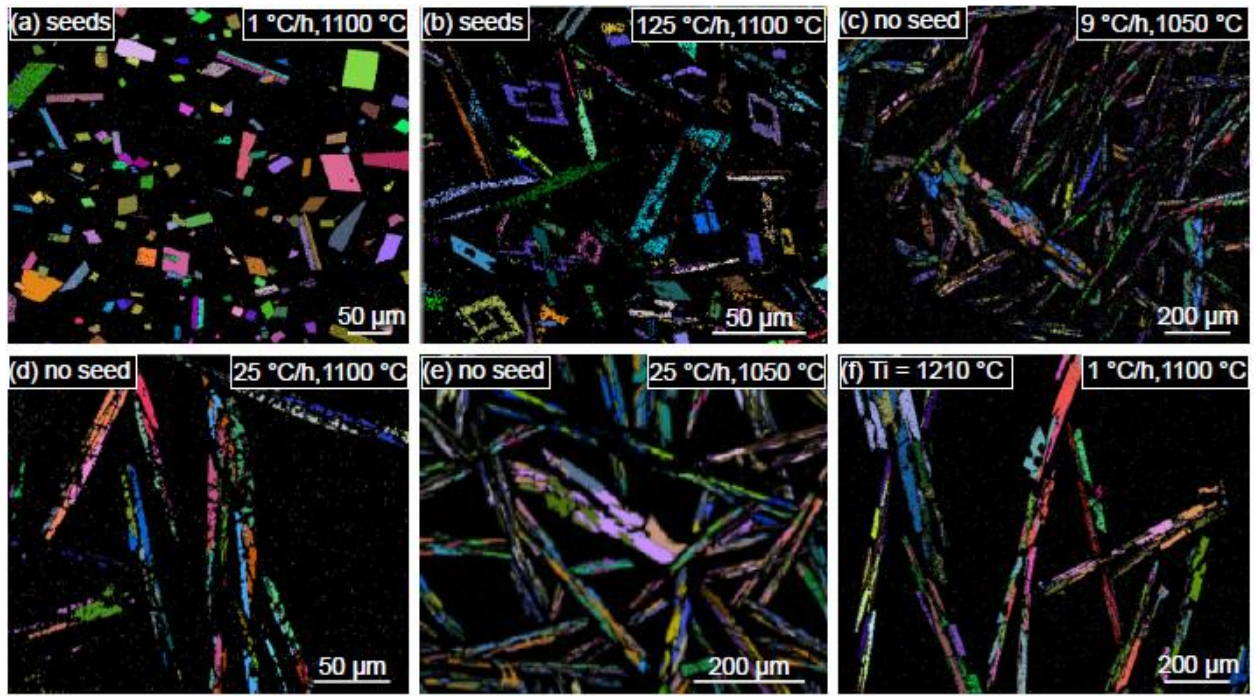


Fig. 5: EBSD images of twins in plagioclase crystals. **a, b** Crystals from seed-bearing experiments cooled at 1 °C/h and 125 °C/h, respectively. Most crystals in these runs do not contain twins. When present, twins are simple, involving only two twins. **c–f** Samples (seed-free) initially superheated to **c–e** $+\Delta T = 15$ °C and **f** $+\Delta T = 35$ °C. These samples contain complex twin boundaries associated with dendritic plagioclase textures.

Textural analysis

As in [Billon et al. \(2025\)](#), textural parameters such as phase proportions (φ_V), surface crystal and bubble number densities (N_A and N_p , determined by point counting in ImageJ), volumetric crystal number density (N_V), and the global mean crystal size (S_N) were calculated from the entire image of each sample as:

$$\varphi_V = \frac{\text{Total Plagioclase area}}{\text{Sample area}} \quad (\text{S1})$$

$$N_A = \frac{\text{Plagioclase crystal number}}{\text{Sample area}} \quad (\text{S2})$$

$$N_p = \frac{\text{Bubble number}}{\text{Sample area}} \quad (\text{S3})$$

$$S_N = \sqrt{\frac{\varphi_V}{N_A}} \quad (\text{S4})$$

$$N_V = \frac{N_A}{S_N} \quad (\text{S5})$$

Nucleation and growth rates calculated by the Batch, l_{mean} , and l_{max} methods) were then calculated from the global images of each experiment as:

