1 Basalt, Unveiling Fluid-filled Fractures, Inducing Sediment Intra-void Transport, Ephemerally:

examples from Katla 1918

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7 Abstract

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8 This article documents textures within Katla 1918 pyroclasts, where particle-filled fractures and

9 bubbles have been observed. These features are analogous to tuffisite veins; particle-filled fractures10 which represent the preserved remains of transient degassing pathways in shallow conduits.

11 Such fractures have long been considered restricted to high viscosity silicic melts. However, through 12 BSE images and compositional maps, we have identified similar tuffisite-like features in crystal-poor basalt pyroclasts from the 1918 A.D. subglacial eruption of Katla, Iceland (K1918). Clast textures record 13 14 transient mobility of juvenile/lithic particles, melt droplets and gas through magmatic fractures and 15 connected vesicles. Key evidence includes (1) the presence of variably sintered fine-ash particles 16 within variably healed fractures and vesicles (present in >80% of clasts analysed), (2) compositional 17 maps that reveal the presence of foreign particles within preserved and healed permeable pathways, 18 and (3) lower vesicularities immediately surrounding 'fracture' walls, suggestive of diffusive volatile 19 loss into a permeable network.

The 1918 A.D. eruption of Katla occurred under a thick glacier, however the ice was quickly breached, owing partly to the explosive nature of the eruption. We propose that the formation and preservation of these transient permeable networks have been facilitated by rapid decompression of a relatively volatile-rich magma, in a confined subglacial environment, with combined magmatic and phreatomagmatic fragmentation, followed by rapid quenching by meltwater.

25 Tuffisite veins in rhyolite demonstrate repeated fracture-healing cycles, which drive incremental 26 release of overpressured gas and help to defuse explosive eruptions. Interestingly, the permeable 27 network at Katla failed to defuse the 1918 A.D. eruption, which involved a particularly violent subglacial eruptive phase. It is unclear whether this demonstrates an inability of mafic tuffisite-like 28 29 features to efficiently degas magma (perhaps owing to the especially transient nature of permeable 30 pathways in low viscosity magmas) or an ability to enhance fragmentation by providing infiltration 31 pathways for external water. The latter scenario may explain the rapid melting of the overlying glacier 32 as the large surface area-to-volume ratio of fractured magma would allow rapid heat transfer.

Nevertheless, we document a previously undescribed texture in basaltic magmas. It is intriguing why it has not, to the best of our knowledge, been documented elsewhere. Have these permeable pathways been over-looked in the past (e.g. mistaken for bad sample preparation or not noticed without high magnification BSE images) and are in fact a widespread phenomenon in subglacial (and other?) basalts; or do our samples in fact represent a rarely preserved texture? Either way, they offer a new insight into the degassing and fragmentation of subglacial basalt. 40 Keywords: degassing; vesicles; open-system degassing; phreatomagmatic; tuffisite veins; Katla

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42 Introduction

43 Due to the ability of volcanic gases to rapidly expand at low pressure, volatiles are often considered 44 the driving force for explosive volcanism (Sparks, 1978; Moore et al., 1998; Blundy et al., 2006), with 45 violent fragmentation typically fuelled by high volatile contents and closed system degassing (i.e. where the volatiles remain trapped within the magma) (Anderson and Fink, 1989; Hoblitt and Harmon, 46 47 1993; Anderson et al., 1995; Martel et al., 1998; Villemant and Boudon, 1998; Villemant and Boudon, 48 1999; Villemant et al., 2003; Adams et al., 2006; Villemant et al., 2008; Humphreys et al., 2009; Owen 49 et al., 2013a). Transitions to effusive volcanism are often characterized by a shift towards open system 50 degassing behaviour, where volatiles are able to efficiently outgas from the magma (Jaupart and Allègre, 1991; Jaupart, 1998; Namiki and Manga, 2008; Owen et al., 2013b). 51

52 Magma outgassing occurs according to different mechanisms that strongly depend on melt 53 composition (Vergniolle and Jaupart, 1986; Houghton and Gonnermann, 2008). The low viscosity of 54 basalt, coupled with high volatile diffusivities in basaltic melts, enables bubbles to rapidly grow and buoyantly rise through the magma column, allowing efficient magma-gas separation and open-system 55 degassing (Wilson, 1980; Vergniolle and Jaupart, 1986; Edmonds and Gerlach, 2007; Menand and 56 Phillips, 2007; Houghton and Gonnermann, 2008). In contrast, the high viscosity and lower volatile 57 58 diffusivities in silicic melts hinder both bubble growth (Proussevitch and Sahagian, 1996) and melt-59 bubble decoupling (Sparks, 1978). However, high-Si magmas can still experience open system 60 degassing by becoming permeable, either with connected bubbles acting as a pathway for gas to flow 61 (Eichelberger et al., 1986; Westrich and Eichelberger, 1994; Klug and Cashman, 1996; Namiki and 62 Manga, 2008), and/or by the creation of tuffisite veins, permeable gas and ash filled fracture networks that temporarily exist within the magma conduit (Sparks, 1997; Tuffen et al., 2003; Saubin et al., 2016; 63 64 Farguharson et al., 2017).

65 Magma fracturing and tuffisite vein formation are currently thought to be limited to high viscosity 66 magmas such as rhyolites (Tuffen et al., 2003; Castro et al., 2012; Saubin et al., 2016), crystal-rich 67 andesites (Kolzenburg et al., 2012; Plail et al., 2014; Kendrick et al., 2016) and dacites (Nakada et al., 68 2005; Noguchi et al., 2008; Gaunt et al., 2014). Tuffisite veins within conduits, are proposed to form 69 when highly viscous magma experiences failure either due to gas overpressure or shear fracture within 70 the glass transition (Tg) interval. These fractures then act as a pathway for gas and fragmented 71 particles that eventually obstruct the vein. Subsequent welding of these particles results in vein 72 healing and permeable pathway occlusion, leading to re-pressurisation (Tuffen et al., 2003). Slow 73 crystallisation rates in rhyolite allow prolonged magma residence in the glass transition interval (hours 74 or days), providing sufficient time for repeated fracture-healing episodes within crystal-poor melt, and 75 creating superimposed generations of tuffisite veins within flow-banded glass (Tuffen et al., 2003; 76 Gonnermann and Manga, 2005).

In contrast, the low viscosity of basalt means that repeated episodes of fracturing and healing within
 the conduit are unlikely. Inherently, it is much harder to fragment/fracture basaltic magma at eruptive

79 temperatures (Papale, 1999; Giordano and Dingwell, 2003) without decreasing temperature, increasing crystallinity (Giordano and Dingwell, 2003; Houghton and Gonnermann, 2008; Namiki and 80 81 Manga, 2008) and/or by invoking rapid decompression and/or magma-water interaction (Giordano and Dingwell, 2003). These processes will all hinder subsequent sintering, as rapid cooling would serve 82 83 to quench the magma and high crystallinities lower the available melt fraction. Therefore, even if 84 fracturing were able to occur in a crystal-free basaltic melt, rapid crystallisation will limit its residence 85 time in the Tg interval, allowing little time for sintering, vein healing or repeated fragmentation cycles 86 (Tuffen et al., 2003). It is perhaps not surprising that, to our knowledge, there is no documented evidence of tuffisite-like features within basaltic magma. 87

In this paper we present some of the first observations supporting transient particle transport and
 sintering within permeable pathways (fractures, connected vesicles and inter-clast void spaces) in a
 high-temperature, crystal-poor basalt. This process likely preceded powerful hydromagmatic
 fragmentation, emphasising a need to better understand the influence of open-system degassing and
 clast recycling in basaltic eruptions.

- 93 2. Materials and methods
- 94 2.1 Geological background and sampling

95 Samples were collected within deposits from the 1918 A.D. VEI 4 (Smithsonian, 2016) subglacial 96 basaltic eruption of Katla (K1918) in South Iceland (Fig. 1a). The eruption took just two hours to melt 97 through ~400 m of overlying ice (Mýrdalsjökull glacier), after which both an ash plume and a 98 jökulhlaup (glacial flood) were observed (Tómasson, 1996; Sturkell et al., 2010). The plume was 14 km 99 high, and deposited ash over 50,000 km² of land (Larsen, 2010). The jökulhlaup transported 0.7-1.6 100 km^3 of tephra (Larsen, 2000), with an inferred peak discharge rate of >300,000 $m^3 s^{-1}$ (Tómasson, 101 1996), making it the 14th most powerful flood of the Quaternary (last 2.6 million years) (O'Connor and 102 Costa, 2004). Both the jökulhlaup and the extreme melting rate of the glacier were exceptional and 103 cannot be readily explained by existing models of convective magma-ice heat transfer (Gudmundsson et al., 1997; Hoskuldsson and Sparks, 1997; Wilson and Head, 2002; Gudmundsson, 2013; Woodcock 104 105 et al., 2014; Woodcock et al., 2015; Woodcock et al., 2016).

Samples characterised in this study were collected from both the air-fall and the jökulhlaup deposit.
 Wind blew ash from the eruption plume in many directions but predominantly to the NE (Larsen, 2010;
 Larsen et al., 2014), with ~300 g m⁻² reaching North Iceland (Larsen et al., 2014). However, the ash was
 poorly preserved following the eruption. Nevertheless, K1918 ash can be found in select soil horizons
 around Katla (Óladóttir et al., 2005; Óladóttir et al., 2008), and the thickest deposit is observed as a
 layer in the Mýrdalsjökull ice, which is now being exhumed (Gudmundsson, 2013, pers. comm).

112 The K1918 jökulhlaup deposit has been well characterised by a variety of studies (e.g. Maizels, 1992; 113 Maizels, 1993; Tómasson, 1996; Duller et al., 2008; Russell et al., 2010). Maizels (1992) identified four 114 units; 1: a basal unit of massive gravels and imbricated clast-supported gravels that represents rising 115 flow, overlain by 2: massive pumice granules interpreted to be part of a flow surge, overlain by 3: 116 trough cross-bedded pumice granules, and finally 4: horizontally bedded pumice granules and pumice 117 sands, with the top two units representing more fluid waning stages of the jökulhlaup. The deposit is 118 ~12 m thick in proximal regions, decreasing to 4 m thick at the coast, some 18 km away (Maizels, 119 1992).

