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1 **Nucleation delay in water-saturated rhyolite during decompression in shallow volcanic systems**
2 **and its implications for ascent dynamics**

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8

9 **Abstract**

10 We developed a model of feldspar nucleation delay during decompression in water-saturated rhyolite
11 based on classical nucleation theory (CNT). With the thermodynamic and kinetic parameters proposed
12 in this study, the model agrees with the experimental results of this study to within a factor of two. To
13 account for the variety of conditions encountered by magma during ascent, we propose expanding the
14 model, developed in this study for isothermal decompression, to a wider range of pressures and
15 temperatures (i.e., non-isothermal conditions). We present new measurements of feldspar growth rates
16 during decompression in water-saturated rhyolite, emphasizing the importance of accurately calculating
17 the nucleation delay in order to estimate growth rates. Using these data, we propose a method for
18 estimating the ascent rate of water-saturated rhyolite based on the nucleation delay of feldspar microlites.
19 The findings of this study hold the promise of a better understanding of volcanic timescales during
20 magma ascent in the conduit, as well as the possibility of extending our model to more complicated
21 scenarios with changing pressure and temperature.

22 **Keywords:**

23 nucleation delay, ascent rate, crystallization, decompression, magmatic timescales

24 **Introduction**

25 Microlites, the crystals less than few hundred micrometers in size, of which plagioclase is the most
26 common, form on the timescales of minutes to a few days during magma ascent and subsequent degassing
27 (e.g., Sharp et al. 1996, Hammer et al. 1999, Hammer and Rutherford 2002, Martel 2012, Mollard et al.
28 2012). Because they form on the timescales of ascent their presence and textures can provide valuable
29 information about magma ascent rates and conditions in the conduit. Moreover, timing of crystallization
30 can affect the production of crystal-rich or crystal-poor magmas, resulting in changes in the textural
31 make-up of the produced rock as well as the explosivity. Magma composition and exsolution of dissolved
32 volatiles are the primary determinants of eruption style and timing. The presence of crystals, on the other
33 hand, can impact volatile exsolution by acting as nucleation sites for bubble nucleation (e.g., Hurwitz
34 and Navon 1994, Gardner and Denis 2004, Shea 2017, Pleše et al. 2018) and magma rheology (D'Oriano
35 et al. 2005, Mader et al. 2013, Vona et al. 2017). The permeability of vesicular magma can be affected
36 by crystallization during ascent, which can lead to changes in eruption style by lowering the percolation
37 threshold and changing the eruptive behaviour from explosive to effusive (e.g., Lindoo et al. 2016,
38 Cassidy et al. 2018, deGraffenried et al. 2019). Notably, magmas with ~4.0-5.5 wt. % water (around the
39 water saturation limit depending on storage conditions) and <30 vol. % crystallinity erupt mostly
40 explosively, but magmas with >40 vol. % crystallinity erupt primarily effusively (Popa et al. 2021).

41 The first order transition i.e., nucleation initiates the process of partial or complete crystallization of a
42 silicate melt (e.g., Kirkpatrick 1981, De Yoreo and Vekilov 2004, Fokin et al. 2006). Nucleation is the
43 process of rearrangement of atoms (or molecules) in a parent phase that results in the formation of a
44 cluster of a new, thermodynamically more stable phase (Kirkpatrick 1981, De Yoreo and Vekilov 2004,
45 Fokin et al. 2006). In magmatic systems, however, nucleation does not occur immediately when the new
46 phase becomes thermodynamically stable, instead can be delayed by minutes, hours, or days (e.g., Fenn
47 1977, Swanson 1977, Couch 2003, Mollard et al. 2012, Polacci et al. 2018, Rusiecka et al. 2020).

48 Understanding the magmatic timescales of volcanic systems can be improved by studying nucleation
49 kinetics, particularly nucleation delay. Modelling nucleation delay allows for the prediction of the

50 timescales on which igneous rocks crystallize from a melt and when they do remain vitreous (Rusiecka
51 et al. 2020). Numerous studies have been conducted to investigate crystallization kinetics in felsic melts
52 (e.g., Fenn 1977, Swanson 1977, Hammer and Rutherford 2002, Couch 2003, Brugger and Hammer
53 2010, Mollard et al. 2012, Riker et al. 2015, Sirbescu et al. 2017, Mollard et al. 2020, Rusiecka et al.
54 2020). However, the majority of these studies have been focused on constraining the crystal growth and
55 nucleation rates. A few of them provided incidental measurements of nucleation delays (e.g., Fenn 1977,
56 Swanson 1977, Couch 2003, Mollard et al. 2012, Sirbescu et al. 2017). Rusiecka et al. (2020) presented
57 a model based on Classical Nucleation Theory (CNT) for the nucleation delay of plagioclase, pyroxene
58 and olivine in basaltic melts, and of quartz and alkali feldspar in a hydrous, rhyolitic melt under isobaric
59 conditions.

60 In this contribution we build on the model presented by Rusiecka et al. (2020) by developing a theoretical
61 model of feldspar nucleation delays in rhyolitic melts during ascent in the conduit. The model was
62 calibrated by a series of decompression-induced crystallization experiments conducted using a hydrated
63 obsidian at temperatures ranging from 800-900 °C and pressures ranging from 25-200 MPa. This
64 temperature range is representative of natural rhyolite temperatures (e.g., Castro and Dingwell 2009) and
65 decompression from 200 to 25 MPa covers the range of pressures prevailing during magma ascent from
66 a deep reservoir (200 MPa, around 7.5 km below the Earth's surface) to near-surface levels (Martel, 2012).

67 **Classical Nucleation Theory**

68 Volmer and Weber (1926), Becker and Döring (1935), and Turnbull and Fisher (1949) developed classical
69 nucleation theory during the first half of the twentieth century on the basis of Gibbs' (1875-1878) earlier
70 work on the thermodynamic description of heterogeneous systems. According to Gibbs' theory, a
71 heterogeneous system, consisting of a parent phase and a new nucleating phase, is replaced for simplicity,
72 by two homogeneous phases (the parent and a nucleus) divided by a mathematical surface of zero
73 thickness. A change in Gibbs free energy between the newly formed nucleus and the parent phase drives
74 nucleation. The parent phase is assumed to be in a metastable state, and nucleation must occur after an

75 activation energy barrier is overcome. The rate of nucleation is affected by the degree of supersaturation,
76 which is commonly measured as undercooling below the liquidus temperature. When the new nuclei are
77 surrounded by the previously existing phase, homogeneous nucleation occurs, and heterogeneous
78 nucleation occurs when the nucleating phase forms on a previously existing surface (Kirkpatrick 1981,
79 De Yoreo and Vekilov 2004, Fokin et al. 2006).

80 **Methods**

81 **Experimental procedure**

82 The starting sample used in the experiments is the Lake County Obsidian (LCO) – a common
83 metaluminous composition (in wt. % on an anhydrous basis: 75.7 SiO₂, 0.1 TiO₂, 13.2 Al₂O₃, 0.7 FeO,
84 0.1 MnO, 0.1 MgO, 0.6 CaO, 4.3 Na₂O, 5.1 K₂O; Maneta and Baker 2014). This is the same material as
85 used to investigate cooling-induced crystallization by Rusiecka et al. (2020) and Rusiecka and Baker
86 (2021). The obsidian was coarsely crushed and melted at 1400 °C for 4 hours before being ground to a
87 fine powder. About 30 mg of the LCO glass powder was loaded into 15 mm length 2.9 mm OD (outer
88 diameter), 2.5 mm ID (inner diameter) gold capsules with enough water to ensure saturation (as
89 calculated using MagmaSat, Ghiorso and Gualda 2015) and welded shut. To check for leaks, the capsules
90 were kept at 120 °C for at least 2 hours and weighed. The experiments were carried out at the Institut des
91 Sciences de la Terre d'Orleans in Orleans (France) in an Internally Heated Pressure Vessel (IHPV) with
92 argon as the pressurizing medium. The capsules were placed in the 3-cm hot spot of the furnace and
93 temperature (± 4 °C) was controlled at the top and bottom of the samples using two S-type thermocouples.
94 The pressure (± 2.5 MPa) was monitored by a high pressure transducer calibrated against a 700 MPa
95 Heise gauge. The liquidus temperature at different pressures was determined using rhyolite-MELTS
96 (Ghiorso and Sack 1995, Gualda et al. 2012) and confirmed experimentally at 200, 150 and 50 MPa to
97 constrain the effective degree of undercooling ($\Delta T_{\text{eff}} = \text{run temperature} - \text{liquidus temperature}$, Table 1).
98 To confirm that the starting material was free of pre-existing crystals we conducted an experiment in
99 which the samples were kept for 24 h at 825 °C and 200 MPa and then decompressed at 10 MPa/min to

100 50 MPa. They were then immediately isobarically quenched. Experiments were first kept at 200 MPa
101 and 825 °C for 24 hours to homogenize the melt with water and then decompressed at 10 MPa/min to
102 90, 75, 50 and 25 MPa (Table 1).

103 **Analytical techniques**

104 After the experiments, the capsules were first weighted to confirm that no water loss occurred during the
105 experiment. Then they were opened, and the samples were recovered as glass chips that were placed in
106 epoxy resin and polished for analyses. The samples were imaged using a Merlin Compact Zeiss scanning
107 electron microscope (SEM) at the Institut des Sciences de la Terre d'Orléans in Orléans. Although most
108 of the crystals were too small for thorough quantitative analysis the observed crystals were identified as
109 plagioclase feldspar (i.e., in the oligoclase field in the experiment conducted at 50 MPa; semi-quantitative
110 composition: $An_{28}Ab_{65}Or_7$) and magnetite using a Bruker Quantax XFlash6 energy dispersive
111 spectrometer coupled to the SEM. The crystallinity of the samples did not exceed ~1-2 vol. %. The water
112 saturation content was calculated using the MagmaSat application, which employs the Ghiorso and
113 Gualda (2015) H_2O-CO_2 saturation model.