$$G_{\text{max,l}} = \frac{l_{\text{max,av}}}{2t} \quad (\text{S6})$$

$$G_{\text{mean,l}} = \frac{l_{\text{mean}}}{2t} \quad (\text{S7})$$

$$J_{\text{Batch}} = \frac{N_V}{t} \quad (\text{S8})$$

Crystal size parameters (l_{mean} , $l_{\text{max, av}}$) were measured from the various subsectional images, and the results were then combined to derive the final data for the overall sample. Incremental nucleation (J_i) and growth (G_i) rates were also determined for runs with various final dwell

durations at the same final temperature to measure the nucleation and growth between two final dwell times (t_2 and t_1):

$$G_i = \frac{(l_{\text{mean}}/l_{\text{max,av}})_2 - (l_{\text{mean}}/l_{\text{max,av}})_1}{2 \times (t_2 - t_1)} \quad (\text{S9})$$

$$J_i = \frac{(N_V)_2 - (N_V)_1}{t_2 - t_1} \quad (\text{S10})$$

Analytical techniques

SEM and EPMA analyses

EDS (Quanta-650F FEG-SEM at Cambridge University in England; Tescan MIRA 4 SEM at KULeuven in BELGIUM) and WDS (CAMECA SX Five Tactics at the LMV in Clermont-Ferrand, France; JEOL JXA-8530F at the Department of Material Engineering, KU Leuven, Belgium; and JEOL JXA-ISP 100 in Aachen, Germany) analyses of glasses (using a defocused beam) and plagioclase crystals (focused beam) were performed in continuity with those of [Billon et al. \(2025\)](#). SEM imaging was performed at a working distance of 10–15 mm, with a dwell time of 10 μs and a voltage of 15 KV (20 KV for EDS analyses, with a beam current of 6–9 nA). Calibration details for WDS analyses are provided below.

The CAMECA SX Five Tactics (Clermont-Ferrand) calibration standards were the same as those of [Bechon et al. \(2022\)](#): wollastonite (Ca, Si), TiMnO_3 (Ti, Mn), Al_2O_3 (Al), fayalite (Fe), forsterite (Mg), albite (Na), Cr_2O_3 (Cr), orthoclase (K), and NiO (Ni).

Standards used for the JEOL JXA-8530F (KULeuven) and JEOL JXA-ISP 100 (Aachen University) were the same as those used by [Billon et al. \(2025\)](#):

- JEOL JXA-8530F (KULeuven):

- Obsidian (SiO_2 , Al_2O_3 , K_2O), apatite (P_2O_5), diopside (CaO), hematite (FeO), jadeite (Na_2O), olivine (MgO), rutile (TiO_2), rhodonite (MnO), chromium oxide (Cr_2O_3), and nickel (NiO) for glasses.
- Albite (Al_2O_3), diopside (SiO_2), apatite (P_2O_5), hematite (FeO), jadeite (Na_2O), olivine (MgO), obsidian (K_2O), diopside (CaO), rutile (TiO_2), chromium oxide (Cr_2O_3), and rhodonite (MnO) for plagioclase.
- JEOL JXA-ISP 100 (Aachen University):
 - Obsidian (SiO_2 , Al_2O_3 , K_2O), jadeite (Na_2O), olivine (MgO), apatite (P_2O_5), diopside (CaO), rutile (TiO_2), hematite (FeO), Mn oxide (MnO), and chromite (Cr_2O_3) for glasses.
 - Plagioclase (SiO_2 , Al_2O_3 , CaO), jadeite (Na_2O), olivine (MgO), apatite (P_2O_5), orthoclase (K_2O), rutile (TiO_2), hematite (FeO), Mn oxide (MnO), and chromite (Cr_2O_3) for plagioclase.
 - Plagioclase (Al_2O_3), olivine (SiO_2 , MgO), rutile (TiO_2), Magnetite (FeO), Mn oxide (MnO), chromite (Cr_2O_3), nickel (NiO) for magnetite.

The type of analysis (EDS/WDS) and the institution where analyses were performed are reported in *Supplementary file 2*. In the case of EDS analyses performed at KU Leuven and WDS data, the same secondary plagioclase and glass standards (compositions provided in *supplementary file 2*) were used to correct the data.