120 Air-fall tephra was collected from Sólheimajökull glacier (Fig. 1a) where a ~40-cm thick layer of typically sub-cm clasts are preserved in the ice (Fig. 1b). These samples are prefixed "Sol". Jökulhlaup 121 122 samples (prefixed "Mul") were collected from the banks of the Múlakvísl river, where river 123 downcutting has exposed a clear vertical cross-section through the deposit (Fig. 1c). In both settings 124 multiple locations were sampled, which is denoted by the second part of the sample name. At 125 Múlakvísl we focussed on a 3 m high exposure (Figs. 1a, 1c). In both locations, there were multiple 126 layers, each of which was sampled. The layer makes up the third part of the sample name. For the air-127 fall tephra six layers were documented and labelled A-F (Fig. 1b). The observed stratigraphy at the jökulhlaup deposit matches the units described in Maizels (1992), Duller et al. (2008) and Russell et al. 128 129 (2010) but with only units 2-4 exposed (Fig. 1c). The lower and upper half of unit 3 appeared more 130 lithic- and juvenile-rich respectively and was separated by a vein. As a result, we collected two samples 131 from unit 3 and labelled them 3a and 3b respectively (Fig. 1c).

132 Grain size distributions revealed that for the samples collected, the largest clast sizes typically fell 133 within the -3 to -4 φ category (8-16 mm) and had a peak at the -1 to -2 φ category (2-4 mm). Typical 134 air fall samples had a second modal group with a high proportion < 125 μ m (>3 ϕ). However, it should 135 be noted that, in both settings, the grain size distribution is probably a reflection of the transportation, 136 and potentially the re-mobilisation, history and not thought to represent the true volcanic deposition. 137 This is especially true of the jökulhlaup deposit where it is thought that the majority of the fines and 138 most of the early material was washed out to sea (Duller et al., 2008). Nevertheless, for each key 139 sample, four representative 8-16 mm and nine 2-4 mm clasts were chosen, numbered and made into 140 thin sections. The clast size and number make up the fourth and fifth (final) parts of the sample name, 141 respectively. In total 100 clasts of this size were thin sectioned and of these, 26 representative clasts 142 (9 air-fall and 17 jökulhlaup samples) were chosen for backscatter imaging. Thin sections were also 143 made of the 250-500 μ m clast size to supplement the geochemistry data.

Each sample name, therefore, has the following format 1) environmental setting 2) location number,3) layer/unit reference, 4) clast size, 5) clast number.



Figure 1: (a) Map of the Katla area showing the sampling locations with the inlet showing theposition in South Iceland; (b) Cross-section through the air-fall deposit on Sólheimajökull glacier

showing units A-F; (c) Cross-section of the jökulhlaup deposit at Múlakvísl showing the different
units sampled (units 2 to 4 from Duller et al. (2008)) with a meter rule for scale.

152 2.2 Geochemistry and imaging (EPMA)

153A Field-emission JEOL Hyperprobe JXA-8500F Electron Probe Micro-Analyser (EPMA) at the154University of Hawaii was used to acquire (a) back-scattered electron (BSE) images, (b) compositional155(X-ray distribution) maps and (c) spot analyses. Accelerating voltages of 15, 20 and 20 keV and beam156currents of 10, 30-50, and 10 nA were used for (a), (b) and (c) respectively. Spot sizes of 1 µm, 2 µm

and 10 µm were used for Fe-Ti oxides, K-rich particles and matrix glass, respectively. On-peak count
times of 30 s were used for Si, Al, Fe, Mn, Na, K, P, and 65-70 s for Al, Mg, Ca, and Cl. Time-dependent
intensity corrections were used for glass analyses when significant Na loss or Si gains were detected
(e.g. Shea et al., 2014).

To assess compositional heterogeneity and relative element abundances in select samples, compositional maps of Fe, Ca and K were obtained using three of the five spectrometers and dwell times of 40-45 msec/pixel. Raw data was used to make single 2D intensity matrices, which were then combined as individual channels into a single RGB composite image. The other two spectrometers measured S and either F or Na. Intensity matrices for all measured elements are provided in Appendix 1 (Figs. A1-A3).

Spot analyses were mainly performed on magnetite and ilmenite crystals for geothermometry (see section 2.3). A limited number of glass analyses were also acquired and compared to EPMA data collected from the University of Edinburgh. There, > 200 glass measurements were made on more than 50 different air-fall and jökulhlaup clasts. All analyses were carried out at 15 kV with a 5 μm spot size. Beam currents of 2 and 80 nA were used for major and minor/trace elements respectively as per Hayward (2011). Analyses with totals < 97 wt.% and those with a clear influence of crystals were rejected.

174 2.3 Estimating eruption temperature (geothermometry)

To better constrain the physical parameters of the melt (e.g. viscosity, diffusion rates, sintering rates etc.) oxide geothermometry was used to estimate the magma eruptive temperature. Using the method described in section 2.2, 61 measurements were made of magnetite and ilmenite crystals within both jökulhlaup and air-fall samples. These were converted into temperatures using the Fe-Ti oxide geothermobarometer model of Ghiorso and Evans (2008). Measurements were rejected if either SiO₂ exceeded 1 wt.%, as this may reflect mixed analyses with surrounding glass, or if they failed the equilibrium test of Bacon and Hirschmann (1988).

182 2.4 Glass H₂O content (FTIR)

The glass water contents of five clasts (one air-fall and four jökulhlaup) were measured using Fourier Transform Infrared Spectroscopy (FTIR). A Thermo Nicolet IR interferometer, with KBr beamsplitter, Continuum Analytical microscope and MCT-A detector were used at Lancaster University. Each measurement (including background analyses) constituted 256 spectra collected at 4 cm⁻¹ resolution over the range 600-5500 cm⁻¹. A minimum of ten measurements were taken per sample with a 100x100 µm aperture.

189 H₂O contents (C) were determined using the Beer-Lambert law (e.g. Stolper, 1982)

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(1)

where M_w is the molecular weight of water (18.02 g mol⁻¹), A is peak height, d is sample thickness (in cm), ρ is density (2,770 g l⁻¹ estimated using the density calculator of Bottinga and Weill (1970) and a

 $C = \frac{M_w A}{d\rho\epsilon}$

representative composition of K1918 basalt taken from Óladóttir et al. (2008)), and ε is the absorption coefficient (I mol⁻¹ cm⁻¹).

Total H₂O (H₂O_t) concentrations were determined using the absorption peak at 3550 cm⁻¹ and an
 absorption coefficient of 63 l mol⁻¹ cm⁻¹ (Dixon et al., 1988). Sample thickness was determined using a
 Mitutoyo digital displacement gauge accurate to ±3 μm.

The characteristic double CO_3^{2-} peaks (1515 and 1435 cm⁻¹) and the molecular H₂O (H₂O_m) peak at 1630 cm⁻¹ were indiscernible, suggestive of very low CO₂ and H₂O_m concentrations. As post quenching hydration favours H₂O_m (Yokoyama et al., 2008; Denton et al., 2009), it is unlikely that our K1918 samples have undergone this process.

- 203
- 204 3 Results
- 205 3.1 Overview of clasts

206 Clasts tend to be brown, black (or a mixture of the two) and contain sparse phenocrysts. The brown 207 material (e.g. Figs. 2b, 3, 4bii) is clear glass (sideromelane), with few microlites (typically 5-25% for air-208 fall and < 5 % for jökulhlaup clasts), in contrast to the black (tachylite) material (e.g. Fig. 4aii), which is 209 relatively microlite-rich (~30-70%), opaque and contains bubbles that are largely obscured in plane-210 polarised light (ppl) microscopy. Air-fall clasts are typically 40-50% vesicular whilst jökulhlaup clasts 211 are ~60-75% vesicular, with a large number of small bubbles (Owen et al., 2017). Bubble and microlite 212 textures are often locally heterogeneous throughout each clast. The clasts often contain fractures, 213 connected bubbles and textural evidence of sintering, which are particularly well preserved in the 214 jökulhlaup samples (see sections below). Note that in this paper, we will use the term 'sintering' in reference to a texture that shows any of the stages of sintering from contact point fusing, to full 215 216 coalescence into a solid mass. In addition to the brown and black clasts, there is a small percentage of 217 pale/clear lithic clasts that are largely void of both phenocrysts and vesicles.

218 3.2 Fractures

219 Most clasts contain fractures that have either a ragged, sharp angular appearance (Fig. 2) or a 220 smoother, more rounded morphology (Fig. 3). Fractures observed are typically millimetric in length 221 and tens to hundreds of μ m wide, however observations were limited by clast size (< 16 mm). It is 222 common for the fractures to be partially filled by ash particles. Ash also occurs within neighbouring 223 vesicles connected to the fractures, but tends to be absent from nearby isolated bubbles (e.g. Fig. 2c). 224 The fracture- and vesicle-occupying particles shall herein be referred to as 'particles', and the host as 225 'clast' to avoid confusion. The fracture-hosted particles are typically of μm scale but can reach 250 μm 226 in the largest fractures. They are predominantly composed of vesicle-free basalt (both sideromelane 227 and tachylite), but lithics are also present, and some of the larger particles can contain vesicles. The 228 particles tend to be either angular or well rounded, and sometimes appear welded to nearby surfaces 229 (Figs. 4, 5a).



Figure 2: Images showing an angular fracture containing ash particles within sideromelane. (a) A BSE mosaic image showing the whole clast - jökulhlaup sample Mul 6 unit 3a-1 8,000-16,000 2a (~5x8 mm). Rectangles with solid lines highlight the area shown in parts b and c, whilst the dashed line rectangles outline areas shown in Figures 4b, 5c and 7. Figure 8 features within the area shown by Figure 5c but is too small to depict here; (b) Photomicrograph in ppl detailing the fracture; (c) Simplified BSE image showing the same area as b, where bubbles have been coloured black, glass white, the background

238 (and bubbles connected to the background) grey, and ash particles in red.



Figure 3: Photomicrographs in ppl showing a rounded fracture within sideromelane, which contains ash particles near the clast margin. (a) mosaic overview of the whole clast – jökulhlaup sample Mul 6 unit 4-1 8,000-16,000 4a (~6x9mm); (b) detail of the fracture, showing a transition from angular (left) to rounded (right) as it approaches the clast margin (far right). The rounded part of the fracture contains ash particles.

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249 Figure 4: Images of fractures containing partially sintered particles. The ash particles are connected to the host, and each other, via 'necks' of various sizes. The suffix refers to the image type, with (i) 250 251 denoting BSE images and (ii) denoting thin section images taken in ppl. (a) A ~20 µm wide partially 252 filled fracture within the tachylite part of a jökulhlaup clast - Mul 6 unit 4-1 8,000-16,000 3a (see 253 Figure 10a for context). Rounded tachylite ash particles have sintered onto the fracture wall with the 254 boundary marked by a microlite chain; (b) A ~100 µm wide fracture that has almost healed within a 255 sideromelane jökulhlaup clast - Mul 6 unit 3a-1 8,000-16,000 2a (see Figure 2a for image location). 256 The fracture is filled with rounded particles, which have sintered to each other and the fracture 257 walls. The area immediately adjacent to the fracture is less vesicular than the surrounding melt.