114 **Theoretical model**

115 Nucleation delay (τ) is the time between when the parent phase (silicate melt) is cooled below its liquidus
116 temperature and the onset of crystallization. It is defined as:

$$117 \quad \tau = \frac{16h}{\pi} \frac{\sigma}{\Delta G_V^2 a^4} \exp \frac{E_a}{RT} \quad (1)$$

118 where h is Planck's constant, R is the gas constant, σ is the interfacial free energy between the parent
119 phase and the forming nucleus. The value for the interfacial free energy was chosen to be 0.18 J/m², this
120 being the value of the interfacial free energy for silicate crystals in the system $Na_2O-CaO-SiO_2$ (Fokin et
121 al. 2006). The interfacial free energy, this being σ , is poorly constrained and for feldspar in a silicate melt
122 can vary between 0.10 and 0.25 J/m² (e.g., Hammer 2004, Baker et al. 2020, Mollard et al. 2020, Rusiecka
123 et al. 2020). Free energy of crystallization per unit volume ΔG_V (Robie and Bethke 1962) was calculated

124 using the MELTS software (Ghiorso and Sack 1995, Gualda et al. 2012). Energy of crystallization, ΔG
125 was obtained using the affinities of feldspar in the melt of the same composition as the starting material
126 at the temperature of the experiments (825 °C) and a range of pressures (10 to 200 MPa). The affinities
127 were plotted against pressure and fitted with a trend line. The equation describing the trend line was used
128 in the calculation of the ΔG in the nucleation delay program. Parameter a in Equation (1) is the size of a
129 structural unit, here approximated using the size of the Si^{4+} ion (0.26×10^{-10} m, Shannon 1976), as Si^{4+}
130 is an essential component of both a silicate melt and its crystals. This value was previously successfully
131 used by Rusiecka et al. (2020) for nucleation of quartz and feldspar in a natural, hydrous rhyolite, and by
132 Baker et al. (2020) for anorthite and diopside in a synthetic composition. The activation energy for
133 nucleation (E_a , the kinetic barrier) was approximated using the diffusivity calculated using the Eyring
134 equation (Eyring 1935):

$$135 \quad E_a = -k_B T \ln \left(\frac{h}{\lambda^3 \eta} \right) \quad (2)$$

136 where η is viscosity and λ is the size of the diffusive jump and k_B is the Boltzmann's constant. The
137 diffusive jump is not well constrained and can generally vary between 1^{-10} to 5^{-10} m (0.1 to 0.5 nm, e.g.,
138 Baker 1990, 1992, Mungall 2002, Gonzalez-Garcia et al. 2017). The variation of the diffusive jump from
139 1^{-10} m to 5^{-10} m corresponds to variation in a nucleation delay of two orders of magnitude. The viscosity
140 for the Eyring equation was calculated using model by Giordano (2008) with water contents calculated
141 at saturation at the different run pressures using the MagmaSat program (i.e., 1.8 to 4.8 wt. % H_2O from
142 25 to 120 MPa). Rusiecka et al. (2020) proposed that activation energy for the nucleation needs to be
143 modified by using diffuse interface theory. However, this is not the case in this study as using the
144 parameters for water saturated melt and changing the water content at saturation with pressure replicates
145 the experimental results to within a factor of two. This variation might be caused by the differences in
146 water content and the distribution of water in the melt. In the case of Rusiecka et al. (2020) water content

147 was 4 wt. %, which is several wt. % below the saturation content at the experimental conditions (600
148 MPa) whereas in this study all the experiments were water saturated.

149 The numerical constant in Equation 1 ($\frac{16}{\pi}$) is that of Fokin et al. (2006); different derivations of Equation
150 (1) (e.g., Collins 1955, Kashchiev 1969, Slezov and Schmelzer 1999, 2002) can lead to slightly different
151 constant values, but they typically vary by less than an order of magnitude (Gutzow and Schmelzer 1995).

152 **Results**

153 **Description of the textural characteristics of the experimentally produced plagioclase**

154 All samples produced in experiments conducted in this study are vesicular and contain magnetite.
155 Although magnetite may be seen as inclusion in some feldspars on the SEM images, it is not considered
156 here as a nucleation site for the silicate crystals for two main reasons. Firstly, the distribution of the oxides
157 in connection to the plagioclase is random (i.e., there are more oxide sites than plagioclase crystals).
158 Secondly, the magnetite distribution within the plagioclase crystals is random (i.e., magnetite is not
159 systematically in the centre of the plagioclase where growth would presumably have started in case of
160 magnetite acting as a preferential nucleation site). The sub-micron oxides were likely included in the
161 plagioclase during rapid growth. Plagioclase was produced at three different pressures: 90, 75 and 50
162 MPa (corresponding to $\Delta T_{\text{eff}} = -10, -20$ and -55 °C). The representative images of the samples produced
163 at those conditions can be observed in Figure 1. Magnetite and plagioclase crystals are homogeneously
164 distributed in the sample and did not preferentially nucleated at capsule walls. At 50 MPa ($\Delta T_{\text{eff}} = -55$ °C)
165 the crystals are highly irregular with skeletal morphologies (Fig. 1a) and numerous embayments and melt
166 inclusions (Fig. 1b). At 75 MPa ($\Delta T_{\text{eff}} = -20$ °C) the crystals are more regular than at 50 MPa and have
167 more prismatic shapes, nevertheless some irregularities, melt embayments and inclusions persist (Fig.
168 1c). At 90 MPa ($\Delta T_{\text{eff}} = -10$ °C) the crystals are elongated, with euhedral and subhedral morphologies,
169 well faceted crystals (Fig. 1d), and some clustering of smaller crystals.

170 **Nucleation delay**

171 The theoretically derived curve describing nucleation delay of plagioclase in hydrous, metaluminous
172 rhyolite at the conditions of the experiments in this study is shown in Figure 2 along with the experimental
173 results. In our study, the shortest delay is 30 h at $\Delta T_{\text{eff}} -50$ °C, and the delay tends to infinity as the
174 undercooling approaches zero. The findings of our study, as well as our modeling of the nucleation delay
175 in decompression induced crystallization, were compared to those of Mollard et al. (2012), who
176 conducted experiments on a rhyolitic melt using a crystal free starting material and reported nucleation
177 delays in their study (nucleation delays are given in Fig. 7 of Mollard et al. 2012). We initialized
178 nucleation delay model with a value for interfacial free energy of 0.18 J/m^2 , and bracketed it with values
179 ranging from 0.10 J/m^2 to 0.25 J/m^2 . Our theoretical modelling is specific to the composition used in this
180 study, but agrees well with other rhyolitic compositions represented by the synthetic composition of
181 Mollard et al. (2012), as shown here in Figure 2. Considering both the experiments conducted in this
182 study as well as the experiments of Mollard et al. (2012), the model agrees with the experimental data
183 by a factor of two, which is also the uncertainty represented by the typical variation in range of the
184 interfacial free energy.

185 **Growth rates**

186 We calculated the growth rates of feldspar (G_{max}) in the samples where crystallization was observed. To
187 do this we used ten largest crystals from each sample and applied growth rate equation (e.g., Couch 2003,
188 Arzilli and Carroll 2013, Arzilli et al. 2020, Rusiecka and Baker 2021) modified to include nucleation
189 delay:

$$190 \quad G_{\text{max}} = \frac{(L/2)}{(t-\tau)} \quad (3)$$

191 Here, L is the length of the longest crystal dimension (in m), t is the duration of the experiment (in s) and
192 τ is the nucleation delay calculated using the model described above. The spreadsheet with all the growth
193 rates obtained is available in the Supplementary Material.

194 Growth rates for pressures of 90, 75 and 50 MPa (corresponding to $\Delta T_{\text{eff}} = -10, -20$ and -55 °C) are shown
195 in Figure 3. At the conditions of these experiments (i.e., moderate to low degrees of undercooling) the
196 growth rates are on the order of 10^{-11} m/s (10^{-8} mm/s), and increases from the lowest degree of
197 undercooling to the highest degree of undercooling.

198 When plotting the growth rates for experiments conducted at 50 MPa ($\Delta T_{\text{eff}} = -55$ °C) with different
199 durations (48, 72 and 120 h, Fig. 4), higher by about a factor of two in the first, shorter experiment than in
200 the subsequent, longer experiment. Growth rates decrease with time is not unexpected. We expect non-
201 linear behaviour due to the rapid decay of initial steep gradients in chemical potential (c.f., Moschini et
202 al. 2021). The decay in the growth rates can be best represented semi-quantitatively by fitting the
203 experimental results with a trend line, as in Figure 4.

204 Another way of looking at growth rates is by comparing them to the equilibrium growth rate (G) as
205 calculated using the dependence of growth rate on the degree of undercooling (Equation 4; Toramaru
206 1991, Moschini et al. 2021):

$$207 \quad G \propto \exp\left(\frac{-E_a}{RT_{\text{exp}}}\right) \left(1 - \exp\left(\frac{\Delta H \Delta T}{RT_{\text{exp}} T_L}\right)\right) \quad (4)$$

208 Here, E_a is the activation energy (approximated as the activation energy of diffusion from Eq. 2 with the
209 same parameters as in the nucleation delay calculation), T_{exp} is the temperature at which the experiment
210 was conducted, ΔH is enthalpy of fusion (extracted from MELTS), and T_L is the liquidus temperature.
211 Growth rates calculated using Equation (4) yield results on the same order of magnitude as the growth
212 rates derived from experimental results with nucleation delay taken into account. Moreover, the growth
213 rate at 50 MPa most closely matches the growth rate derived from the longest experiment suggesting that
214 after dissipation of the initial steep chemical potential gradients, the growth rate trends to equilibrium
215 over time. In the case of growth of plagioclase, at 50 MPa the equilibrium growth rate is achieved after
216 ~95 h (120 h of the experimental duration minus ~25 h for the nucleation delay).

217 **Discussion**

218 **Using nucleation delay to estimate the ascent rate in microlite-free and microlite-bearing rhyolites**
 219 Textures of volcanic products are strongly linked to the history of magma ascent (e.g., Noguchi et al.
 220 2006, Polacci et al. 2006, Bain et al. 2021) . On eruptive timescales, degassing of volatile-rich magma
 221 can induce feldspar microlite formation, although this growth can be hindered if the ascent is too fast
 222 (c.f., Hammer and Rutherford 2002). For volcanic hazard reduction, estimating the ascent rates during
 223 an eruptions is crucial, as it can determine the style of eruption (e.g., Dingwell, D.B. 1996, Castro and
 224 Gardner 2008, Mujin and Nakamura 2014). Typical ascent rates derived for rhyolitic eruptions are in the
 225 range of 0.00005 to 5 MPa/s, corresponding to ascent velocities of 0.0001 to 200 m/s (Rutherford 2008).
 226 Figure 5 presents the comparison between depressurization equivalent ascent rates of 0.0005 MPa/s,
 227 0.001 MPa/s, 0.005 MPa/s and 0.01 MPa/s, and the nucleation delay of feldspar in water saturated
 228 rhyolite. The starting pressure for the ascent paths, simulating the initial storage conditions, is 120 MPa.
 229 For the composition considered at 825 °C these conditions are above the liquidus. The heterogeneous
 230 nature of nucleation requires using the correction factor (τ_{het}):

$$231 \quad \tau_{het} = \tau\varphi^{\frac{1}{3}} \quad (5)$$

232 where

$$233 \quad \varphi = \frac{1}{2} - \frac{3}{4}\cos\theta + \frac{1}{4}\cos^3\theta \quad (6)$$

234 here θ is the interior contact angle between the surface upon which nucleation occurs and the
 235 nucleating phase (Gutzow and Schmelzer 1995; Fokin et al. 2006). The angle used here (25°) is the
 236 median wetting angle for feldspar crystals in rhyolite (Holness 2006). In the case of homogeneous
 237 nucleation, none of the ascent paths cross the nucleation delay curve except for the path with an ascent
 238 rate of 0.0005 MPa/s. The presence of microlites in a water-saturated rhyolite under conditions
 239 considered in this study would mean that if the ascent started at the superliquidus storage conditions of
 240 120 MPa and 825 °C, then the minimum ascent rate would be between 0.0005 and 0.001 MPa/s. If no
 241 feldspar microlites were produced, the maximum ascent rate would be 0.001 MPa/s. Presence of pre-

242 existing crystals in the melt subjected to decompression may influence the nucleation kinetics during
243 ascent by decreasing the interfacial free energy between the parent phase (melt) and new nuclei,
244 consequently decreasing the nucleation delays (c.f., Martel 2012, Rusiecka et al. 2020). In case of
245 heterogeneous nucleation delay (Fig. 5) the ascent path crosses the nucleation delay curve at 0.005
246 MPa/s and not at 0.01 MPa/s, suggesting that in the absence of microlites or overgrowths on pre-
247 existing crystals the maximum ascent rate was 0.01 MPa/s. If microlites or overgrowths are present the
248 minimum ascent rate declines, for this case, to 0.005 MPa/s. This simple model makes it possible to
249 estimate the maximum ascent rate of the crystal free rhyolite, and the minimum ascent rate of
250 microlite-bearing rhyolite.