EBSD analyses

All electron backscatter diffraction (EBSD) measurements were conducted at the University of Lille using a HITACHI SU5000 field-emission scanning electron microscope (FE-SEM) equipped with an Oxford Instruments Symmetry EBSD detector (CMOS). Analyses

were performed at an accelerating voltage of 20 kV, with a beam current of a few nA, under a partial vacuum of 30 Pa, removing conductive coating, unsuitable for EBSD measurements. The samples were tilted 70° towards the EBSD detector. Diffraction patterns were acquired at high acquisition speed with a resolution of 156×128 pixels, integrating 2–3 frames per measurement.

Prior to EBSD analysis, samples were polished using $\frac{1}{4} \mu\text{m}$ diamond suspension, followed by final surface preparation with a Fishione Model 1061 ion polisher operated at 5 keV accelerating voltage, 5° milling angle, 50 % beam focus, and continuous rotation of the sample for 5 minutes.

Chemical results

Plagioclase analysis

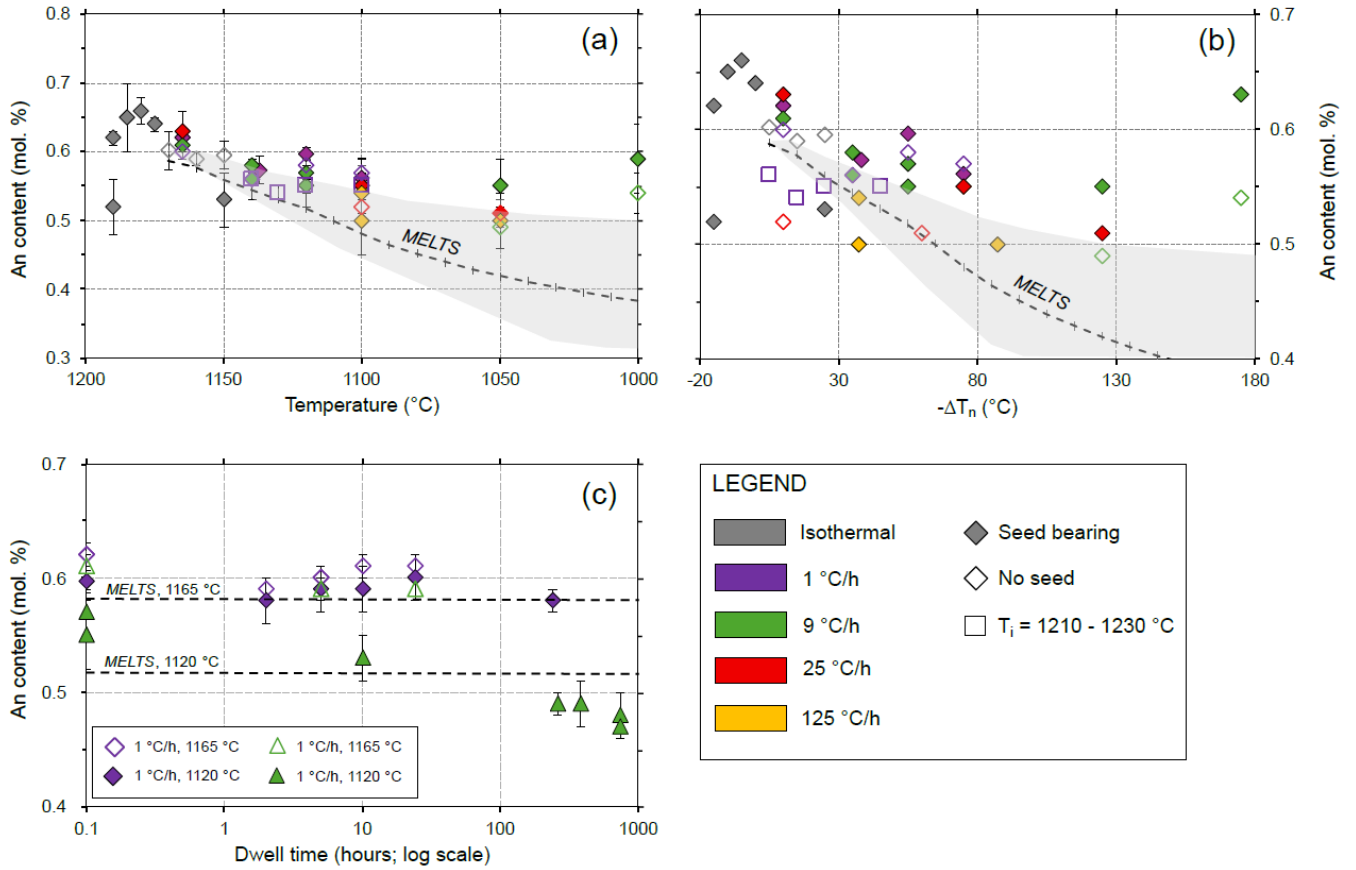


Fig. S1: Evolution of plagioclase anorthite content as a function of the solidification path. **a, b** Evolution during isothermal and cooling experiments, plotted against temperature (**a**) and nominal degree of undercooling (**b**). **c** Evolution as a function of the final dwell duration following cooling at 1 and 9 °C/h.

As explained by [Billon et al. \(2025\)](#), the anorthite contents of newly nucleated crystals or overgrowth rims were quite homogeneous, with 1σ standard deviations < 0.05 (see **Supplementary File 3**). The slightly higher anorthite content observed in our experiments compared to the Rhyolite-MELTS predictions, particularly below 1100 °C (**Fig. S1a, b**), can be attributed to the lower nucleation temperature of clinopyroxene in our runs (~ 1050 °C), as opposed to 1110–1115 °C estimated by Rhyolite-MELTS. This delay in clinopyroxene crystallization resulted in a relative enrichment of Al_2O_3 and CaO in the melt, thereby increasing the anorthite content of crystallizing plagioclase. Increasing the cooling rate and the degree of initial superheating appears to have decreased the anorthite content of crystallizing plagioclase (**Fig. S1a and b**). For example, in seeded experiments cooled at 25 and 125 °C/h, An contents