As can be seen in Figure 4, the particle-filled fractures, which are fairly evident in the BSE images on
the left, are not at all clear in the ppl images on the right. This is especially true for the tachylite
sample (Fig. 4a) where the opaqueness of the matrix makes the feature (and many of the bubbles)
invisible in ppl (Fig. 4aii). This is not only true for particle filled fractures but also particle filled

262 bubbles and in fact most examples of sintering.

Particles within fractures and bubbles are noticeably vesicle-poor compared to their host clasts, likely reflecting their grain size being smaller than the predominant bubble size (Fig. 4ai). Most particles are

in fact void of vesicles. There are, however, some exceptions, for example the larger fracture-filling

266 particles in Figure 4b share similar bubble contents and textures to the melt immediately surrounding

the fracture. In turn, this area is less vesicular, with typically smaller and less abundant bubbles than
 material >100 μm away.

269 Compositional mapping of a particle-filled fracture (Fig. 5a) shows that most particles have 270 compositions similar to the surrounding microlite-free host basaltic melt (sideromelane). However, 271 the fracture also contains a significant percentage of K-rich (blue) particles (Fig. 5a), which are mostly 272 also Na-rich, although some are Na-poor (Appendix 1; Fig. A1). There is also a large Na-rich phenocryst (dark green in Fig. 5a) and microlites of this composition within some of the K-rich particles. These 273 274 K/Na-rich particles likely represent fragments of silicic glass, alkali feldspar, and albitic plagioclase 275 from a more evolved magma. Particles are predominantly sub-angular, but a significant proportion, 276 especially of sideromelane, display rounded morphologies. Some rounded sideromelane particles 277 have viscously deformed around K-rich particles, whilst others are sintered to the fracture walls, with 278 the former boundaries expressed by oxide microlite chains (pink in Fig. 5a). Additional microlite chains 279 occur in the basaltic glass surrounding the fracture, outlining domains that are similar in size and shape 280 to the particles within the fracture. These microlite chains are S-enriched (Fig. A1). Also present in the 281 surrounding glass are a few additional K-rich fragments and partially collapsed vesicles.



Figure 5: Compositional maps showing a particle-filled fracture (a), a particle-filled bubble (b), and an 285 286 area of extensive particle-bearing microlite chains (c). Colours represent relative element abundances 287 according to the legend in the bottom left corner (Fe-rich=red, Ca-rich=green, K-rich=blue, Fe and Ca 288 -rich=yellow, Ca and K –rich=turquoise, K and Fe –rich=pink and areas rich in Fe, Ca and K=white). (d) 289 offers an interpretative summary. (a) A $^{\sim}200~\mu m$ wide particle filled fracture within a sideromelane 290 jökulhlaup clast - Mul 6 unit 3a-1 2,000-4,000 3a. The particles are both basaltic sideromelane and 291 fragments from a more evolved melt, and have both sub-angular and rounded morphologies. Microlite 292 chains are present in the surrounding basaltic glass. See Figure 16 for fracture location and ppl/BSE 293 images; (b) A particle-filled irregularly shaped bubble which appears to be connected to other particle-294 filled bubbles (e.g. upper right). The host is microlite-rich tachylite from an air-fall clast - Sol 3E 8,000-295 16,000 3a, but the particles are predominantly sideromelane basalt and K-rich particles from a more evolved melt. These particles are highly angular and small, and show slight near-horizontal lineation
with larger particles being near the upper and lower bubble walls. There is clear sample preparation
damage to the upper bubble wall as labelled; (c) An area of extensive microlite chains within a
sideromelane clast from jökulhlaup sample Mul 6 unit 3a-1 8,000-16,000 2a (see Figure 2a for context).
The microlites (pink) occupy the upper right part of the image and coincide with small fragments of
various compositions. The lower left part of the image is void of microlites and particles but contains
near-spherical bubbles. The dashed line rectangles outline areas shown in Figures 8a and 8b.

303 3.3 Infilled bubbles

Interconnected bubble networks also frequently contain particles (Figs. 5b, 6). As with the fracture filling particles, these are predominantly angular, but include a significant proportion that are well

306 rounded and sintered onto bubble walls (Fig. 7).

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309 Figure 6: BSE images showing examples (a and b) of particle-rich bubble chains in sideromelane

310 jökulhlaup clasts. In both examples, the bubble chain appears to widen as it approaches the clast

311 margin (upper right corner), however, the particles are most abundant at the opposite end. Particles 312 are predominantly absent from nearby isolated bubbles. (a) is from Mul 6 unit 2-1 8,000-16,000 2a

are predominantly absent from nearby isolatedand (b) is from Mul 6 unit 3a-1 8,000-16,000 3a.



316 Figure 7: Images (a: BSE; b: ppl) showing sintered particles within bubbles in a sideromelane jökulhlaup 317 sample - Mul 6 3a-2 8,000-16,000 2a (see Figure 2a for context). A round particle (~20 μm) is sintered 318 to the wall of a large bubble (right side of the image), with a microlite chain marking the boundary. On 319 the far left of the image is the edge of a bubble with two smaller sintered particles adhered to the 320 bubble wall. The centre of the image is occupied by an area with a high number density of small 321 bubbles, which appears flow-banded in part (b) and is outlined by a microlite chain. In the 322 neighbouring glass beneath, there are partially collapsed vesicles, clearly visible in part (a), which 323 pinch out into additional microlite chains; these also appear as flow bands in part (b).

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A compositional map was acquired within an irregular, highly deformed particle-filled bubble, which 325 326 appears to form part of a series of interconnected bubbles (Fig. 5b). The particles are very small (<<63 327 μ m), generally angular and crudely sorted, with linear bands of finer grains in the bubble centre and 328 larger particles on either side. The orientation of this band matches the orientation of the main axis 329 of the bubble. The surrounding tachylite matrix consists of Ca-rich (green) and Fe- and K-rich (pink) 330 microlites, which are most likely plagioclase, diopsidic pyroxene and Fe/Ti oxides (± orthopyroxene) 331 typical of mafic magmas and Katla basalts (Lacasse et al., 2007; Budd et al., 2016). This phase 332 assemblage contrasts with particles inside the bubble, which are predominantly either K-rich (blue) 333 lithics, more likely belonging to a more evolved melt, or sideromelane (grey) juvenile glass particles. 334 There are only two significant places where the particles match their host composition and their 335 geometry suggests they are fragments of broken bubble wall and therefore are most likely a product 336 of sample preparation (as labelled in Figure 5b). The foreign nature and the apparent sorting and 337 imbrication of the remaining particles leads us to believe this is a magmatic feature.

338 3.4 High density microlite chains

The last compositional map (Fig. 5c) focused on an area containing abundant microlite chains within sideromelane. The microlite chains seem to connect crudely oval vesicles (Figs. 5c, 8), some of which appear partially collapsed (Fig. 8b). Many of the bubbles are surrounded by microlites (Fig. 8b). Various Fe-rich (red) and K-rich (blue) phases coincide with the location of microlite chains (Fig. 5c). These phases have both angular (Fig. 8a) and rounded (Fig 8b) forms, suggesting that they are variably relaxed particles of a fragmented melt. The Fe-rich fragments (Fig. 5c) are either Fe-sulphides or Fe-Ti 345 oxides. The K-rich fragments (Fig. 5c) are likely feldspar (both alkali and plagioclase) and silicic glass.

346 EPMA analyses on select K-rich fragments reveal compositions consistent with dacite and plagioclase

347 feldspar (see section 3.8). K-rich particles within all the Figure 5 maps are therefore interpreted to be

fragments of silicic glass and/or crystal fragments derived from a more evolved melt. In contrast, the lower left hand region in Figure 5c is almost devoid of both microlite chains as well as Fe/K-rich

350 particles and appears more vesicular with relatively large, near spherical bubbles.

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353 Figure 8: Images showing dark grey fragments of Si-rich material (revealed to be dacite and

354 plagioclase feldspar through EPMA spot analyses; see section 3.8 within a basaltic glass (pale grey)

riddled with microlite chains (white lines) and small bubbles (black), some of which appear partially collapsed. This area has also been mapped compositionally (Fig. 5c) and the whole clast (jökulhlaup

collapsed. This area has also been mapped compositionally (Fig. 5c) and the whole clast (jökulhlat
 sample Mul 6 unit 3a-1 8,000-16,000 2a) can be seen in Figure 2a. The Si-rich material appears in

- both angular (a) and rounded (b) forms.
- 359 3.5 Other sintered textures

360 Sintering can be at times so extensive that it is unclear whether the original void was a bubble or a

- 361 fracture (Figs. 9, 10). In total, of the basaltic clasts for which there are BSE images, 12 of the 15
- 362 jökulhlaup clasts and 8 of the 9 air-fall clasts, show evidence of particle sintering.



365 Figure 9: Images (in ppl (a) and BSE (b-d)) showing sintered particles within void spaces. The rectangles 366 indicate areas detailed in other parts of this image as labelled. (a) Mosaic overview of the whole clast 367 which is a tachylite jökulhlaup sample - Mul 6 unit 4-1 8,000-16,000 2a (~5x11 mm). There is a ~2 mm 368 sized dark lithic in the upper part of the image and abundant particle filled bubbles, which appear pale 369 brown in this image and appear to occupy distinct areas; (b) An overview of the upper part of the clast, 370 with the lithic appearing dark grey in the bottom right hand corner. Bubbles (black) are nearly spherical 371 on the left, but are significantly more irregular in the centre of the image. Irregular void spaces often coincide with sintered particles; particularly sintered areas are indicated with arrows; (c) An example 372 373 of one of the highly sintered areas in part (b). The void in the image centre contains a large number of 374 rounded particles; some of which are partially sintered onto the walls and others becoming 375 indistinguishable from the matrix; (d) The boundary between the host clast (pale grey, upper right) 376 and the lithic (dark grey) which contains a highly distorted phenocryst (white). There is a \sim 20 μ m wide 377 void space between the two enclosing rounded particles of tachylite, which in some cases are sintered 378 onto the host clast.





381 Figure 10: Examples of sintering within the jökulhlaup clast Mul 6 unit 4-1 8,000-16,000 3a. (a) Photomicrograph mosaic image in ppl showing an overview of the whole clast (~6x9 mm), which is 382 383 mixed tachylite (left) and sideromelane (right). Solid and dashed line rectangles express the areas 384 shown in other parts of this figure and other figures, respectively, as labelled; (b) a semi-circular 385 feature with a high abundance of rounded tachylite and sideromelane particles. It is unclear whether 386 this was once a fracture which has welded shut or whether the whole area shows variably sintered 387 clasts (i: BSE; ii: ppl); (c): detail of the area outlined in part (b) showing partially collapsed bubbles that 388 contain rounded, sintered particles with microlite chains extending from the pinched point of each 389 vesicle. (i: BSE; ii: ppl); Parts d-g show BSE images of the upper part of the clast; (d) overview mosaic 390 image. The area shows extensive but heterogeneously distributed sintering. Arguably the large dense 391 areas in the upper left and right-hand corners could represent former large particles spanning several 392 hundred microns in diameter that have fused with the host. (e) and (f) illustrate the potential lower 393 boundaries of these larger former particles, with small sintered particles prominent in this void space. 394 The dashed rectangle in part e outlines the 'fracture' shown in Figure 4a. (g) Detail showing the sintering interface between two rounded particles of tachylite, which is marked by a microlite chain 395 396 of oxides.