251 An example of how presence or absence of microlites can be used to estimate the ascent rate is the
252 2008 eruption of Chaitén (Northern Patagonia, Chile). There has been discussion as to the reason of the
253 lack of microlites in the products of this eruption (Andrews and Befus 2020, Huggins et al. 2021).
254 Andrews and Befus (2020), who in presenting their Supersaturation Nucleation and Growth of
255 Plagioclase (SNGPlag) numerical model, suggested that the reason their model was unable to
256 reproduce the crystal poor textures of the 2008 Chaiten eruption was the fact that they did not include
257 nucleation delay in their calculations. Castro and Dingwell (2009) performed decompression
258 experiments to determine the minimum decompression rate which resulted in the textures observed in
259 the products of the 2008 eruption. They observed that decompression at 10 MPa/h and 20 MPa/h
260 (~ 0.005 MPa/s) produced an overgrowth on pre-existing plagioclase crystals but the rim did not form
261 as a result of decompression at 40 MPa/h (~ 0.01 MPa/s). This result agrees well with the model
262 presented here, where the nucleation delay curve for heterogeneous nucleation crosses the ascent path
263 at 0.005 MPa/s suggesting formation of plagioclase at this ascent rate, but not at 0.01 MPa/s. This
264 indicates that the ascent of 0.01 MPa/s is too fast for the plagioclase to form.

265 **Modelling nucleation delay in Pressure-Temperature (PT) space**

266 Because just one isothermal decompression scenario is used, the nucleation delay model described above
267 may have limited utility in calculating nucleation delays of feldspar in rhyolitic compositions across a
268 wide range of conditions. However, the successful comparison of the nucleation delay model to the
269 decompression results of this study, and the isobaric tests of Rusiecka et al. (2020), allow for the
270 construction of a model of nucleation delay change with pressure and temperature. The nucleation delay
271 at different pressures and temperatures was calculated using a 40 by 40 grid of P and T with a pressure
272 step of 25 bars (2.5 MPa). This corresponds to the uncertainty of the pressure in the experiments and the
273 temperature step of 5 °C. Figure 6 shows the results of modelling of nucleation delay from 100 MPa to
274 10 MPa and for temperatures from 800 °C to 650 °C. The highest nucleation delays are observed close
275 to the liquidus, at high pressure and temperature, and at the highest degree of undercooling, at low
276 pressure and temperature. At low pressure and temperature there is a high degree of undercooling and
277 the nucleation delay is primarily controlled by the activation energy. Therefore the barrier preventing
278 nucleation is mainly kinetic. At high pressure and temperature, close to the liquidus, the nucleation is
279 mostly hindered by the thermodynamic barrier, i.e., the interplay between the surface and bulk free
280 energy.

281 Our findings may expand the use of nucleation delay and crystallization of microlites into more
282 complicated magma ascent scenarios, for example: one in which the temperature is not constant during
283 decompression. Thermodynamic modelling of magma degassing without crystallization indicates that
284 both heating and cooling is possible (c.f., Mastin and Ghiorso 2001, Glazner 2019). Not only can the
285 degassing result in a change in temperature during decompression, but also the latent heat released during
286 crystallization of phases not considered in the nucleation delay model may increase the temperature of
287 the magma (c.f., Couch et al. 2001, Blundy et al. 2006). Such phases include those that may nucleate
288 before plagioclase, such as Fe-Ti oxides, pyroxene, and amphibole. The results from our modelling are
289 available in Supplementary material.

290 **Re-evaluating feldspar growth rates**

291 Growth rates when the nucleation delay is considered are twice to three times faster than when the growth
292 is considered throughout the whole experiment (Fig. 3). The same can be observed when comparing
293 growth rates obtained at 50 MPa and for different durations, with the largest difference being visible
294 when comparing growth rates for the shortest experiments (Fig. 4). Growth rates calculated with and
295 without consideration of the nucleation delay have been fitted with an exponential trend as described in
296 Figure 4. The trend line for growth rates calculated without nucleation delay has the same, but the
297 intercept two times lower.

298 It is especially important to account for the nucleation delay when the timescales being considered are
299 on the same order of magnitude as the delay. This is the case for our experiments, as well as when
300 considering the formation of microlites in an ascending magma where timescales are short i.e., velocities
301 and decompression rates are fast. Experimental growth rates adjusted for delayed nucleation and applied
302 to volcanic systems, can thus help to constrain the minimum ascent rate based on the presence and size
303 of the microlites. When calculating growth rates we need to, however, not only consider the delayed
304 nucleation, but also the deviation from the equilibrium growth rate.

305 Growth rates obtained in this study fall within the range of growth rates determined in previous studies
306 on decompression induced crystallization in felsic melts (e.g., Hammer and Rutherford 2002, Couch
307 2003, Martel 2012, Mollard et al. 2012, Riker et al. 2015). Our growth rates are compared to the results
308 of other experimental studies on silicic melts in which the single step decompression (SSD) method was
309 employed in (Fig. 3).

310 Mollard et al. (2012) obtained the growth rates one order of magnitude higher than in our study at similar
311 ΔT_{eff} whereas the growth rates in Hammer and Rutherford (2002) and Couch (2003) are of the same order
312 of magnitude as obtained here. One reason for this discrepancy may be the effect of composition. This
313 study and the studies of Hammer and Rutherford (2002) and Couch (2003) used compositions with SiO_2
314 contents of less than 75 wt. % and containing K_2O . Mollard et al. (2012) used a high silica composition
315 ($\text{SiO}_2 = 78.7$ wt. %) and with no K_2O . The growth rates of Hammer and Rutherford (2002) and Couch

316 (2003) are, however, consistently lower than those obtained here (with exception of two short duration
317 experiments of Couch 2003). However, the results of Hammer and Rutherford (2002) and Couch (2003)
318 fall within the uncertainty of our growth rates calculated without taking the nucleation delay into
319 consideration. This further strengthens the importance of accurately estimating nucleation delays when
320 trying to obtain and compare growth rates.

321 **Conclusions**

322 Theoretical (Classical Nucleation Theory) and experimental nucleation delays for decompression-
323 induced crystallization of feldspar in rhyolitic melt agree within a factor of two. This finding opens up
324 the possibility of quantitatively predicting feldspar nucleation during decompression in water-saturated
325 rhyolite. This, thus, provides an opportunity to better understand the timescales of ascent, decompression
326 and nucleation. As part of this, nucleation delay is particularly important to consider for accurate growth
327 rate estimation, particularly under disequilibrium conditions resulting in rapid growth. Nucleation delay
328 can be used to calculate the maximum ascent rate of microlite-free rhyolites and the minimum ascent rate
329 of microlite-bearing rhyolites. We thus extend the simple model for cooling-induced and decompression-
330 induced crystallization, allowing the nucleation delay of feldspar in rhyolites to be estimated under a range
331 of eruptive conditions in shallow volcanic systems.

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340 **References:**

- 341 1. Andrews, B. J., Befus, K. S., 2020. Supersaturation nucleation and growth of plagioclase: a
342 numerical model of decompression induced crystallization. *Contributions to Mineralogy and*
343 *Petrology*, 175, 1-20. <https://doi.org/10.1007/s00410-020-1660-9>
- 344 2. Arzilli, F., Carroll, M. R. (2013). Crystallisation kinetics of alkali feldspars in cooling and
345 decompression-induced crystallisation experiments in trachytic melt. *Contributions to*
346 *Mineralogy and Petrology*, 166, 1011–1027. doi: 10.1007/s00410-013-0906-1
- 347 3. Arzilli, F., Stabile, P., Fabbrizio, A., Landi, P., Scaillet, B., Paris, E., Carroll, M. R. (2020).
348 Crystallisation kinetics of alkali feldspar in peralkaline rhyolitic melts: implications for
349 Pantelleria volcano. *Frontiers in Earth Science*, 8, 177. doi: 10.3389/feart.2020.00177
- 350 4. Baker, D. R. (1990). Chemical interdiffusion of dacite and rhyolite: anhydrous measurements at
351 1 atm and 10 kbar, application of transition state theory, and diffusion in zoned magma
352 chambers. *Contributions to mineralogy and petrology*, 104, 407-
353 423. <https://doi.org/10.1007/BF01575619>
- 354 5. Baker, D. R. (1992). Estimation of diffusion coefficients during interdiffusion of geologic
355 melts: application of transition state theory. *Chemical Geology*, 98, 11-21.
356 [https://doi.org/10.1016/0009-2541\(92\)90089-N](https://doi.org/10.1016/0009-2541(92)90089-N)
- 357 6. Baker, D. R., Rusiecka, M. K., Bilodeau, M., Kwon, S. Y. (2020). Nucleation delay in the
358 anorthite-diopside binary system: Models and experiments. *Journal of Non-Crystalline*
359 *Solids*, 546, 120255. <https://doi.org/10.1016/j.jnoncrysol.2020.120255>
- 360 7. Bain, A.A., Kendrick, J.E., Lamur, A., Lavallée, Y., Calder, E.S., Cortés, J.A., Cortés, G.P.,
361 Gómez Martínez, D., Torres, R.A. (2021). Micro-textural control on magma rheology and
362 vulcanian explosion cyclicality. *Frontiers in Earth Science*, 8:611320.
363 <https://doi.org/10.3389/feart.2020.611320>

- 364 8. Becker, R. and Döring, W. (1935). Kinetische Behandlung der Keimbildung in übersättigten
365 Dämpfen. *Annales of Physics*, 416, 719–752.
- 366 9. Blundy, J. D., Cashman, K., Humphreys, M. C. S. (2006). Magma heating by decompression-
367 driven crystallization beneath andesite volcanoes. *Nature*, 443(7107), 76-80.
368 <https://doi.org/10.1038/nature05100>
- 369 10. Brugger, C. R., Hammer, J. E. (2010). Crystallization kinetics in continuous decompression
370 experiments: implications for interpreting natural magma ascent processes. *Journal of*
371 *Petrology*, 51, 1941–1965.
- 372 11. Cassidy, M., Manga, M., Cashman, K., Bachmann, O. (2018). Controls on explosive□effusive
373 volcanic eruption styles. *Nature Communications*, 9, 2839.
- 374 12. Castro, J.A., Gardner, J.E. (2008). Did magma ascent rate control the explosive-effusive
375 transition at the Inyo volcanic chain, California? *Geology*, 36, 279-282.
376 <https://doi.org/10.1130/G24453A.1>
- 377 13. Castro, J.A., Dingwell, D. B. (2009). Rapid ascent of rhyolitic magma at Chaitén volcano,
378 Chile. *Nature* 461, 780–783. <https://doi.org/10.1038/nature08458>
- 379 14. Couch, S. (2003). Experimental investigation of crystallization kinetics in a haplogranite
380 system. *American Mineralogist*, 88, 1471–1485.
- 381 15. Couch, S., Sparks, R.S.J., Carroll, M.R. (2001). Mineral disequilibrium in lavas explained by
382 convective self-mixing in open magma chambers. *Nature* 411, 1037–1039.
383 <https://doi.org/10.1038/35082540>
- 384 16. Cramer, F. (2018). Scientific colour-maps. Zenodo. <https://doi.org/10.5281/zenodo.1243862>
- 385 17. Cramer, F., Shephard, G. E., Heron, P. J. (2020). The misuse of colour in science
386 communication. *Nature Communications*, 11, 5444. [https://doi.org/10.1038/s41467-020-19160-](https://doi.org/10.1038/s41467-020-19160-7)