ranged from 50 to 54 An%, whereas in experiments cooled at 1 and 9 °C/h, they ranged from 57 to 62 An%.

The addition of a final dwell duration shorter than 24 h did not impact plagioclase anorthite content (**Fig. S1c**). Runs quenched after final dwell durations >240 hours (particularly experiments previously cooled at 9 °C/h) showed a significant decrease of the anorthite content to around 47–49 An%. This strong decrease was simultaneous to the second nucleation event (**Fig. 6f**).

Glass analysis

Isothermal and cooling runs

Despite minor variations, the overall liquid composition in isothermal runs (with or without pre-heating) was consistent with Rhyolite-MELTS predictions. [Billon et al. \(2025\)](#) showed that in seed-bearing runs cooled at 1 to 9 °C/h, the melt evolves from basaltic-andesitic/andesitic to dacitic as temperature drops to 1000 °C. Samples initially quenched at 25 and 125 °C/h follow a similar trend, though with a residual enrichment in MgO, Al₂O₃, and CaO (**Fig. S2 a, c, g**). The sharp increase in MgO at 1050 °C from 9 to 125 °C/h correlates with decreasing clinopyroxene content, down to 1% and 0% at 25 and 125 °C/h, respectively.

In runs superheated to 1450 °C to remove plagioclase seeds, the residual melt composition depended on cooling rate. Despite slight enrichments in FeO(t), likely due to lower Fe-Ti oxide content, and CaO, the general melt evolution remains comparable at 1 °C/h (**Fig. S2 b, d, h**). However, samples cooled at 25 °C/h showed distinct melt compositions. The decrease in nucleation temperature between 1165 and 1100 °C did not significantly affect chemistry, which we attribute to the limited plagioclase proportion: from <1% at 1165 °C to only ~5% (mostly along sample edges) at 1100 °C. At 1050 °C, plagioclase proportions reached ~36% and MgO content increased (possibly due to the absence of other phases like clinopyroxene), while Al₂O₃ and CaO contents decreased slightly (**Fig. S2 b, d**). A similar Al₂O₃

enrichment between 1165 and 1130 °C was observed in runs initially heated to 1210–1230 °C, likely also due to delayed nucleation.

The strong FeO(t) depletion observed (**Fig. S2 f**) is harder to explain. It likely involves more than just the absence of clinopyroxene and Fe-Ti oxides. Iron loss during the experiments, possibly due to the run durations, may be a contributing factor.