397

398 3.6 Geothermometry

399 Fe-Ti oxides yield an average temperature of T=1,045 ± 31 °C (n=45 pairs) for K1918 basalts (Fig. 11).

- 400
- 401





403

Figure 11: Magma temperatures plotted against oxygen fugacity, estimated from magnetite and ilmenite compositions using the model of Ghiorso and Evans (2008). NNO refers to a nickel-nickel oxide oxygen buffer. Jökulhlaup and air-fall samples are prefixed by "Mul" and "Sol" and are coloured blue and red respectively. The error bars represent average absolute deviation (44°C and 0.34 log units for temperature and oxygen fugacity (fo₂) respectively), calculated by Blundy and Cashman (2008) for the Ghiorso and Evans (2008) model.

410 3.7 FTIR

One air-fall clast was measured and found to have a glass H₂O content of 0.08 wt.% (Table 1) consistent
 with degassing to atmospheric conditions (e.g. Tuffen and Castro, 2009). K1918 jökulhlaup clasts have

413 glass H_2O contents of 0.15-0.31 wt.% (Table 1), consistent with a pressure elevated beyond

414 atmospheric, but not equivalent to the full weight of the glacier. Using the pressure-solubility model 415 VolatileCalc (Newman and Lowenstern, 2002) and assuming a Si-content of 47 wt.% (consistent with 416 Óladóttir et al. (2008)), 0 ppm of CO₂ (as measured) and a temperature of 1,045 $^{\circ}$ C (estimated using geothermometry; section 3.6), the jökulhlaup clasts likely quenched under 0.29-1.11 MPa of pressure. 417 418 The loading pressure (P in Pa) can be estimated by multiplying gravity (g; 9.81 m s^{-2}) by the density (ρ 419 in kg m⁻³) and thickness of the load (h in m).

420

$$P = \rho g h$$

421

(2)

422 Thus a 400 m thick glacier (as was inferred to be the ice thickness over the vent in 1918 A.D.; (Sturkell 423 et al., 2010)), with a density of 917 kg m⁻³ (Tuffen et al., 2010) would exert a pressure of 3.60 MPa, 424 approximately four times greater, or more than the jökulhlaup clasts experienced syn-quenching. 425 There is no systematic variation in H₂O content within the stratigraphic section sampled in Figure 1c. 426 Therefore taking the average jökulhlaup glass H_2O content (0.22 wt.%), we can estimate that the 427 average load during quenching was 0.59 MPa. This equates to approximately 65 m of ice, 60 m of 428 water, 20 m of solid rock or 10 m of 50% vesicular basalt, assuming densities of 917, 1,000 and 2,770 429 kg m⁻³ for ice, water and K1918 basalt respectively, with errors of ~20%. However, natural samples 430 often experience loading by a combination of materials (Tuffen et al., 2010; Owen et al., 2012; Owen 431 et al., 2013b; Owen, 2016; Owen et al., in press).

432

433 Table 1: FTIR results showing glass H₂O contents for five K1918 clasts.

Environmental Setting	Sample name	Mean H₂O content (wt. %)	Standard deviation (wt. %)	Number of successful analyses
Air-fall	Sol1A 8,000-16,000 4b	0.08	0.01	11
Jökulhlaup	Mul6 unit 2-1 8,000-16,000 2b	0.22	0.02	9
Jökulhlaup	Mul6 unit 2-1 8,000-16,000 4b	0.21	/	1
Jökulhlaup	Mul6 unit 4-1 8,000-16,000 1b	0.31	0.04	2
Jökulhlaup	Mul6 unit 4-1 8,000-16,000 4b	0.15	0.05	2

434

435 3.8 Geochemistry

436 There is strong consistency between the EPMA data gathered at the University of Hawaii, that gathered at the University of Edinburgh and that already published for Katla (Fig. 12). EPMA data (Fig. 437 438 12) shows a bimodal distribution with sideromelane and tachylite clasts having ~47 wt.% SiO₂ 439 consistent with K1918 data published by Óladóttir et al. (2008), although the tachylite data is more 440 scattered, likely due to these clasts being more microlite-rich (e.g. Fig. 4). In addition to the brown 441 (sideromelane) and black (tachylite) clasts, most samples had a small percentage of clear/pale clasts

which have a trachyte/trachydacite to rhyolite composition consistent with older silicic Katla eruptions 442 443 (Lacasse et al., 1995; Newton, 1999; Larsen et al., 2001; Lacasse et al., 2007; Óladóttir et al., 2008). 444 Sintered sideromelane particles have similar compositions to the larger discrete sideromelane clasts 445 (Fig. 12). Sintered K-rich particles found within an area of extensive microlite chains within a 446 sideromelane clast (Fig. 5c) were found to have compositions consistent with plagioclase feldspar and 447 dacite (Table 2). Although a relatively high Fe content (potentially from crystal contamination) meant 448 they were excluded form Figure 12, they are also consistent with the published data from older silicic 449 Katla eruptions.









- 461
- 462 Table 2: EPMA analyses of three particles within the area of extensive microlite chains in Figure 5c
- (sample Mul 6 unit 3a-2 8,000-16,000 2a). The analyses are consistent with the compositions of dacite
 (top two) and plagioclase feldspar (third analysis).

Field Code Changed

Analysis number	SiO ₂	TiO ₂	Al ₂ O ₃	FeO	MnO	MgO	CaO	Na₂O	K₂O	P ₂ O ₅	Cl	TOTAL	Inferred material
1	63.76	1.85	13.45	7.39	0.16	1.31	4.19	2.09	2.31	0.21	0.08	96.79	dacite
2	65.70	1.67	14.83	6.66	0.15	1.35	3.64	4.13	2.17	0.21	0.07	100.57	dacite
3	54.45	0.33	28.60	1.40	0.00	0.29	11.33	4.99	0.23	0.00	0.00	101.63	plagioclase feldspar

466 4. Discussion

467 4.1 Evidence of gas and ash transport within permeable pathways

468 Many of the samples contain fractures and/or have connected bubbles, some of which contain small 469 particles of fragmented material. It is possible for such features to form during poor sample 470 preparation. Grinding during thin section making may fracture the rock, break thin bubble walls 471 connecting vesicles and produce small particles which may then lodge in void spaces. It is also possible 472 for fractures to form during quenching. However, both cooling-contraction cracks and sample 473 preparation cracks should be brittle as they happen < Tg, whereas most of our observed fractures 474 show ductile morphologies (Figs. 3b,4,5a), suggesting they experienced temperatures > Tg. Also, we 475 would expect particles distributed during sample preparation to be random. However, what we have 476 observed in the K1918 deposits is a very organised distribution of particles throughout the clasts.

477 Most of the particles are found in fractures and bubbles that are connected, and rarely within isolated 478 bubbles (Figs. 2-6). When particles do occur in apparently isolated bubbles, they are often in close 479 proximity to a bubble chain (e.g. Figs. 2c, 6a) and we suspect that they are actually connected in the 480 third dimension. Figure 9 shows clear areas of infilled bubbles and clear areas with none. These 481 distribution patterns are hard to explain with poor sample preparation. Furthermore, the particle filled 482 bubble in Figure 5b seems to show layering parallel to the axial plane of the bubble, consistent with 483 clastic deposition of particles within a stream of gas, as documented in silicic tuffisite veins where 484 internal laminae are parallel to fracture walls (Tuffen et al., 2003).

485 What is more, compositional maps (Fig. 5) reveal that within both fractures (Fig. 5a) and bubbles (Fig. 486 5b) the phase assemblage and composition of the particles is at least partly distinct from that of the 487 host, yet consistent with K1918 textures and geochemistry. All three compositional maps in Figure 5, 488 reveal silicic particles and felsic crystal fragments that have compositions consistent with older silicic 489 eruptions (Fig. 12). It is most likely that particles introduced through grinding in thin section preparation would be exclusive to the host. In Figure 5b, there is almost a 100% inconsistency; there 490 491 are only a few particles consistent with the composition and textures of the tachylite host, with the 492 vast majority being particles of silicic and basaltic glass. We hypothesise that based on the tachylite 493 particles being in close proximity to the host and with a jigsaw like fit, that these particles do represent 494 poor sample preparation. However, the vast majority of the particles cannot be explained by this and 495 we speculate that they are a primary feature.

The presence of sintered particles (Figs. 4,5a,7,9,10) is further evidence that particle deposition was a syn-eruptive process as sintering suggests that some of the particles experienced temperatures > Tg within the fractures/connected bubbles. Some fractures and particle-particle contacts are almost fully healed (e.g. Fig. 4b), making it difficult to deduce the original fracture wall or particle boundary (e.g Fig. 5a). Therefore, fractures may have been wider than currently observed. The presence of K-rich particles, collapsed vesicles and microlite chains in the host immediately surrounding the fracture in
 Figure 5a further supports this notion. This thorough healing and compaction is analogous to welded
 rhyolitic tuffisites (Tuffen and Dingwell 2005).

504 Furthermore, there are textures consistent with clast re-amalgamation; Figures 9 and 10, seem to 505 show former clasts embedded within larger clasts, with small particles sintered to the former clast 506 walls and to each other, within boundary zones (Figs. 9d, 10f). This is germane to tuffisite vein interiors 507 (Saubin et al., 2016) again suggesting larger permeable pathways than the preserved features.

Evidence for volatile transportation can be seen as local heterogeneity in bubble textures. We
speculate that the vesicle-poor area neighbouring the fracture in Figure 4b failed to vesiculate as much
as the surrounding glass due to volatile depletion following diffusive loss into the degassing fracture
(see section 4.6). Similar textures have been observed in rhyolitic Chaitén bombs (Saubin et al., 2016;
Webb et al., 2017).

513 We therefore hypothesize that bubbles (Figs. 5b,7,9) and fractures (Figs. 2,3,4,5a) have acted as 514 pathways for the transportation of both juvenile and lithic material, analogous to tuffisite veins in 515 silicic pyroclastic material (Rust et al., 2004; Saubin et al., 2016). Systematic transport is shown by the 516 organised distribution of particles and was likely facilitated by gas flow evidenced by diffusive loss 517 shown by the apparent vesicularity gradient surrounding the fracture in Figure 4b. Ductile fracture 518 morphologies and sintering suggest that both the fractures and particles within them experienced 519 temperatures > Tg and thus we believe it to be a syn-eruptive process that likely occurred within the 520 conduit.