- 388 18. deGraffenried, R. L., Larsen, J. F., Graham, N. A., Cashman, K. V. (2019). The influence of
389 phenocrysts on degassing in crystal-bearing magmas with rhyolitic groundmass
390 melts. *Geophysical Research Letters*, 46, 5127– 5136.
- 391 19. De Yoreo, J. J., Vekilov, P. G. (2003). Principles of crystal nucleation and growth. *Reviews in*
392 *mineralogy and geochemistry*, 54, 57-93.
- 393 20. Dingwell, D.B. (1996). Volcanic dilemma: Flow or blow? *Science*, 273, 1054-1055. DOI:
394 10.1126/science.273.5278.1054
- 395 21. D’Oriano, C., Poggianti, E., Bertagnini, A., Cioni, R., Landi, P., Polacci, M., Rosi, M. (2005).
396 Changes in eruptive style during the A.D. 1538 Monte Nuovo eruption (Phlegrean Fields, Italy):
397 the role of syn-eruptive crystallization. *Bulletin of Volcanology*, 67, 601-621.
- 398 22. Eyring, H. (1935). The activated complex in chemical reactions. *The journal of chemical*
399 *physics*, 3, 107. <https://doi.org/10.1063/1.1749604>
- 400 23. Fenn, P.M. (1977) The nucleation and growth of alkali feldspars from hydrous melts, *Canadian*
401 *Mineralogist* 15, 135-161.
- 402 24. Fokin, V. M., Zanutto, E. D., Yuritsyn, N. S., Schmelzer, J. W. (2006). Homogeneous crystal
403 nucleation in silicate glasses: a 40 years perspective. *Journal of Non-Crystalline Solids*,
404 352(26), 2681-2714.
- 405 25. Gardner, J. E., Denis, M. H. (2004). Heterogeneous bubble nucleation on Fe-Ti oxide crystals in
406 high-silica rhyolitic melts. *Geochimica et Cosmochimica Acta*, 68(17), 3587-3597.
407 <https://doi.org/10.1016/j.gca.2004.02.021>
- 408 26. Gibbs, J.W. (1875–1878). On the Equilibrium of Heterogeneous Substances. *Transactions of the*
409 *Connecticut Academy of Arts Sciences*, 108, 343.
- 410 27. Giordano, D., Russell, J. K., Dingwell, D. B. (2008). Viscosity of magmatic liquids: a
411 model. *Earth and Planetary Science Letters*, 271(1-4), 123-134.

- 412 28. Ghiorso, M. S., Sack, R. O. (1995). Chemical Mass Transfer in Magmatic Processes. IV. A
413 Revised and Internally Consistent Thermodynamic Model for the Interpolation and
414 Extrapolation of Liquid-Solid Equilibria in Magmatic Systems at Elevated Temperatures and
415 Pressures. *Contributions to Mineralogy and Petrology*, 119, 197-212.
- 416 29. Ghiorso, M. S., Gualda, G. A. R. (2015). An H₂O–CO₂ mixed fluid saturation model compatible
417 with rhyolite-MELTS. *Contributions to Mineralogy and Petrology*, 169(6), 1-30.
418 <https://doi.org/10.1007/s00410-015-1141-8>
- 419 30. Glazner, A. F. (2019). The ascent of water-rich magma and decompression heating: A
420 thermodynamic analysis. *American Mineralogist: Journal of Earth and Planetary
421 Materials*, 104(6), 890-896. <https://doi.org/10.2138/am-2019-6925>
- 422 31. González-García, D., Behrens, H., Petrelli, M., Vetere, F., Morgavi, D., Zhang, C., Perugini, D.
423 (2017). Water-enhanced interdiffusion of major elements between natural shoshonite and high-
424 K rhyolite melts. *Chemical Geology*, 466, 86-101.
425 <https://doi.org/10.1016/j.chemgeo.2017.05.023>
- 426 32. Gualda G.A.R., Ghiorso M.S., Lemons R.V., Carley T.L. (2012). Rhyolite-MELTS: A modified
427 calibration of MELTS optimized for silica-rich, fluid-bearing magmatic systems. *Journal of
428 Petrology*, 53, 875-890.
- 429 33. Gutzov, I., Schmelzer, J. W. P. (1995). The vitreous state: thermodynamics, structure, rheology,
430 and crystallization. Springer.
- 431 34. Hammer, J. E. (2004). Crystal nucleation in hydrous rhyolite: Experimental data applied to
432 classical theory. *American Mineralogist*, 89, 1673–1679.
- 433 35. Hammer, J. E., Cashman, K. V., Hoblitt, R. P., & Newman, S. (1999). Degassing and microlite
434 crystallization during pre-climactic events of the 1991 eruption of Mt. Pinatubo,
435 Philippines. *Bulletin of Volcanology*, 60(5), 355-380. <https://doi.org/10.1007/s004450050238>

- 436 36. Hammer, J.E. and Rutherford, M. (2002). An experimental study of the kinetics of
437 decompression-induced crystallization in silicic melt. *Journal of Geophysical Research*, 107, 1-
438 23.
- 439 37. Holness, M. B. (2006). Melt–solid dihedral angles of common minerals in natural rocks. *Journal*
440 *of Petrology*, 47(4), 791-800. <https://doi.org/10.1093/petrology/egi094>
- 441 38. Huggins, E.G., Ruprecht, R., Ghiorso, M.S. (2021). Using Chemical Affinities to Understand
442 Disequilibrium Textures of Plagioclase Preserved in Magmatic Systems. *Geophysical Research*
443 *Letters*, 48 (10), e2021GL092884. <https://doi.org/10.1029/2021GL092884>
- 444 39. Hurwitz, S., Navon, O. (1994). Bubble nucleation in rhyolitic melts: Experiments at high
445 pressure, temperature, and water content. *Earth and Planetary Science Letters*, 122(3-4), 267-
446 280. [https://doi.org/10.1016/0012-821X\(94\)90001-9](https://doi.org/10.1016/0012-821X(94)90001-9)
- 447 40. Kashchiev, D. (1969). Solution of the non-steady problem in nucleation kinetics. *Surface*
448 *science*, 14, 209-220.
- 449 41. Kirkpatrick, R. J. (1981). Kinetics of Crystallization of Igneous Rocks in: Lasaga, A.C.,
450 Kirkpatrick, R. J. (Eds.), *Kinetics of Geochemical, Reviews in Mineralogy*, 8, 321-397.
- 451 42. Lindoo, A. N., Larsen, J. F., Cashman, K. V., Dunn, A. L., Neill, O. K. (2016). An experimental
452 study of permeability development as a function of crystal-free melt viscosity. *Earth and*
453 *Planetary Science Letters*, 435, 45– 5.
- 454 43. Mader, H. M., Llewelin, E. W., Mueller, S. P. (2013). The rheology of two-phase magmas: A
455 review and analysis. *Journal of Volcanology and Geothermal Research* 257, 135–158.
- 456 44. Maneta, V., Baker, D. R. (2014). Exploring the effect of lithium on pegmatitic textures: An
457 experimental study. *American Mineralogist*, 99(7), 1383-1403.
458 <https://doi.org/10.2138/am.2014.4556>

- 459 45. Mastin, L. G. (2002). Insights into volcanic conduit flow from an open-source numerical
460 model. *Geochemistry, Geophysics, Geosystems*, 3(7), 1-18.
461 <https://doi.org/10.1029/2001GC000192>
- 462 46. Martel, C. (2012). Eruption dynamics inferred from microlite crystallization experiments:
463 application to Plinian and dome-forming eruptions of Mt. Pelée (Martinique, Lesser
464 Antilles). *Journal of Petrology*, 53(4), 699-725. <https://doi.org/10.1093/petrology/egr076>
- 465 47. Mollard, E., Martel, C., Bourdier, J-L. (2012). Decompression-induced Crystallization in
466 Hydrated Silica-rich Melts: Empirical Models of Experimental Plagioclase Nucleation and
467 Growth Kinetics, *Journal of Petrology*, 53, 8, 1743–
468 1766, <https://doi.org/10.1093/petrology/egs031>
- 469 48. Mollard, E., Martel, C., Le Trong, E., Rogerie, G. (2020). Theoretical models of
470 decompression-induced plagioclase nucleation and growth in hydrated silica-rich
471 melts. *Frontiers in Earth Science*, 8, 203.
- 472 49. Moschini, P., Mollo, S., Gaeta, M., Fanara, S., Nazzari, M., Petrone, C. M., Scarlato, P. (2021).
473 Parameterization of clinopyroxene growth kinetics via crystal size distribution (CSD) analysis:
474 Insights into the temporal scales of magma dynamics at Mt. Etna volcano. *Lithos*, 396, 106225.
475 <https://doi.org/10.1016/j.lithos.2021.106225>
- 476 50. Mujin, M., Nakamura, M. (2014). A nanolite record of eruption style transition. *Geology*, 42,
477 611-614. <https://doi.org/10.1130/G35553.1>
- 478 51. Mungall, J. E. (2002). Empirical models relating viscosity and tracer diffusion in magmatic
479 silicate melts. *Geochimica et Cosmochimica Acta*, 66(1), 125-143.
480 [https://doi.org/10.1016/S0016-7037\(01\)00736-0](https://doi.org/10.1016/S0016-7037(01)00736-0)
- 481 52. Noguchi, S., Toramaru, A., Shimano, T. (2006). Crystallization of microlites and degassing
482 during magma ascent: Constraints on the fluid mechanical behaviour of magma during the

- 483 Tenjo eruption on Koze island, Japan. *Bulletin of Volcanology* 68, 432-449.
484 <https://doi.org/10.1007/s00445-005-0019-4>
- 485 53. Pleše, P., Higgins, M. D., Mancini, L., Lanzafame, G., Brun, F., Fife, J. L., Casselman, J.,
486 Baker, D. R. (2018). Dynamic observations of vesiculation reveal the role of silicate crystals in
487 bubble nucleation and growth in andesitic magmas. *Lithos*, 296, 532-546.
488 <https://doi.org/10.1016/j.lithos.2017.11.024>
- 489 54. Polacci, M., Baker, D.R., Mancini, L., Tromba, G., Zanini, F. (2006). Three dimensional
490 investigation of volcanic textures by X-ray microtomography and implications for conduit
491 processes. *Geophysical Research Letters* 33: L13312. <https://doi.org/10.1029/2006GL026241>
- 492 55. Polacci, M., Arzilli, F., La Spina, G., Le Gall, N., Cai, B., Hartley, M. E. Di Genova, D., Vo, N.
493 T., Nonni, S., Atwood, R. C., Llewellyn, E. W., Lee, P. D., Burton, M. R. (2018). Crystallisation
494 in basaltic magmas revealed via in situ 4D synchrotron X-ray microtomography. *Scientific*
495 *Reports* 8, 8377.
- 496 56. Popa, R.G., Bachmann, O., Huber, C. (2021). Explosive or effusive style of volcanic eruption
497 determined by magma storage conditions. *Nature Geoscience*, 14, 781–786.
498 <https://doi.org/10.1038/s41561-021-00827-9>
- 499 57. Riker, J.M., Cashman, K.V., Rust, A.C., Blundy, J.D. (2015). Experimental constraints on
500 Plagioclase crystallization during H₂O- and H₂O-CO₂- saturated magma decompression. *Journal*
501 *of Petrology* 56, 1967-1998.
- 502 58. Robie, R. A., Bethke P. M. (1962). Molar volumes and densities of minerals. In: Trace elements
503 investigations, USGS Publication Warehouse, 39 pp. <https://doi.org/10.3133/70159012>
- 504 59. Rusiecka, M. K., Bilodeau, M., Baker, D. R. (2020). Quantification of nucleation delay in
505 magmatic systems: experimental and theoretical approach. *Contributions to Mineralogy and*
506 *Petrology*, 175, 47. DOI: <https://doi.org/10.1007/s00410-020-01682-4>