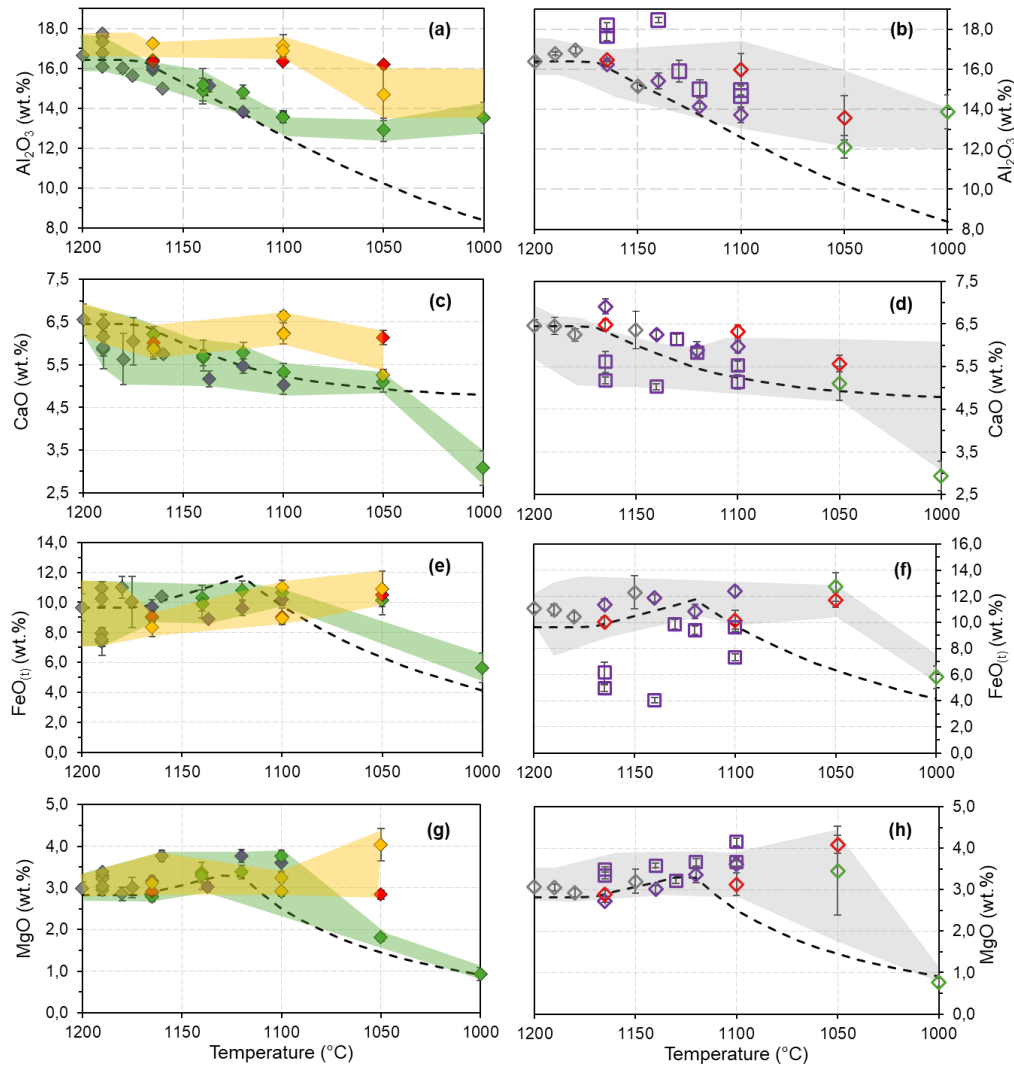


Fig. S2: Evolution of the residual liquid composition as a function of quench temperature. **a, c, e, g** Seed-bearing starting materials. Green and yellow shaded areas refer to the compositions at slow (1 and 9 °C/h) and fast cooling rates (25 and 125 °C/h), respectively. **b, d, f, h** Crystal-free starting materials at various degrees of initial superheating ($+\Delta T = 15, 35\text{--}55$ °C). For reference, the gray shaded area indicates the range of chemical evolution observed in the seed-bearing runs.