521 4.2 Evidence of former gas and ash transport within healed permeable pathways

Partially collapsed vesicles surround many of the features of interest e.g. the fracture in Figure 5a, the
vesicles containing sintered particles in Figure 7, the area of intense microlite chains and Si-rich
particles in Figure 8 and the sintered particles in Figure 10c. These collapsed vesicles always transition
into microlite chains (Figs. 5a, 7, 8b and 10c). Furthermore, the bubbles in Figure 8b are surrounded
by microlites. Thus, we believe some of the microlite chains to represent healed vesicles.

527 Microlite chains also seem to have formed between sintered particles (e.g. Figs. 4ai and 10g). 528 Arguably, the similarity in size and morphology of the microlite chains in Figure 5a to the intra-fracture 529 particles could mean that they represent the boundaries of former particles that have since melted 530 into the surrounding host.

531 Whether microlite chains represent collapsed vesicles (e.g. Fig. 8b) or the margins of former particles 532 (e.g. Fig. 10g), they represent healed void-spaces. This is further evidenced by the relatively high S 533 content (Fig. A1) of the microlite chains surrounding the fracture shown in Figure 5a. Furthermore, it 534 suggests that these facilitated volatile transport with vapour-phase precipitation occurring on former 535 fracture/bubble/clast walls. In fact, in compositional maps, the microlite chains often appear similar 536 in colour to partially exposed bubbles (Fig. 5). The coincidence of these microlite chains with silicic 537 particles (e.g. Figs. 5c, 8) suggests that these healed voids also transported particles. Thus, we believe 538 the dense area of microlite chains and foreign particles in Figure 5c to show a healed transport 539 pathway for a particle-rich fluid phase.

540 There is an apparent compositional similarity between the matrix delineated by these microlite chains 541 and the surrounding microlite-free host basalt glass (Fig. 5c). However, the compositional maps shown 542 in Figures 5a and 5b indicate that basaltic fragments make up the majority of fragmented particles. 543 Therefore, it is likely that the permeable pathways that carried the K-rich particles in Figure 5c, also 544 transported basaltic particles. Evidence for this comes from the fragment of anorthitic plagioclase in 545 Figure 5c; an An-rich plagioclase likely has a mafic source. Apart from the mineral fragment, the basalt 546 is now indistinguishable from the host and therefore likely juvenile in origin. This suggests that Figure 547 5c represents a juvenile magma that fragmented (along with a minor percentage of felsic origin) and was then transported in a permeable pathway, which then collapsed and healed. Small vesicles then 548 549 grew in the scarred remains (Fig. 8).

550 Microlite chains also often surround fractures/permeable pathways (Figs. 5a, 10) indicating that they 551 may have partially healed. However, in Figure 5a, there are K-rich particles embedded into the basaltic 552 glass even beyond the zone of microlite chains surrounding the particle-filled fracture. This is evidence 553 of near complete healing. The apparent progressive welding around Figure 5a suggests that the 554 fractures were once considerably wider than they appear in some of the images; this healing of 555 pathways is akin to tuffisite veins (Tuffen et al., 2003).

556 4.3 Origin of the basaltic particles

557 Most of the bubble- fracture- filling particles have compositions consistent with K1918 basalt (Figs. 5 558 and 12) suggesting that it is juvenile material. This suggests that at least two fragmentation events 559 were involved in the production of the K1918 clasts: firstly to produce the small particles, which are 560 now trapped in the clasts, and lastly to produce and expel the clasts that were sampled. Sintering and 561 healing is apparent between these two events and it is possible that there were further fragmentation 562 events, however the rapid sintering rates of basalt makes it difficult to deduce how many 563 fragmentation events occurred in total.

The fact that the first fragmentation event did not expel the particles, suggests that it may have happened within the conduit, but at shallow level owing to the low H₂O contents (Table 1), low vesicularities of the particles (Figs. 2-10) and incomplete sintering (Figs. 2-10), suggestive of short residence times (see below).

Some clasts also show evidence of incomplete fragmentation (fracturing without significant displacement or expulsion) and brittle deformation (Fig. 13). The fragments of glass between the fractures share similar sizes, morphologies and vesicle textures to the fracture and bubble filling particles seen throughout the samples (e.g. Figs. 5, 6). This sample therefore represents an inefficient fragmentation event which failed to expel particles. It is likely linked with one of the early fragmentation events that produced fragments of glass that then infiltrated the remaining melt through connected bubbles and fractures.



Figure 13: A BSE image showing highly fractured sideromelane within jökulhlaup sample Mul 6 unit 4-1 8,000-16,000 1a, which could have acted as a source for small particles that later sintered with fractures and bubbles elsewhere (e.g. Figs. 5, 6). The dark grey shape at the bottom is a phenocryst.

579 4.4 Origin of the felsic particles

580 Although the majority of particles are basalt, every compositional map (Fig. 5) revealed the presence 581 of some K-rich particles which spot analyses indicated where fragments of feldspar and dacite (Table 582 2). Clear/pale clasts were also found to have compositions of a more evolved melt consistent with that 583 of older silicic Katla eruptions (Fig. 12) (Lacasse et al., 1995; Newton, 1999; Larsen et al., 2001; Lacasse et al., 2007; Óladóttir et al., 2008). In fact, there are extensive silicic outcrops within the Katla caldera, 584 including some close (<4 km) to the inferred 1918 A.D. vent (Jóhannesson and Sæmundsson, 2014). 585 586 We therefore hypothesize that the glassy K-rich particles represent fragments of evolved country rock 587 (dacite, trachyte/trachydacite and rhyolite) that were mobilised during fragmentation and 588 incorporated into connected fracture-bubble networks by a mobile fluid phase during ascent. This 589 possibility was recognised by Budd et al. (2016) who used mineral-melt equilibrium cystallisation 590 pressures to infer multiple magma storage regions at Katla with the potential for new rising magma 591 to intersect evolved magma within shallow-storage regions.

592 4.5 Particle morphology, sintering timescales, and link with composition

The morphology of the silicic particles is generally angular, in contrast to the basaltic particles, which are more often rounded (Fig. 5a). Within magmatic fractures that appear to have largely healed, the angular morphology of glassy silicic lithics (Figs. 5c & 8a) is surprising, as surface tension-driven shape relaxation would occur rapidly at basaltic eruptive temperatures.

597 We consider two possible explanations for why the silicic particles are generally less deformed than 598 basalt ones (Fig. 5a). Firstly, even though the Si-rich material was considerably hotter than its usual eruptive temperature, it would still have had a significantly higher viscosity than the basalt (Fig. 14)
and therefore the timescales for deformation, shape relaxation and sintering would have been
considerably longer (Vasseur et al., 2013). Secondly, being part of the country rock, the silicic material
was likely incorporated at far lower temperatures than the juvenile basalt (1,045 °C), and would thus
have needed to first rise to temperatures >Tg in order to deform, further hindering sintering.







Figure 14: viscosity as a function of composition, temperature and H₂O content, according to the
viscosity model of Giordano et al. (2008). For the 'basalt' and 'silicic' composition the analyses that
most closely resembles published data was used (Fig. 12). The red line depicts our inferred eruptive
temperature (1,045 °C). (a) viscosity plotted against temperature, for different compositions (colours)
and H₂O contents (line styles); (b) viscosity plotted against H₂O concentration for our inferred eruptive
temperature of 1,045 °C.

To investigate these two scenarios, and to produce some crude estimates of heating, relaxing and sintering timescales, we performed some basic first-order estimations (Appendix 2). The results show that owing to the small particle size, all of the intravoid particles shown in this study (typically << 100 μ m) would have likely reached thermal equilibrium within milliseconds (Fig. A4). The time taken to heat silicic particles (of this size) to basaltic magmatic temperature therefore seems insignificant.

619 Our inferred rounding/sintering timescales are only slightly longer (fractions of seconds) than 620 timescales for thermal equilibrium, but are strongly viscosity-dependent (Vasseur et al., 2013). We 621 therefore conclude that the rounding/sintering rate is the main process that controlled particle shape. 622 Even at 1,045 °C, which is significantly higher than its usual eruptive temperature, the viscosity of the 623 silicic magma will be high enough to extend the rounding/sintering timescale by one or two orders of 624 magnitude compared to basalt (Fig. A5). Consequently, for particles of comparable size, the basaltic 625 particles can thoroughly sinter and heal before the silicic particles have relaxed to a rounded shape. 626 These viscosity-controlled differences in rounding/sintering rate can therefore explain the angularity 627 of silicic particles within a healed matrix of basalt in Figure 5c, and the apparent deformation of a 628 basaltic particle around a similarly sized angular silicic particle in Figure 5a.

Estimating exact sintering times is difficult since timescales are extremely sensitive to composition,
 H₂O content and particle size (Fig. A5). These parameters will also be changing throughout the process,
 and it is difficult to deduce the original starting conditions from what is preserved in the samples
 (Pope, 2015).

Nevertheless, our basic calculations (Appendix 2) suggest that the sintering times, for the preserved particles within the fractures/bubbles/voids, was at most a few seconds. Any longer would have likely resulted in complete healing. For angular clasts, this process would have been even quicker, and/or the particles were injected below Tg; either way suggests near instantaneous injection, fragmentation and quenching. The fact that some of the voids contain both angular and rounded particles of similar sizes (e.g. Fig. 5a) suggest that there was momentarily a stream (albeit very short-lived) of gas and ash with particles entering the space at slightly different times.

Final quenching was likely closely linked with fragmentation at Katla. Fragmentation may have even
been triggered or enhanced by melt-water interaction. Even if not phreatomagmatic, located under a
glacier, there would have been abundant ice and meltwater present in close proximity to the vent.
FTIR analyses reveal that the jökulhlaup samples quenched under pressures consistent with loading
from ice or water (see section 3.7). The glassy nature of the sideromelane also implies rapid
quenching.

646 It should be noted that the inferred residence time of variably sintered particles is not necessarily an 647 indicator of the full duration of void opening. These voids, when first opened, may have been vapour-648 rather than particle-filled. Additionally, small (<100 µm) basaltic particles will fully sinter very rapidly (in fractions of seconds; Fig. A5), after which it will be difficult to recognise the former presence of 649 650 particles. Some fractures are neighboured by K-rich fragments, collapsed vesicles and microlite chains that form crude particle-shaped patterns (Fig. 5a). We hypothesise that these textures represent 651 652 former particles that fully annealed to the void walls, and that microlite chains often indicate former 653 permeable pathways/healed voids (e.g. Figs. 5c, 8; see section 4.2). However, it is difficult to 654 determine the extent of healed pathways, because chemical diffusion and viscous flow will 655 homogenise the sintered melt, ultimately leaving little/no trace of the former particle boundaries. For 656 these reasons, we suggest that the voids were open over seconds, and only the final split seconds of 657 the void life is preserved within our samples. We hypothesise that the preservation (particularly of 658 angular particles) was possible due to rapid quenching in water, which must have occurred almost 659 simultaneously with the final injection of particles. This indicates a very rapid succession of 660 deformation that was brittle (to form the initial particles), ductile (to sinter the particles) and then 661 brittle again (to expel and quench the particles).