- 507 60. Rusiecka, M.K., Baker, D.R. (2021). Growth and textural evolution during crystallization of
508 quartz and feldspar in hydrous, rhyolitic melt. *Contributions to Mineralogy and*
509 *Petrology*, 176, 48. <https://doi.org/10.1007/s00410-021-01809-1>
- 510 61. Rutherford, M. J. (2008). Magma ascent rates. *Reviews in Mineralogy and*
511 *Geochemistry*, 69(1), 241-271. <https://doi.org/10.2138/rmg.2008.69.7>
- 512 62. Schmelzer, J. W. P. (2008). Crystal nucleation and growth in glass-forming melts: Experiment
513 and theory. *Journal of Non-Crystalline Solids*, 354, 269-278.
- 514 63. Shannon, R. D. (1976). Revised effective ionic radii and systematic studies of interatomic
515 distances in halides and chalcogenides. *Acta Crystallographica*, A32, 751-767.
516 <https://doi.org/10.1107/S0567739476001551>
- 517 64. Sharp, T. G., Stevenson, R. J., & Dingwell, D. B. (1996). Microlites and "nanolites" in rhyolitic
518 glass: microstructural and chemical characterization. *Bulletin of volcanology*, 57(8), 631-640.
519 <https://doi.org/10.1007/s004450050116>
- 520 65. Shea, T. (2017). Bubble nucleation in magmas: a dominantly heterogeneous process?. *Journal*
521 *of Volcanology and Geothermal Research*, 343, 155-170.
522 <https://doi.org/10.1016/j.jvolgeores.2017.06.025>
- 523 66. Sirbescu, M. C., Schmidt, C., Veksler, I. V., Whittington, A. G., Wilke, M. (2017). Experimental
524 crystallization of undercooled felsic liquids: generation of pegmatitic texture. *Journal of*
525 *Petrology*, 58, 539-568.
- 526 67. Slezov, V. V., Schmelzer, J. W. P. (1999). Kinetics of nucleation–growth processes: the first
527 stages. *Nucleation Theory and Applications*, 6-81.
- 528 68. Swanson, S.E. (1977). Relation of crystal-growth rate to the granitic textures. *American*
529 *Mineralogist*, 62, 966-978.

- 530 69. Toramaru, A. (1991). Model of nucleation and growth of crystals in cooling magmas.
531 Contributions to mineralogy and petrology, 108, 106-
532 117. <https://doi.org/10.1007/BF00307330>
- 533 70. Turnbull, D., Fisher, J. C. (1949). Rate of nucleation in condensed systems. The Journal of
534 chemical physics, 17, 71-73.
- 535 71. Volmer, M., Weber, A.Z. (1926) Nucleus Formation in Supersaturated Systems. Zeitschrift
536 für Physikalische Chemie, 119, 277-301.
- 537 72. Vona, A., Di Piazza, A., Nicotra, E., Romano, C., Viccaro, M., Giordano, G. (2017). The
538 complex rheology of megacryst-rich magmas: The case of the mugearitic “cicirara” lavas of Mt.
539 Etna volcano. Chemical Geology, 458, 48–67.

540 **Figure and Table Captions:**

541 **Figure 1.** BSE (back-scattered electron) images of the experimental run products at a, b) 50 MPa (ΔT_{eff}
542 = -55 °C): glass with skeletal feldspar crystals and magnetite, in b) several melt inclusions can be
543 observed; c) 75 MPa (ΔT_{eff} = -20 °C) – glass with feldspar and magnetite crystals and visible vesicle; d)
544 90 MPa (ΔT_{eff} = -10 °C) well faceted feldspar crystals in glass with magnetite e) low magnification BSE
545 image showing one of the glass chips recovered after the experiment

546 **Figure 2.** Nucleation delay of plagioclase in metaluminous rhyolite, empty symbols – no crystals
547 observed, filled symbols – crystals observed, circles – this study, squares – Mollard et al. 2012, solid
548 black curve – nucleation delay calculated using Eq. 1. dotted and dashed lines – nucleation delay curves
549 calculated using the range of interfacial free energy of 0.10 to 0.25 J/m². Pressure relates to this study,
550 while ΔT_{eff} relates to both this study and the study of Mollard et al. 2012.

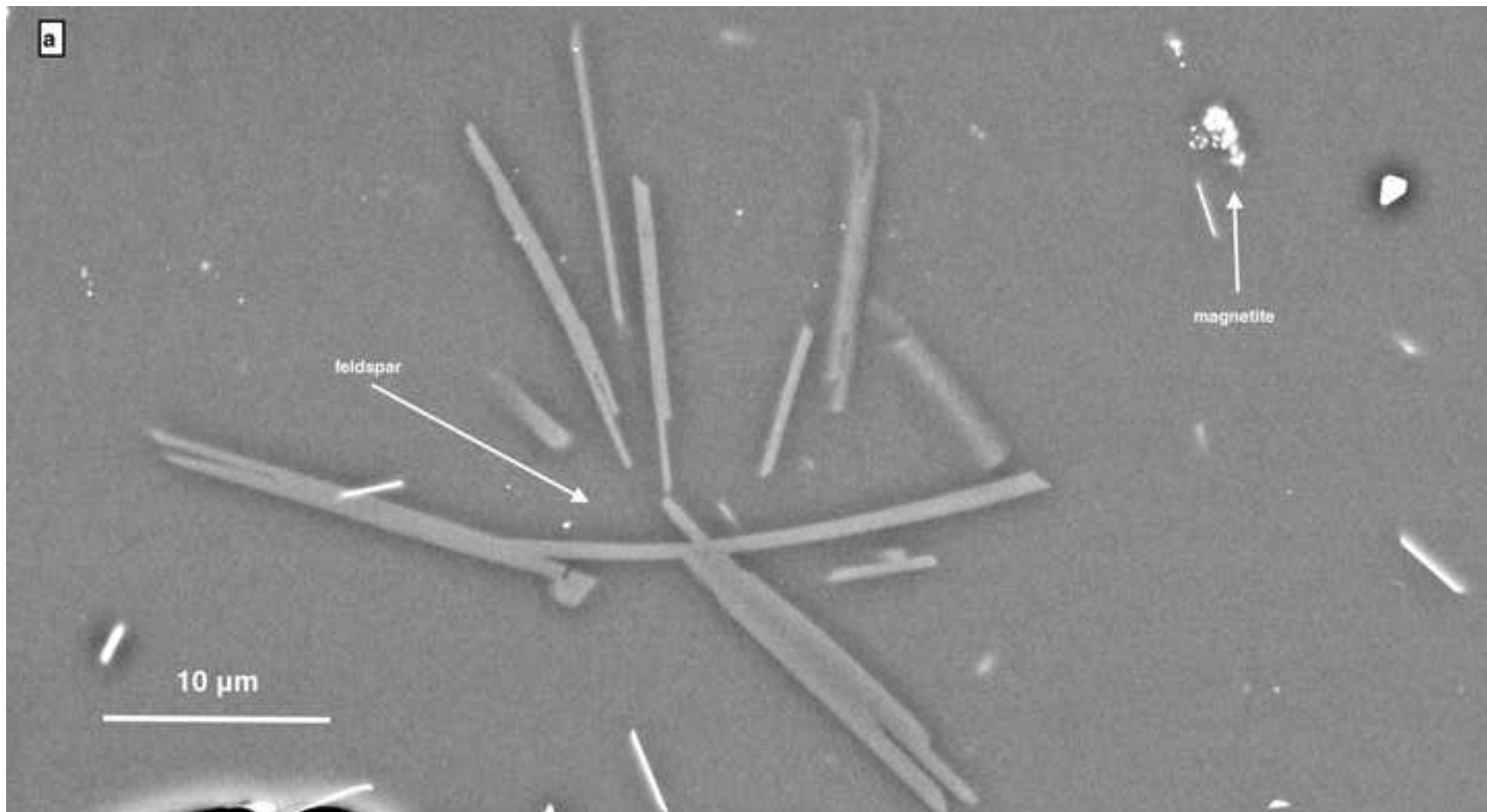
551 **Figure 3.** Growth rates calculated using Eq. 3 at various experimental conditions for average of 10 largest
552 crystals for each samples, circles – this study, squares – Couch 2003, stars – Hammer and Rutherford
553 2002; Pressure relates to this study, while ΔT_{eff} is relevant for all the presented studies.

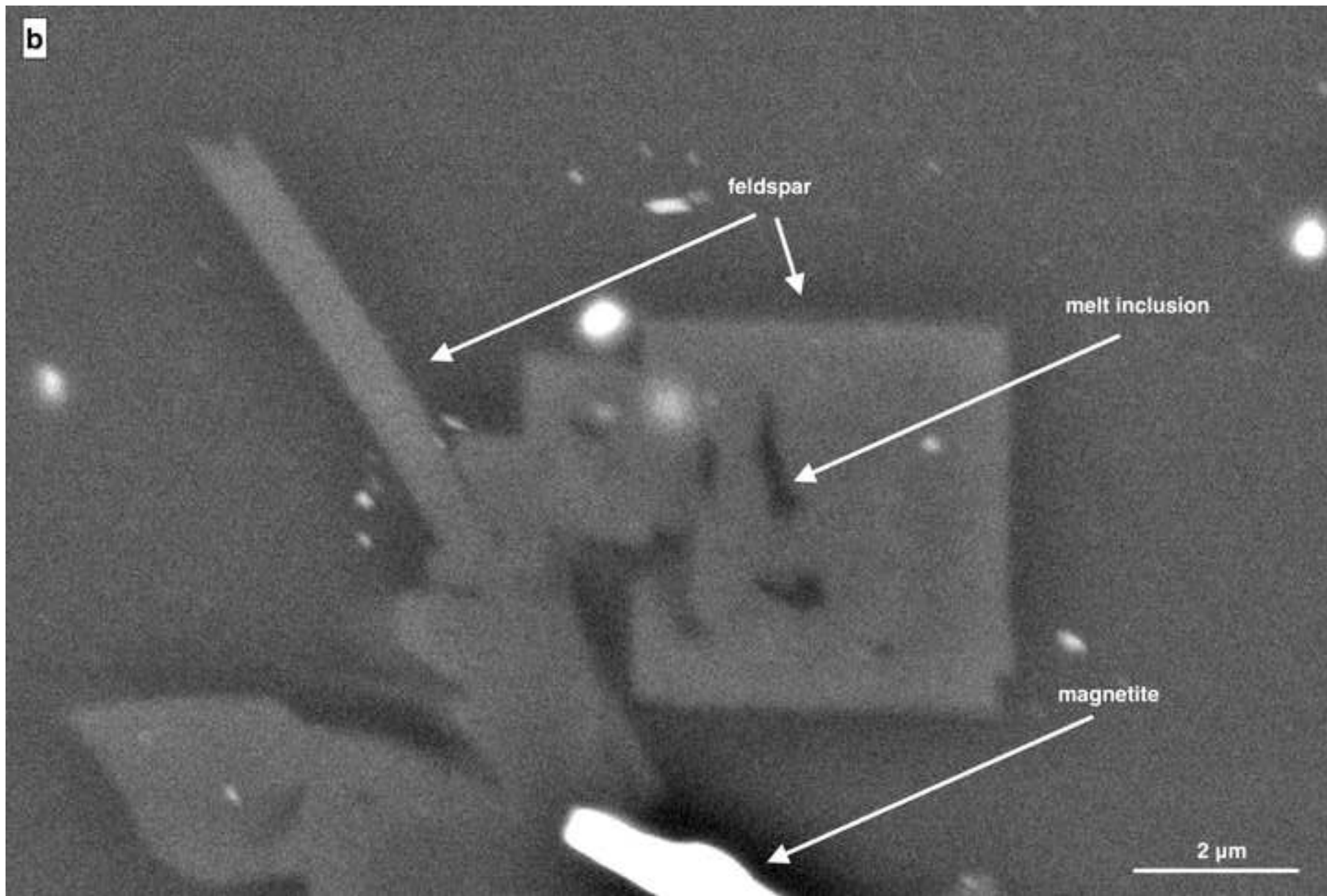
554 **Figure 4.** Growth rates calculated using Eq. 3 at 50 MPa at various experimental durations (48, 72 and
555 120 h) for average of 10 largest crystals for each sample, black symbols – growth rates calculated with
556 taking nucleation delay into consideration, grey symbols – growth rates calculated without taking
557 nucleation delay into consideration, dashed line – exponential trend line fitted to the data.