The maturation step

It is difficult to discern any clear trend in the chemical evolution of the residual liquid. Despite minor variations, the composition at the onset of crystallization (1165 °C) remained relatively stable for dwell durations between 2 and 24 h. More pronounced fluctuations nevertheless occurred at 1120 °C ($-\Delta T_n = 55$ °C). Samples previously cooled at 1 °C/h generally showed relatively stable Al_2O_3 , CaO, MgO, and FeO(t) contents throughout the dwell period from 5 to 240 h (Fig. S3). For the longest dwell times (>240 hours), a decrease in FeO(t) and MgO contents was observed between 264 h (10.18 wt.% FeO(t) and 3.7 wt.% MgO) and 744 h (8.1 wt.% FeO(t) and ~3.5 wt.% MgO), possibly due to the crystallization of clinopyroxene (as evidenced by abundant quench crystals and a few phenocrysts) and a late increase in Al_2O_3 content (Fig. S3a, c, d).

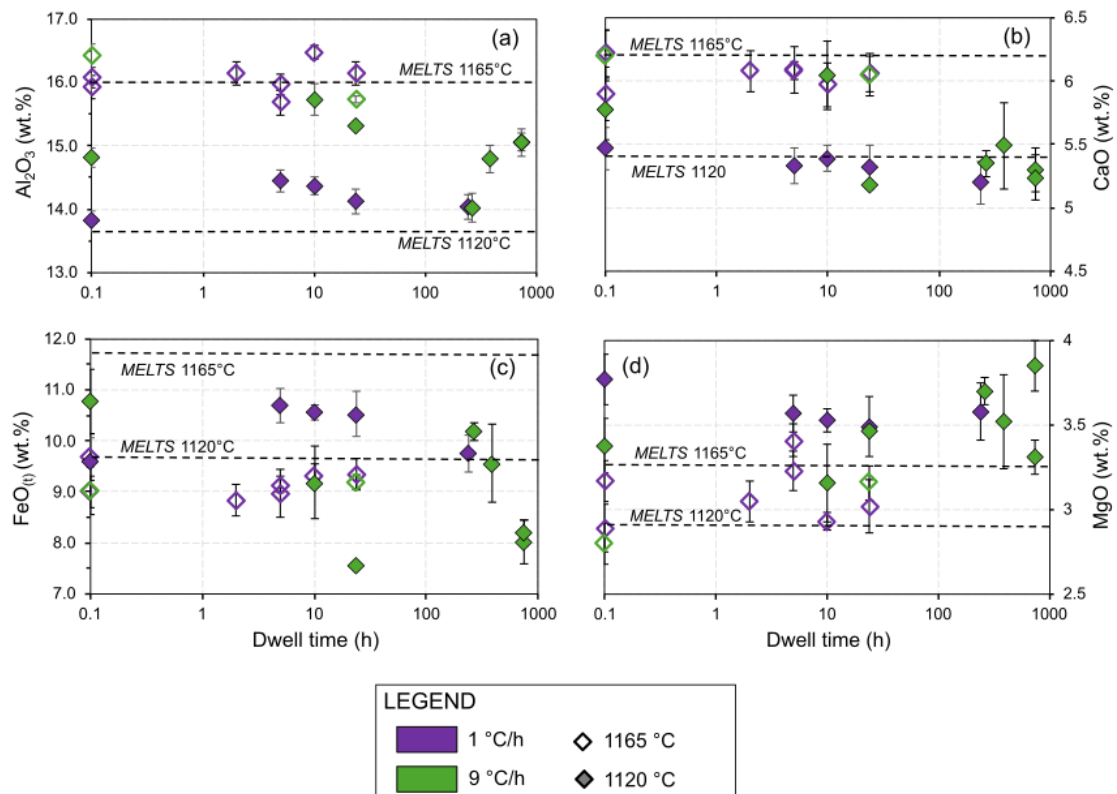


Fig. S3: Evolution of the residual liquid composition as a function of the final dwell duration following cooling at 1 and 9 °C/h. **a–d** Concentrations of Al_2O_3 , CaO, FeO(t), and MgO, respectively.

REFERENCES

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- Billon, M., Vander Auwera, J., Namur, O., Faure, F., Holness, M. B. & Charlier, B. (2025). Plagioclase crystal size distributions, growth and nucleation rates in an anhydrous arc basaltic andesite. *Contributions to Mineralogy and Petrology* **180**.