662 4.6 Diffusive loss of volatiles through fractures, and timescale for open system degassing

To further investigate the lifetime of the permeable networks we provide estimates of diffusion timescales (Appendix 3). There are several lines of evidence that permeable networks have facilitated diffusive loss of volatiles. Firstly, microlite chains and bubble walls are usually S-rich (Figure A.1). These microlite chains were likely former voids and particle boundaries (see section 4.2), where S-rich gases circulated and deposited along the suture zones. Secondly, the glass surrounding some of the voids (e.g. Figure 4b) is noticeably deficient in terms of bubble textures compared to the surrounding melt.

669 Bubble-poor, fracture-adjacent basaltic glass could indicate diffusive volatile loss into the void-space. 670 If a low pressure void opens within magma, it will allow volatile escape, drawing H_2O out of the melt 671 through diffusion. Subsequent decompression of the melt during ascent, will then yield lower vesicularities than surrounding melt which has not experienced diffusive loss (Saubin et al., 2016; 672 673 Webb et al., 2017). Furthermore, the vesicularity and bubble size of particles within the Figure 4b 674 fracture are extremely similar to the glass adjacent to the fracture. This further supports the notion of diffusive loss and suggests that the glass within and immediately surrounding the fracture was able to 675 676 diffuse to a similar H₂O concentration which was lower than the host melt, prior to the final 677 vesiculation event.

678 Using 1D diffusion calculations to quantify the development of an 80 μ m wide H₂O depleted zone 679 (Appendix 3), we estimate that the fracture shown in Figure 4b experienced volatile loss by diffusion 680 towards the melt-fracture interface for ~40 seconds. This particular fracture has been extensively 681 healed. We therefore infer that the pre-quenching lifetime of other less-sintered fractures (e.g. Figs. 682 4a, 5a, 10) was much shorter. Our diffusion timescales are therefore close to timescale estimates for 683 sintering and residence times of inter-void particles discussed in the previous section. Nevertheless, 684 there is clearly a range of sintering and quenching timescales preserved within those clasts, as 685 expected in fragmented magma with a wide range of grainsizes and textures.

686 4.7 The nature of the void spaces

687 Small particles occupy many different void spaces e.g. angular fractures (Fig. 2), rounded fractures 688 (Fig. 3), tuffisite vein-like features (Fig. 4, 5a), bubbles and bubble chains (Figs. 5b, 6, 7) and the 689 boundary space between sintered clasts (Figs. 9, 10). At times, the sintering is so extensive that it is 690 difficult to deduce the original void shape (Fig. 9c). For instance, some features could be interpreted as a vein-like fracture (Fig. 4a) or a boundary zone between clasts (Figs. 10d, 10e). Nevertheless, there 691 692 is textural and compositional evidence for gas and particle transfer in fractures, bubble chains and 693 sintered breccias (often in the same clast; Figures 2a, 10a), and in reality, all are likely to occur, as they do in rhyolitic magma (Stasiuk et al., 1996; Schipper et al., 2013). 694

We propose a hypothetical model where localised explosions fragment both the country rock and some of the juvenile material, which are jetted into fractures, vesicles and mobile breccias, either moments before or during ejection from the vent (Fig. 15).



Figure 15: Our interpretative model to explain the various textures seen within the K1918 clasts. In (a)
a vesicular basaltic melt (pale grey) is rising next to older silicic country rock (dark grey) from past Katla
eruptions. A small localised explosion, either caused by magmatic fragmentation, or more likely

702 phreatomagmatic fragmentation from H_2O percolating down through cracked country rock, shatters both the silicic country rock and the neighbouring basaltic melt. This creates a hot mobile phase of 703 704 particles which begin to sinter together (b), infiltrate chains of connected bubbles (c) and infiltrate the 705 transient fractures formed by the explosion (d). The melt surrounding the blast zone has been 706 shattered (e) representing an incomplete version of the fragmentation within the blast zone. These 707 features, would have only existed for, at most, seconds before thorough welding took place. 708 Preservation requires further fragmentation to expel and quench the clasts into a water-rich 709 environment, which was perhaps facilitated by meltwater percolating down through cracks in the 710 damaged magma and wall rock. Parts b, c, d and e represent Figures 10f, 6a, 5a and 13 respectively.

711

712 4.8 Comparisons with rhyolite-hosted tuffisite veins

Tuffisite veins are the quenched remains of fractures that form in silicic melts, which allowed a pathway for gas and ash particle transport. As described in the previous section, it is unclear whether some of the K1918 textural features represent true 'veins', bubble chains or inter-clast spaces; we appear to have a spectrum of void spaces that include all end-members. Nevertheless, these samples clearly show evidence of permeable pathways for gas and ash particle transport, that heal shut with time in repeated fragmentation cycles (Fig. 15) analogous to tuffisite veins.

719 The circulation of gas through a permeable network is clear from S-rich microlite chains (Fig. A1) and 720 gradients in bubble textures surrounding voids (Fig. 4b). The mobilisation of particles is demonstrated 721 by the systematic distribution of particles within clasts (Figs. 2c, 6a, 9a) as well as bedding within 722 bubbles (Fig. 5b), which was also reported in rhyolitic tuffisites from Torfajökull (Tuffen et al., 2003). 723 Furthermore, foreign particles (silicic particles within basalt (Figs. 5a, b and c) and sideromelane 724 particles within tachylite (Fig. 5b)) must have been transported to their current location. Transport 725 clearly happened at magmatic temperatures because numerous particles show evidence of 726 rounding/sintering, which is also a feature of rhyolitic tuffisites (Tuffen and Dingwell, 2005).

727 The inter-void particle size in the K1918 clasts (10-200 µm diameter) is similar to those found in 728 tuffisite veins and, like rhyolitic tuffisites, angular fragments dominate over rounded ones (Tuffen et 729 al., 2003; Tuffen and Dingwell, 2005). It is difficult to compare the size of the fractures themselves. In 730 rhyolite tuffisite veins can be > 5 m (Tuffen et al., 2003) or < 100 µm (Saubin et al., 2016). The fractures 731 we observed tended to be tens of μm in width (Figs. 4 and 5). We suspect that pathways were once 732 bigger and have narrowed through welding processes (section 4.2) but it is also worth noting that our 733 observations were limited to small particles (<16,000 µm). This begs the question; were the permeable 734 pathways in the K1918 basalt limited to a small size, or did significantly larger fractures exist, and have been destroyed by the explosive nature of this eruption? Indeed the sintered breccia in Figure 10 could 735 736 be evidence for a larger scaled feature, as are the xenoliths trapped within clasts (e.g. Fig. 9). Perhaps 737 the whole conduit was acting as a permeable pathway for fragmentation and sintering.

738However, there is one clear difference; the permeable pathways in the K1918 basalt seem to be much739more transient than rhyolitic tuffisites. Thermal (Appendix 2) and diffusion (Appendix 3) estimates740suggest that the K1918 fractures were open for just seconds before quenching, whereas inferred741tuffisite vein lifetimes in rhyolite span ~10³-10⁵ seconds (Castro et al., 2012). This difference can be742explained by the lower melt viscosities and higher H₂O diffusivities that will occur in the hotter basaltic

system. Interestingly, however, diffusion distances are similar; typically tens to a few hundred μm
(Castro et al., 2012; Berlo et al., 2013; Saubin et al., 2016).

745 4.9 Links with eruptive behaviour

746 In silicic systems, permeable pathways in magma (whether connected bubbles or tuffisite veins) often 747 serve to enhance magma degassing and outgassing, and may contribute to a transition towards more 748 effusive activity (Jaupart and Allègre, 1991; Jaupart, 1998). Permeable outgassing is also thought to 749 occur during subglacial volcanism and contribute towards transitions in eruptive behaviour (Owen et 750 al., 2013a; Owen et al., 2013b; Owen, 2016). Whilst the K1918 samples show plenty of visual evidence 751 for permeable pathways (Figs. 2-10), evidence of an effusive phase is lacking from the 1918 A.D. Katla 752 eruption, although deposits from such a phase could be obscured under the present glacier (Owen, 753 2016). Nevertheless, the clasts collected were small fragments from the large-volume jökulhlaup 754 deposit that originated from explosive fragmentation. In rhyolitic melts, tuffisite veins alone are 755 thought to be inefficient at degassing and outgassing magma, with spacing of < 1mm required to 756 outgas the magma sufficiently to cause the transition to effusive behaviour (Castro et al., 2012). 757 Therefore, it is possible that the magmatic fractures observed within the K1918 samples were simply 758 insufficient to degas the magma efficiently. However, tuffisite veins that intersect permeable foams 759 are considered a highly efficient mechanism of magma degassing, and may induce a transition to 760 effusive behaviour, e.g. at Chaitén (Castro et al., 2012; Saubin et al., 2016). A large proportion of 761 bubbles within the K1918 samples contain ash particles. Therefore, gas was presumably transported 762 in connected bubbles as well as fractures (e.g. Fig. 6). If exsolved volatiles were able to efficiently 763 outgas why was the eruptive style so explosive?

One explanation could be the extremely transient nature of the permeable pathways in the basalt.
Simple calculations to estimate sintering (Appendix 2) and diffusion (Appendix 3) timescales suggest
vein lifetimes of seconds or less, approximately five orders of magnitude shorter than in rhyolitic
systems (Fig. A5) (Castro et al., 2012). However, diffusion in basalt would have also been fast, and our
inferred diffusion profile is of similar length scale to those seen in rhyolite (Castro et al., 2012).

769 An alternative model is that the permeable pathways (fractures and connected bubbles), in the K1918 770 magma, potentially served as mechanisms for enhancing explosivity rather than to defuse the 771 eruption. There are two explanations for this: (1) the pathways connected shallow magma to that of 772 a deeper source, allowing gas to be transported up from depth and thus adding to, rather than 773 reducing the volatile content of the fragmenting magma (Houghton and Gonnermann, 2008; Castro 774 et al., 2012); (2) the pathways allowed meltwater (from the overlying melting glacier) to efficiently 775 infiltrate into the magma column, facilitating fuel coolant interaction (FCI) and thus explosive 776 fragmentation.