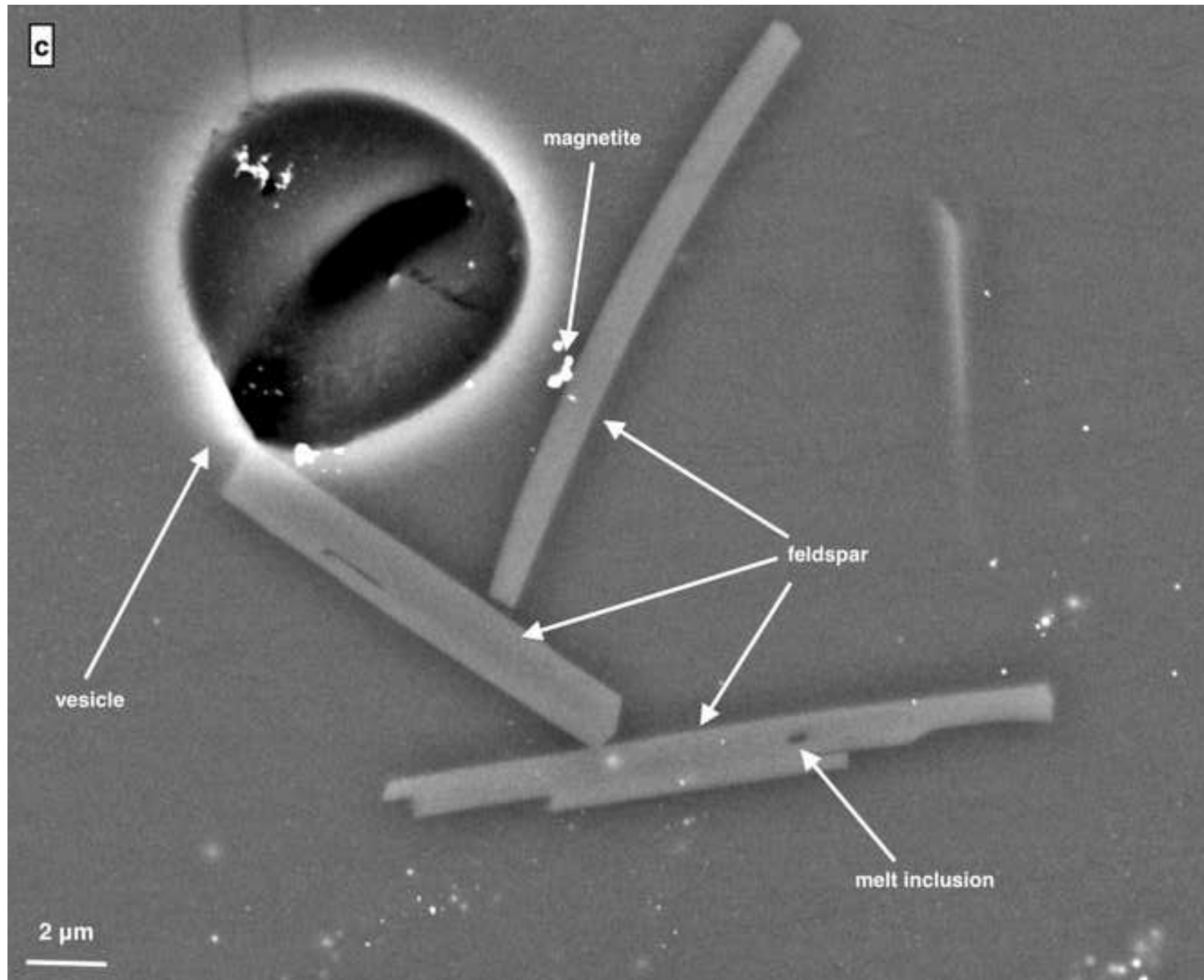
558 **Figure 5.** Nucleation delay curves for homogeneous (solid black curve) and heterogeneous (dotted black
559 curved) nucleation of feldspar calculated using the theoretical model for feldspar nucleation in water-
560 saturated rhyolite, grey dashed lines – ascent paths.

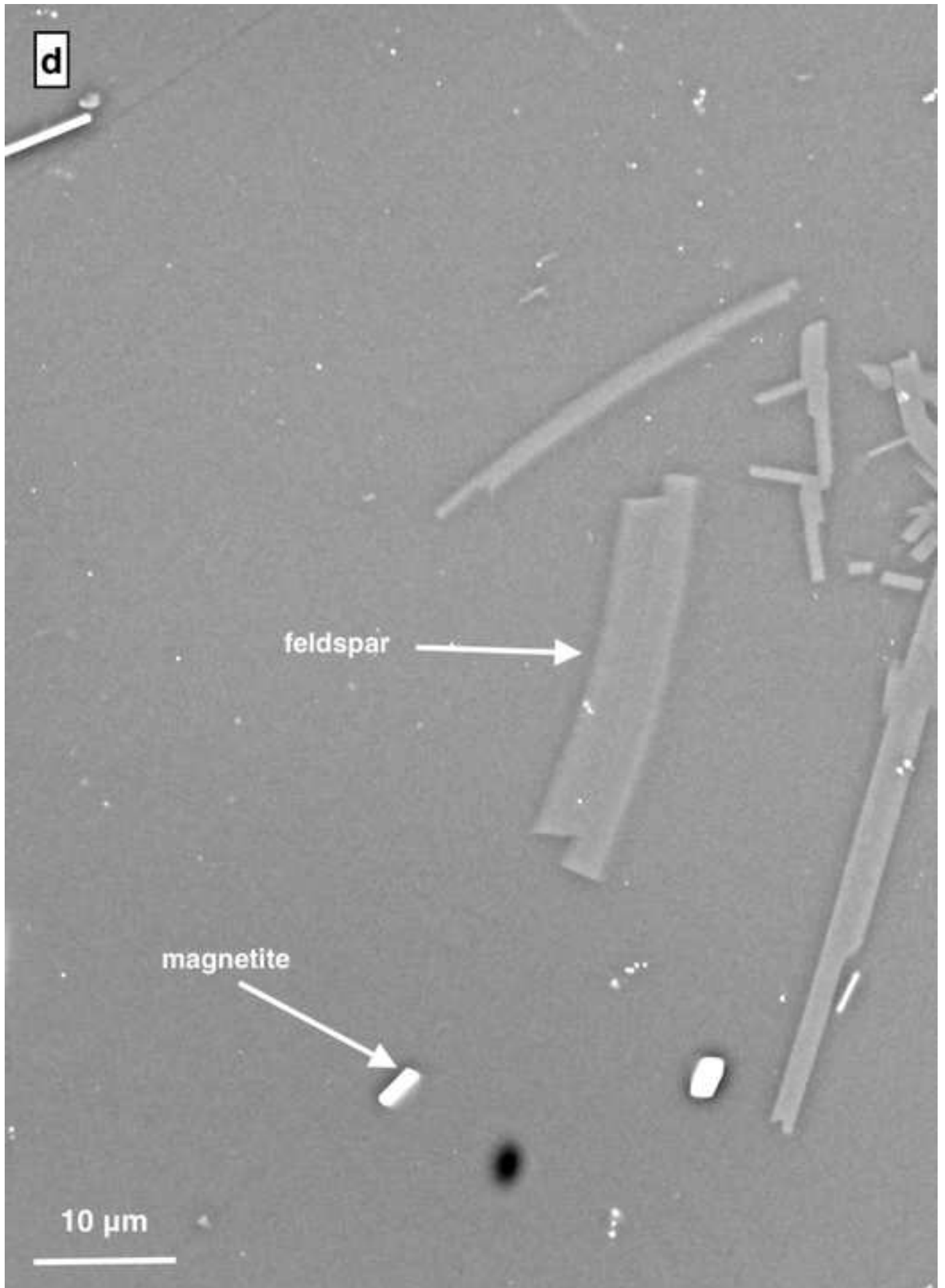
561 **Figure 6.** Nucleation delay calculated in the PT space for water-saturated rhyolite with the model and
562 parameters described in the text of the manuscript. Colormap ‘hawaii’ (Crameri 2018, Crameri et al.
563 2020) was used to ensure accessibility for readers.

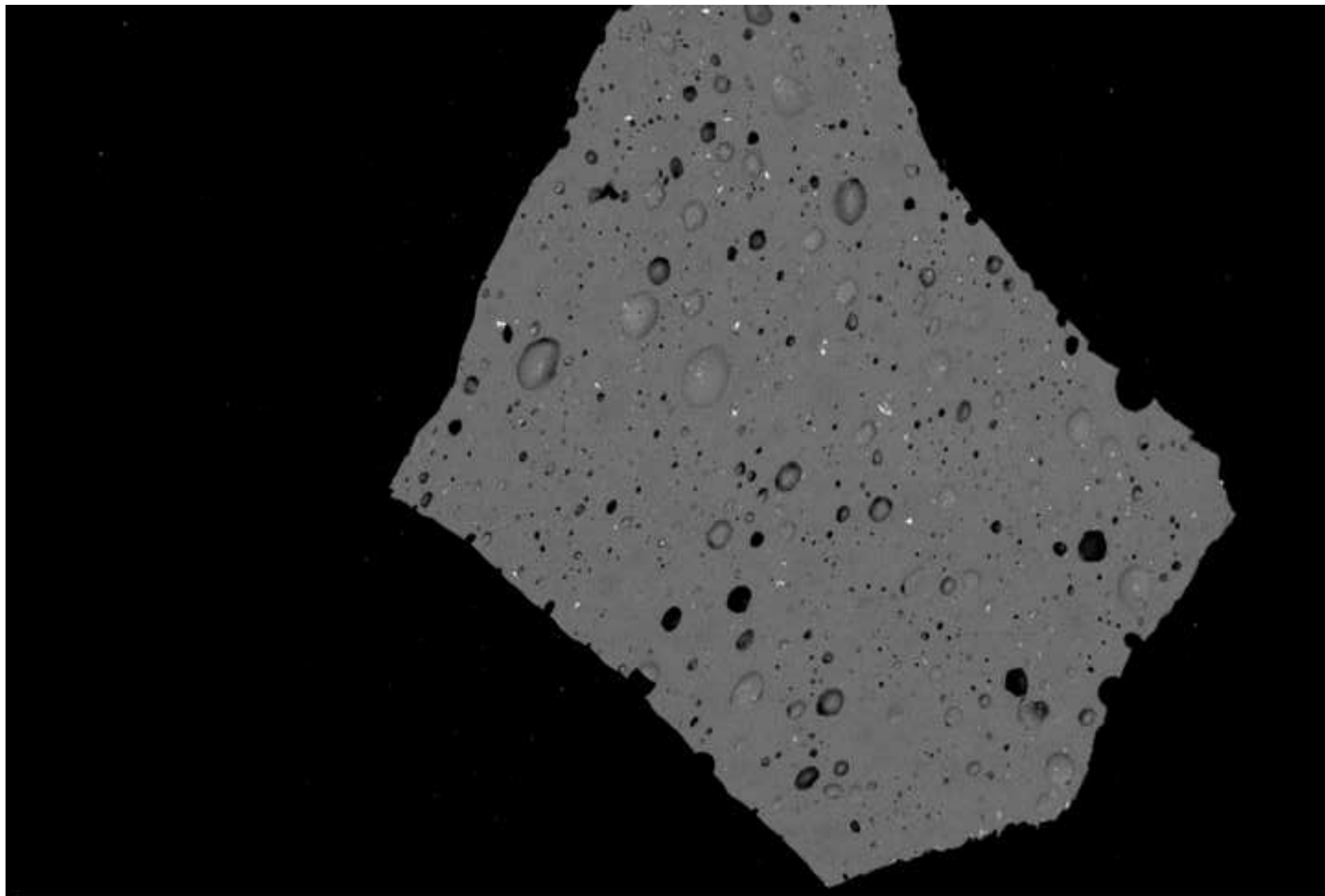
564 **Table 1.** Experimental run table, LCOL – experiments conducted to confirm the location of the liquidus
565 calculated using MELTS, LCOD – decompression single step decompression (SSD) experiments; P_i –
566 initial pressure, P_f – final pressure, ΔT_{eff} – effective undercooling, dP/t – rate of decompression, gl –
567 glass, mgt – magnetite, fds – feldspar.











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ISTO

200 μ m
|-----|

EHT = 15.00 kV
Mag = 33 X

Signal A = BSD4 A
WD = 11.0 mm

Width = 3.465 mm



Figure 2

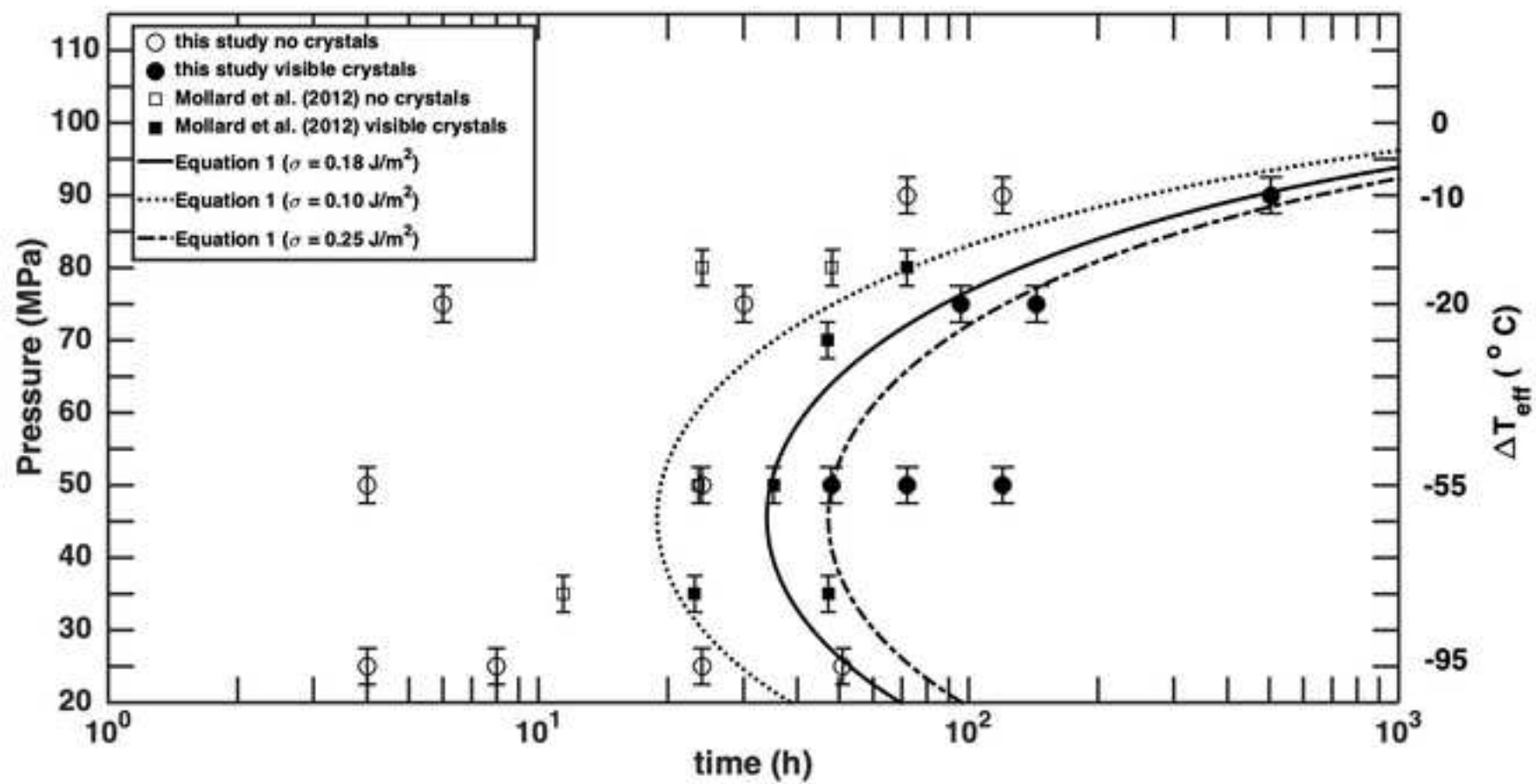
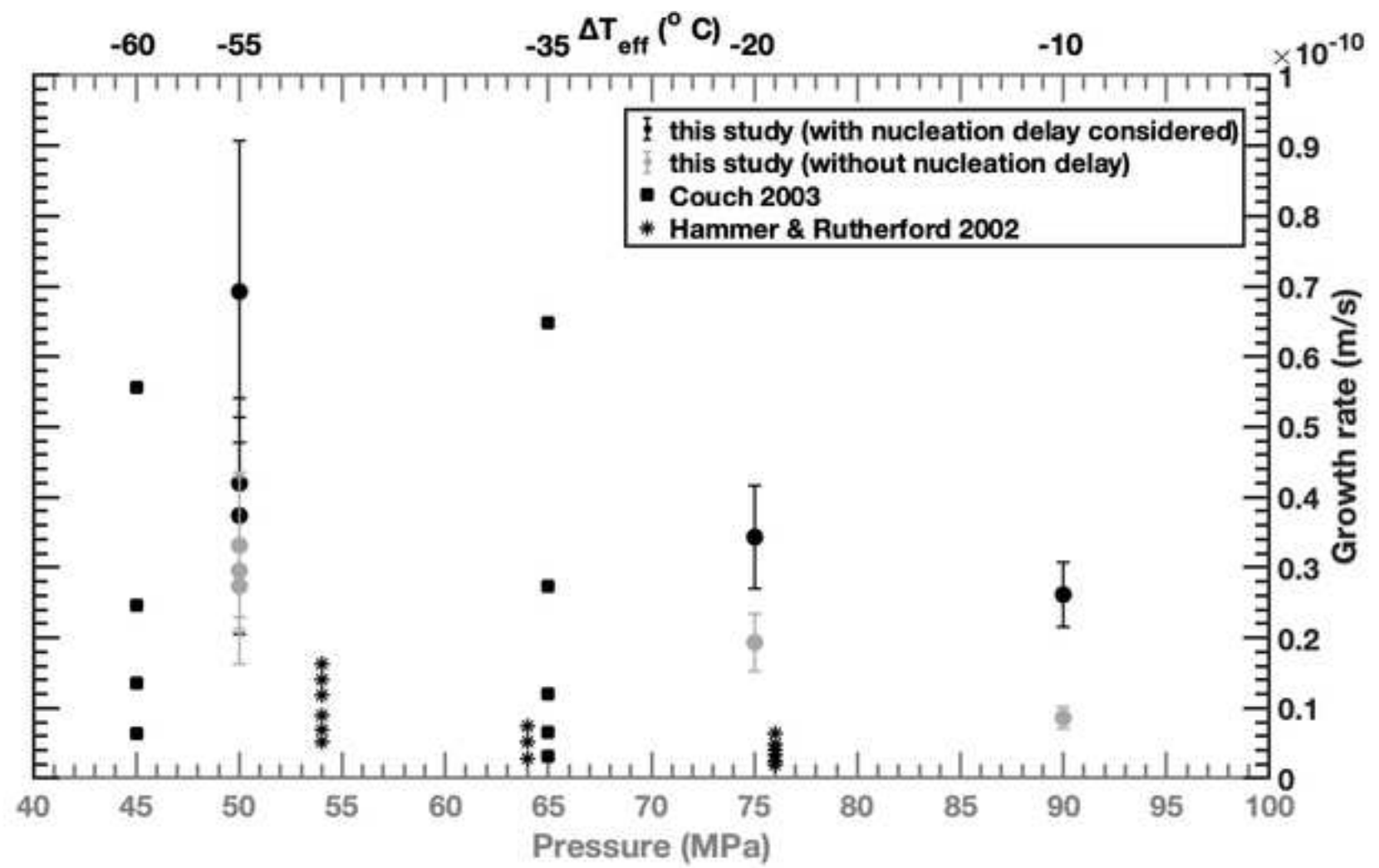
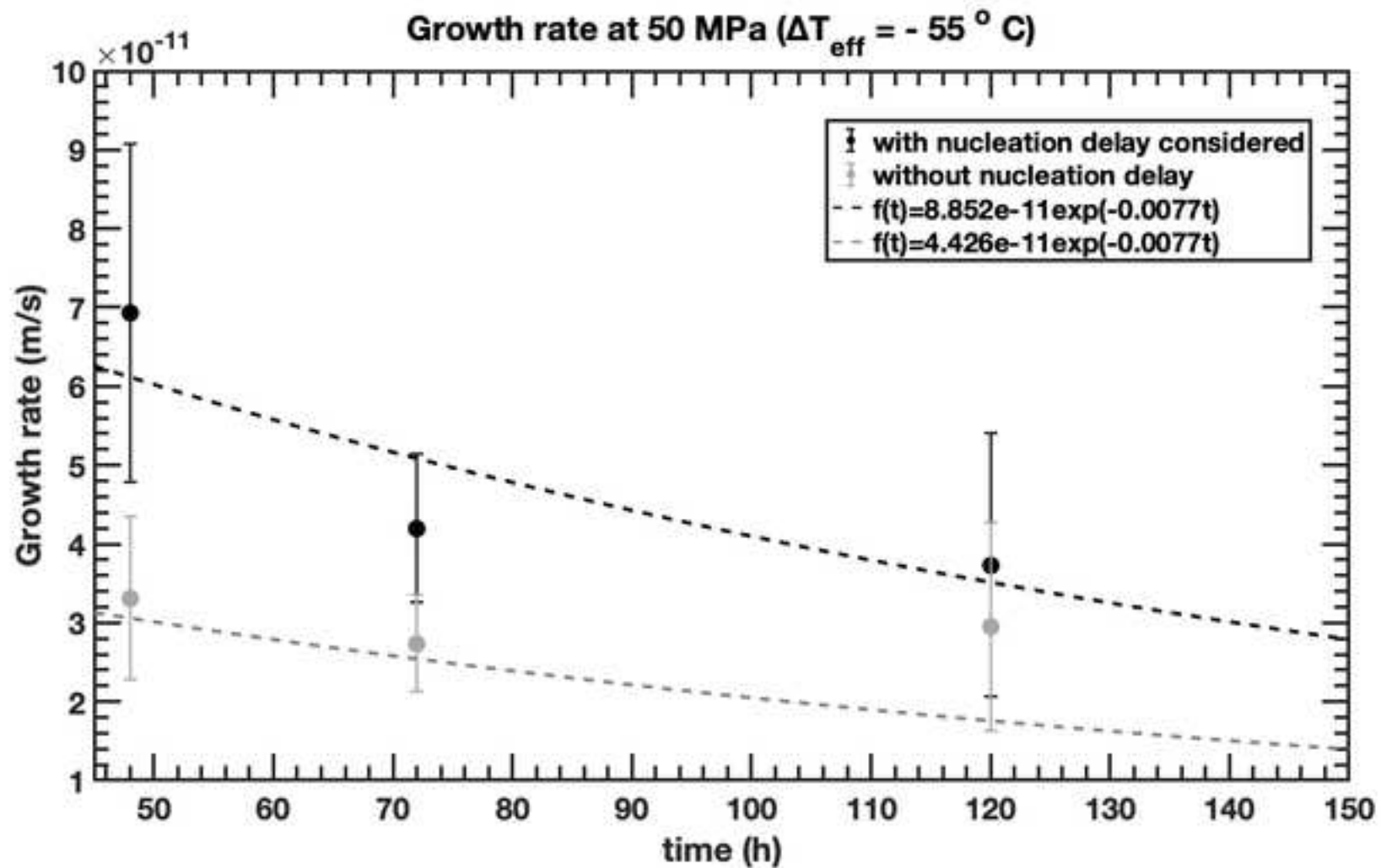