The former, has been used to explain periods of heightened activity at Stromboli where open system degassing brings high levels of CO₂ from a deep magma source (Allard, 2010). However, if this was the case, one would expect volatiles to diffuse from the fracture into the melt and thus for the magma adjacent to fractures to be volatile-rich compared to the surrounding melt. We saw no evidence for this in our samples. In fact, if anything the opposite is true, as there is an apparent bubble-poor zone around the fracture in Figure 4b. 1783 It is more likely that the permeable pathways served to enhance fragmentation by meltwater 1784 infiltration from the overlying glacier triggering FCIs. Abundant meltwater would have been present 1785 at the time of the eruption and there is evidence that the jökulhlaup samples quenched under loading 1786 from water/ice (section 3.7). The microlite-poor nature of the glass also suggests rapid cooling.

787 We therefore propose the following hypothesis to explain the sintered permeable pathways (Fig. 15): 788 1) rising vesicular magma melted overlying ice producing water; 2) As the magma neared the surface 789 it began to fragment (Fig. 15a). It is difficult to underpin the initial cause of the fragmentation; some 790 of the clasts are highly vesicular with high bubble number densities which could indicate magmatic 791 fragmentation but there would have also been abundant meltwater which could have filtered down 792 to the magma through cracks triggering phreatomagmatic fragmentation. Nevertheless, this fragmentation event did not expel the melt fragments but instead injected them (Fig. 15b) into 793 794 connected bubbles (Fig. 15c) and fractures (Fig. 15d) in the remaining intact melt. The blast also 795 fragmented country rock which also got incorporated into these permeable pathways; 3) the hot 796 magma induced sintering of the particles and some of the pathways partially or fully healed; 4) only 797 seconds (or fractions of seconds) later, a further explosive event expelled the now fully fragmented 798 magma out of the vent into a watery environment inducing rapid quenching. The cause of this 799 fragmentation event was likely meltwater infiltrating the now highly fragmented magma through the 800 permeable pathways created by the first fragmentation event; 5) this explosion likely produced more 801 small particles and opened further fractures, allowing more meltwater to infiltrate the conduit in a 802 self-fuelling and repeating process with a cycle of seconds or less.

803 Repeating fragmentation events (Fig. 15) not only explain the particle filled void spaces but the 804 apparent clasts welded within clasts (e.g. Fig. 9). Our model (Fig. 15) could also help to explain why 805 the K1918 eruption was so powerful; magma-water mixing massively promotes fuel coolant 806 interactions (Zimanowski et al., 1991; Morrissey et al., 2000), and meltwater ingressed within 807 permeable pathways would have allowed plenty of opportunity for this. It could also explain why the 808 glacier melted so quickly and produced such a powerful flood; if the under-side of the glacier is constantly being bombarded with a hot slurry of fine particles there will be both mechanical erosion 809 810 as well as rapid thermal melting from the large-surface area to volume ratio.

Finally, our model (Fig. 15) may help to explain why the sintering textures have been preserved so 811 well, particularly in the jökulhlaup clasts; abundant meltwater, perhaps also within the permeable 812 813 pathways would promote rapid quenching. The jökulhlaup samples, unlike the air-fall samples, also 814 show that they quenched under elevated pressure (section 3.7), which could perhaps be explained by 815 being emplaced into water, which would ensure the continued quenching of the particles and the best 816 preservation of features. Note that although there is evidence of sintered particles within the air-fall 817 samples, all of the best examples of particles sintered within permeable pathways were found in the 818 jökulhlaup clasts.

4.10 The extent of the permeable network within Katla 1918 pyroclasts

Sintered particles perhaps present the best evidence for clast transportation around the glass transition and therefore the presence of a permeable network within the K1918 magma. Although sintering is present in 80% of the K1918 clasts examined, rounded particles only make up a very small percentage of the bubble and fracture-filling particles, which are predominantly angular (e.g. Figs. 5,6). When particles are angular, infilling could be thought of as an artefact of poor thin section preparation (e.g. lack of sufficient ultrasonic cleaning to rid the sample surface of adhering ash). However, the layered structures and exotic compositions, in Figure 5b, consistent with older silicic Katla magma (Fig. 12 and section 4.2) emphatically indicate that these are primary textures. As the particles in Figure 5b are almost exclusively angular, this conclusion can potentially be applied to all infilled bubbles and fractures (regardless of particle morphology) which are found in a significant proportion of all K1918 clasts (e.g. Fig. 9a).

Similarly, it is easy to quickly dismiss angular fractures as a feature formed through quenching or
 sample damage during thin section preparation. However, even the angular fractures contain particles
 favourably over nearby isolated bubbles (Figs. 2c, 3b, 6) suggestive of permeable pathways.

Thus we believe that the vast majority of the particles, fractures and connected bubbles seen in the K1918 pyroclasts to be primary features indicative of the transportation of small particles of juvenile and lithic material in a mobile fluid phase through a permeable network within the 1918 Katla magma.

837 4.11 How common are permeable gas and ash transporting pathways within basalt?

838 Although sintered particles within permeable pathways are common in rhyolite, they are thought to be exclusive to silicic eruptions, yet particle sintering was found in 80% of the basaltic K1918 clasts 839 840 examined. There are three explanations for this absence from the literature: 1) the process that forms 841 them is extremely rare but there were some conditions specific to the K1918 eruption that allowed 842 them to be formed; 2) they are commonly created during basaltic eruptions but rarely preserved; near 843 simultaneous fragmentation and quenching during K1918 allowed this; 3) they are common features, 844 preserved readily in basaltic samples, but often overlooked, perhaps when observed, mistaken for bad 845 sample preparation.

We are inclined to think that the latter option is the most likely, based on the high abundance of sintered/transported particles and permeable pathways within our samples (section 4.10). These features are difficult to detect in conventional forms of observation (e.g. Fig. 4), requiring high resolution analytical instruments (e.g. Field-emission microprobes or SEMs) for imaging and compositional mapping. Most of the features were not obvious or else completely invisible in ppl (e.g. Fig. 4aii).

Angular bubble-occupying particles are fairly common in natural volcanic samples, as are flowbands. Careful image comparison has revealed that in the K1918 clasts, flow bands in ppl images tend to represent microlite chains in BSE images (Figs. 7,10, 16), which in turn we infer to represent healed void-spaces (e.g. Figs. 5,8,10,16). It therefore seems, at least in K1918 samples, that extensive flowbanding represents (partially) healed fractures which may be filled with sintered particles (e.g. Fig. 16).



Figure 16: A clast bearing a particle filled sintered fracture which is inconspicuous in ppl, instead appearing as an area of intensive flow-banding. (a) A ppl overview of jökulhlaup clast Mul 6 unit 3a-1

2,000-4,000 3a (~3 x 3 mm). The clast is predominantly sideromelane but has two dark horizontal stripes of intensive flowbanding dissecting the sample; (b) Detail of the topmost dark stripe (see Figure 5a for a compositional map of the same area); (bi) a BSE image showing a partially healed fracture full of sintered particles, surrounded by microlite chains; (bii) a ppl image of the same area showing flow bands around a feature which is much less obviously a particle filled sintered fracture.

The features described in this paper are extremely widespread; evidence of particle sintering was present in most clasts, with particularly good examples of fracture and bubble-filled particles present in jökulhlaup samples. It would be scientifically useful to know whether such ash filled magmatic fractures and bubbles exist in other basaltic magmas; and if so whether they are exclusive to subglacial settings. Are they features which are actually common and have simply been overlooked, being merely attributed to bad sample preparation and/or thought to show flowbanding? Or are they actually a rare feature, uniquely preserved by the K1918 eruption?

873 5. Conclusions

We use textures and compositions to infer that fractures and connected bubbles acted as pathways for gas and ash to be transported within a basaltic melt. Evidence appears primarily in the form of angular to healed fractures, sintered ash particles, differing compositions between the ash particles and host, apparent sorting of particles, S precipitation and a zone of low vesicularity surrounding a partially healed fracture. Silicic particles and mineral fragments that belong to a more evolved melt suggest that material was incorporated from the country rock, and that both juvenile and lithic particles were transported through these systems, some of which have partly healed.

There are some similarities between these observed features and rhyolitic tuffisite veins. However, by comparison, the pathways seem considerably smaller and more transient suggesting extremely rapid, near simultaneous and successive episodes of brittle-ductile-brittle deformation. These observations are significant, since it was previously thought that magmatic fractures and permeable gas/ash networks only formed in high viscosity melts. The presence of such features at Katla could be explained by rapid quenching which has allowed the preservation of such features.

887 This discovery challenges our conceptions of magma degassing, fluid and particle transport, and the rheological properties of basaltic magma. In rhyolitic melts, tuffisite veins can cause a transition to 888 889 more effusive activity. The fractures within the Katla basalt do not appear to have significantly 890 degassed the magma as the clasts are still highly vesicular and erupted explosively. An alternative 891 explanation is that the fractures served to enhance explosivity by providing pathways by which 892 meltwater could infiltrate, enhancing both quenching and phreatomagmatic fragmentation. The 893 discovery of these fractures therefore could have important implications for our understanding of the 894 way in which basalt fragments.

We propose similar textures could be widespread in basaltic tephras from other settings but have
been largely ignored to date as dismissed as sample preparation artefacts. However, these textures
likely record key phases of magma damage, recycling and preparation for fuel coolant interactions.

898

899 6. Acknowledgements.

900 Jacqueline Owen is the beneficiary of a post-doctoral grant from the AXA Research Fund. Thomas 901 Shea and Hugh Tuffen were supported by a National Science Foundation grant EAR 1250366 and a 902 Royal Society University Research Fellowship respectively. Chris Hayward and Debbie Hurst assisted 903 greatly in lab analysis. Robert Duller, Lionel Wilson and Margherita Polacci contributed to fascinating 904 discussions as did a great number of people from the University of Iceland, including Magnús 905 Guðmundsson and William Moreland. We are grateful for the thoughtful and thorough edits 906 provided by Jérémie Vasseur and an anonymous reviewer. Final thanks go to James Gardner for 907 professional editing and insightful comments.

908

909 APPENDIX

910 Appendix 1: Compositional maps

911 As described in the methods, EPMA was used to make compositional (x-ray distribution) maps. Five 912 spectrometers were used to measure the relative abundance of S, Ca, Fe, K and either Na or F. The 913 data was used to make single 2D intensity matrices as shown in Figures A1, A2 and A3. These images 914 were then combined to create the RGB composite image shown in Figure 5. The relative abundance 915 of all five elements was considered when making compositional interpretations as described in the 916 text.

917



- 919 A1: 2D single intensity matrices for the fracture shown in Figure 5a. Each image represents the
- relative abundance of a single element (Na, Fe, S, K or Ca) as indicated by the chemical symbol in thetop left corner of each image. Hot and cold colours represent high and low abundances respectively.