Figure 3





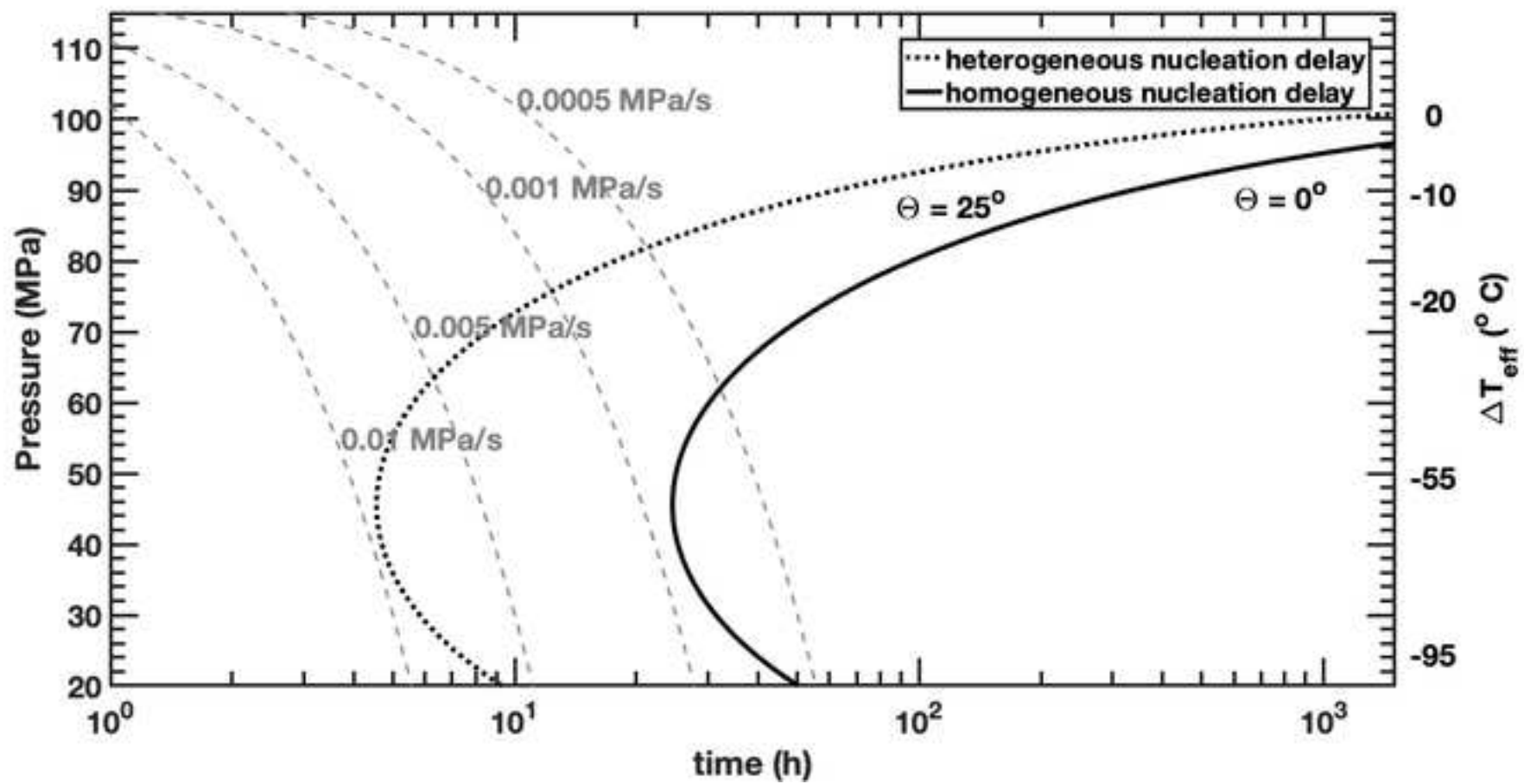
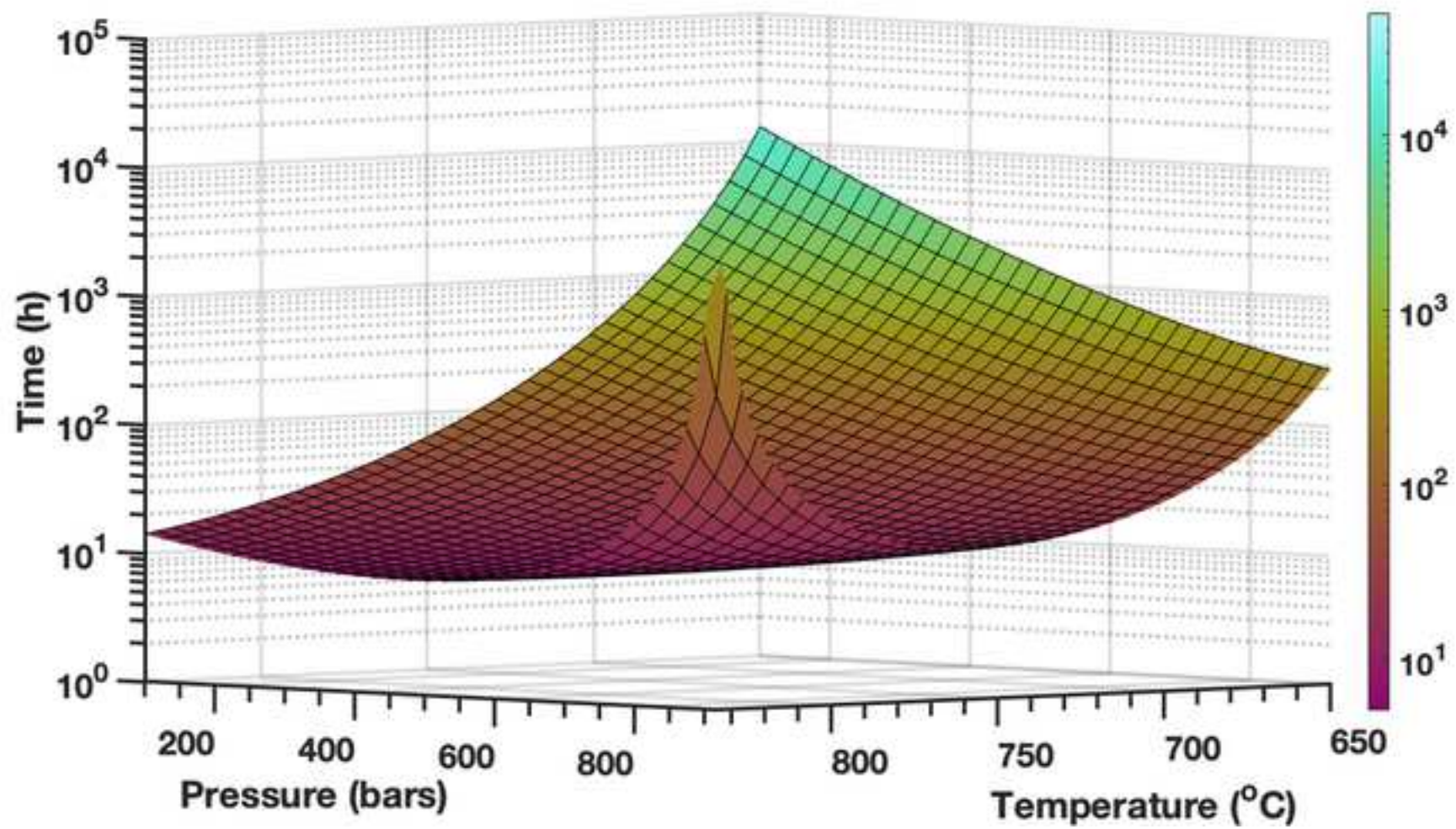


Figure 6



Sheet1

sample	T (°C)	P _i (MPa)	P _f (MPa)	dP/t (MPa/h)	ΔT _{eff} (°C)	time at P _f (h)
LCOD-50	825	200	50	600	-55	0
LCOD-25-4	825	200	25	600	-95	4
LCOD-25-8	825	200	25	600	-95	8
LCOD-25-24	825	200	25	600	-95	24
LCOD-25-51	825	200	25	600	-95	51
LCOD-50-4	825	200	50	600	-55	4
LCOD50-24	825	200	50	600	-55	24
LCOD-50-48	825	200	50	600	-55	48
LCOD-50-72	825	200	50	600	-55	72
LCOD-50-120	825	200	50	600	-55	120
LCOD-75-6	825	200	75	600	-20	6
LCOD-75-30	825	200	75	600	-20	30
LCOD-75-96	825	200	75	600	-20	96
LCOD-75-144	825	200	75	600	-20	144
LCOD-90-72	825	200	90	600	-10	72
LCOD-90-120	825	200	90	600	-10	120
LCOD-90-504	825	200	90	600	-10	504
LCOL200	800	200	200	-	-	168
LCOL200	825	200	200	-	-	168
LCOL200	850	200	200	-	-	168
LCOL200	900	200	200	-	-	168
LCOL150	825	200	200	-	-	168
LCOL50	850	200	200	-	-	168
LCOL50	900	200	200	-	-	168

* phases: gl – glass; mgt – magnetite; fds – feldspar;

Sheet1

nucleation	phases
no	gl+mgt
no	gl+mgt
no	gl+mgt
no	gl+mgt
no	gl+mgt
no	gl+mgt
no	gl+mgt
yes	gl+mgt+fds
yes	gl+mgt+fds
yes	gl+mgt+fds
no	gl+mgt
no	gl+mgt
yes	gl+mgt+fds
yes	gl+mgt+fds
no	gl+mgt
no	gl+mgt
yes	gl+mgt+fds
-	gl+mgt+fds
-	gl+mgt
-	gl+mgt
-	gl+mgt
-	gl+mgt
-	gl+mgt+fds
-	gl+mgt