Figure A2: 2D single intensity matrices for the particle filled bubble shown in Figure 5b. Each image 924

- 925 represents the relative abundance of a single element (Na, Fe, S, K or Ca) as indicated by the
- 926 chemical symbol in the top left corner of each image. Cold and hot colours represent low and high
- 927 abundances respectively.



930 Figure A3: 2D single intensity matrices for the area of intensive microlite chains shown in Figure 5c.

Bach image represents the relative abundance of a single element (F, Fe, S, K or Ca) as indicated by
the chemical symbol in the top left corner of each image. Cold and hot colours represent low and
high abundances respectively.

934 Appendix 2: Thermal calculations to estimate sintering timescales

935

To estimate sintering timescales we assume a three step process: (1) thermal equilibration, (2) clastrounding, (3) welding to 'healed' state.

938 The fact that so many of the particles are angular, suggests brittle fragmentation and therefore that

939 the magma was below Tg. Furthermore, we hypothesise that the more evolved particles represent

940 older silicic Katla material, which was incorporated into permeable pathways during magma accent,

941 and therefore presumably entered the system at temperatures closer to ambient. Therefore, for

942these particles to sinter, the first step will be for them to gain magmatic temperature i.e. reach943thermal equilibrium. The timescale for thermal equilibrium to be reached (t_{eq} in s) can be estimated

 t_{eq}

944 using this equation (Wilson and Mouginis-Mark, 2003)

$$\sim \frac{r^2}{k}$$

946

945

(3)

947 where r is particle radius (in m) and κ is thermal diffusivity (in m² s⁻¹). Thermal diffusivity varies (0.5-948 2.0 x 10⁻⁶ m² s⁻¹) as a function of temperature and to a lesser effect composition (Vosteen and

949 Schellschmidt, 2003; Whittington et al., 2009; Eppelbaum et al., 2014), however, in the interests of

950 simplicity we will take an ~average value of 1 x 10⁻⁶ m² s⁻¹ which is considered the general thermal

951 diffusivity for all silicates (Wilson and Mouginis-Mark, 2003)





953

954 Figure A4: Expected times for particles to meet thermal equilibrium as a function of particle size

Typical intra void particles are <100 μm and therefore would have reached thermal equilibrium in a
 matter of milliseconds (Fig. A4). We therefore, deem this process insignificant in effecting the

957 timescale of sintering.

958 Many papers discuss the timescales of sintering e.g. (e.g. Uhlmann et al., 1975; Ristić and Milosević, 959 2006; Pope, 2015), however, these models assume that the starting media consists of spherical 960 particles. Natural ash particles are not spherical. It seems from our samples that some of the ash 961 particles have experienced rounding, whereas others have not, therefore, it would be interesting to 962 estimate the timescale of this rounding/relaxation process that occurs prior to sintering. We are only 963 aware of one study (Pope, 2015) that has tried to estimate rounding rates of natural samples however, this was performed on rhyolitic particles from Cordon Caulle, Chile, and no general 964 965 relationships were observed between rounding rate, grain size and temperature. We therefore 966 turned to the equation used to estimate the rounding rate of vesicles within magma (Gardner et al., 967 2017):

$$\lambda =$$

 $\frac{\eta r}{\sigma}$

969

970 where λ is the relaxation time (in s) that a non-spherical bubble will take to relax into a spherical

971 form, η is melt viscosity (in Pa s), r is particle radius (in m) and σ is melt surface tension (in N m⁻¹).

972 When this equation was applied to the parameters of the particles used by Pope (2015) the

calculated relaxation time was very similar to the timescales of clast rounding observed in their
experiments. Therefore, we will use equation (4) as a proxy for the relaxation/rounding rate of Katla
clasts.

976 Viscosities were estimated using the model of Giordano et al. (2008) our EPMA data and the

 $\,$ 977 $\,$ assumption that the magma was 1045 °C. We also modelled typical rhyolite from Chaitén at 800 °C $\,$

978 (Castro and Dingwell, 2009), where tuffisite veins and sintering are common (Castro et al., 2012;

Berlo et al., 2013; Saubin et al., 2016). Although surface tension (σ) varies as a function of various
 magmatic parameters (Bagdassarov et al., 2000; Mangan and Sisson, 2005; Gardner and Ketcham,

magmatic parameters (Bagdassarov et al., 2000; Mangan and Sisson, 2005; Gardner and Ketcham,
2011; Gardner et al., 2013), H₂O content is the only parameter considered to have a significant

effect, with melt composition and temperature only playing a very minor role (Walker and Mullins,

1983 1981; Bagdassarov et al., 2000; Gardner et al., 2013; Gardner et al., 2017). We therefore chose a

single value of 0.3 N m⁻¹ for all modelling which is consistent with literature values for relatively dry

985 silicate melts (Taniguchi, 1988; Phillips et al., 1995; Gardner and Denis, 2004; Sumner et al., 2005).

986





988

(4)

989 Figure A5: expected times for particles to round/sinter as a function of particle size and dissolved

 $990 \qquad H_2O \ content \ (0.1 \ -2.0 \ wt.\% \ as \ shown \ in \ the \ legend). \ Note \ that \ the \ rounding \ timescale \ and$

991 sinetering/healing timescale were calculated in the same way and therefore have the same value.

992 Therefore, the combined rounding and sintering rate can be found by simply doubling the values in

this figure. Green and blue lines refer to samples within this study at an assumed temperature of
1045 °C. The orange lines model Chaiten rhyolite at 800 °C (using composition data from Castro and

995 Dingwell (2009)) where tuffisite veins are often found.

996

997Once connections are made between particles, the pore spaces can 'heal' through viscous flow998(Ristić and Milosević, 2006; Vasseur et al., 2013). The following equation can be used to estimate the999timescale τ_s of viscous sintering (Vasseur et al., 2013)

 $\tau_{s=\frac{R_i\eta}{\gamma}}$

1000

1001

(5)

where R_i is the initial radius, η is melt viscosity and γ is the melt-vapour interfacial tension. Note that
this is essentially the same equation as that for the viscous relaxation of bubbles (Vasseur et al.,
2013). As we are using the latter to estimate the rounding rate of the particles, and because we
disregard the timescales for thermal equilibrium as being insignificantly small, then our combined
timescale for the complete sintering of the sub millimetric 1918 A.D. Katla particles to become
healed glass can be estimated by doubling equation (4) or (5) and thus the values in Figure A5.

1008 Typically, inter-void particles are approximately \leq 100 μ m in diameter. For basaltic particles with 0.2-1009 0.3 wt.% H_2O (as measured with FTIR) this equates to rounding times of ≤ 0.1 seconds, and thus full 1010 healing times of approximately ≤ 0.2 seconds. These resultant times, are perhaps a little 1011 unrealistically short. However, they give an idea of relative differences between particles e.g. it can 1012 be seen that silicic particles of the same size will take considerably longer to sinter (Figs. A5, A6). 1013 Furthermore, even if the absolute values were out by a few orders of magnitude (it is difficult to 1014 suggest reasons for errors larger than this), then it still equates to extremely short residence times 1015 for the permeable networks and times much shorter than for Chaitén rhyolite at 800 °C (Fig. A5) 1016 (Castro et al., 2012) where our estimated values closely match observations (Pope, 2015). Thus we 1017 can be confident that our estimates are reasonable ball park amounts.



Figure A6: The compositional map from Figure 5a, with estimated residence times (in seconds),
assuming low H₂O concentrations (consistent with observations) shown in red.

1022 The thermal estimations can help explain some of the particle morphologies as well as shed insights 1023 into timescales. For example, our calculations suggest the large rounded basaltic particle in Figure A6, had to be hot for at least 0.13 seconds to be deformable, however if it had been at 1045 °C for 1024 1025 more than 0.26 seconds it would have completely annealed with the surrounding melt. The 1026 angular silicic particle (blue) next to it required ~8 seconds at 1045 °C to relax into a sphere. The fact 1027 that it has not rounded, therefore suggests a residence time < 8 seconds. The smaller rounded basaltic particles require residence times of 0.05-0.1 seconds, whilst the angular clasts must have 1028 1029 been resident for <0.05 seconds. If we assume that the microlite chains (in pink above the fracture) 1030 represent the former bounderies of annealed particles then using their approximate dimensions, 1031 these particles must have been present for more than >0.14 seconds to anneal together. All of this is 1032 consistent with a fracture opening for a fraction of a second, and having a momentary stream of 1033 particles before quenching. However, it is impossible to tell the extent of the annealed particles 1034 bejond the pink microlites (presumably prolonged heating will completely homogenise the melt), 1035 therefore a more conservative esimate for the lifetime of each permeable network would be on the 1036 order of a second or so.

1037 We estimate that the fracture in Figure 5a was open for a slightly longer duration than the
 1038 connected bubbles in Figure 5b where nearly all of the particles are incredibly small (~5 μm) and
 1039 angual. Our thermal calculations suggest that these particles could not have been resident for > 0.05
 1040 seconds suggesting a near instanenous injection and quenching process.

1041 For Figure 5c, we estimate that the network was permeable for ~ 0.05-0.4 seconds. The first number 1042 represents the time taken to completely sinter and heal basalt, and the latter is the time required to 1043 round dacite; both assuming a particle size of ~5 μ m which seems to be the approximate average 1044 size of the remaining silicic particles.

1045

1046 Appendix 3 - Diffusion estimates

1047 Bubble-poor zones next to voids can be interpreted to show areas of diffusive volatile loss into the 1048 void-space (Saubin et al., 2016; Webb et al., 2017). The void in Figure 4b shows a clear zone of bubble-1049 poor glass extending approximately equidistant from the void wall. The average width of the bubblepoor zone is ~80 μ m, which can be inferred to represent the volatile diffusion distance. The fact that 1050 1051 these fractures have been preserved, suggests that they formed at a relatively shallow level in the 1052 conduit, thus is it likely that H_2O will be the predominant species that is diffusing. The diffusivity of H_2O (D) in m²s⁻¹ for a basaltic melt of 0.2 wt.% H_2O (a reasonable assumption given the measured FTIR 1053 1054 concentrations, and assuming our interpretation of shallow level is correct) can be expressed with 1055 the following equation from Zhang and Stolper (1991):

 $lnD = -(12.49 \pm 2.35) - (15200 \pm 3900)/T$

(6)

(7)

where T is temperature in Kelvin, inferred to be 1318 K (1045 °C) based on our oxide thermometry
results. Assuming that the opening of a fracture results in disequilibrium within the melt, the H₂O
diffusion distance (L) in m, is roughly approximated by the following equation from Zhang and Stolper
(1991):

 $L \cong 2\sqrt{Dt}$

1064 where *t* is time in seconds. Combining equations (6) and (7), we obtain a diffusion time of ~40s for H_2O 1065 to diffuse ~80 µm into the fracture.

1066

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