GEOLOGY

E: geology@geosociety.org
W: geosociety.org/gsa
T: @geosociety

Mid-loaf crisis: Internal breadcrust surfaces in rhyolitic pyroclasts reveal dehydration quenching

Tracking no: G49959R

Authors:
Hugh Tuffen (Lancaster University), Jamie Farquharson (Independent), Fabian Wadsworth (Durham University), Cameron Webb, Jacqueline Owen (Lancaster University), Jonathan Castro (University of Mainz), Kim Berlo, C. Schipper (Victoria University of Wellington), and Katia Wehbe (Diamond Light Source Ltd)

Abstract:
Breadcrust bombs are pyroclasts displaying fractured, dense surfaces enveloping expanded interiors, associated with Vulcanian explosions. Here, we document pyroclasts from the 2008-2009 eruption of Chaitén (Chile) that are internally as well as externally breadcrusted. The pyroclasts are cut by intersecting µm-mm thick tuffisites with dense glassy walls, which grade into strongly inflated pumiceous material. We find H2O diffusion gradients proximal to the breadcrusted surfaces, such that H2O is depleted from far-field magma (0.68{plus minus}0.04 wt.%) into dense, fractured vein walls (0.2-0.3 wt.%), indicating a spatial association between H2O mass transfer within the pyroclast interior and both suppressed vesiculation and breadcrusting. We experimentally confirm that diffusive H2O depletion suppresses bubble growth at shallow conduit conditions. Therefore, we interpret the breadcrust formation to be induced by H2O diffusion and the associated rise in viscosity, rather than by cooling in the classical breadcrust-formation models. We posit that a 'dehydration quench' is important as degassing continues to very low H2O contents in shallow conduit magma that continues to vesiculate.

Mid-loaf crisis: Internal breadcrust surfaces in rhyolitic pyroclasts reveal dehydration quenching

Hugh Tuffen[1], Jamie I. Farquharson[1], Fabian B. Wadsworth[2,] Cameron Webb[1,3], Jacqueline Owen[1], Jonathan M. Castro[4], Kim Berlo[5], C. Ian Schipper[6], and Katia Wehbe[7]

[1]Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, UK

[2]Department of Earth Sciences, Durham University, Durham, DH1 3LE, UK

[3]Environment Agency, Rotherham, S60 1BY, UK

[4]Institute of Geosciences, Johannes Gutenberg Universität, Mainz, D-55128, Germany

[5]Department of Earth & Planetary Sciences, McGill University, Montreal, H3A 0E8, Canada

[6]School of Geography, Environment and Earth Sciences, Victoria University of Wellington, Wellington 6012, NZ

[7]B22: MIRIAM, Diamond Light Source Ltd, Oxford, OX11 0DE, UK

ABSTRACT

Breadcrust bombs are pyroclasts displaying fractured, dense surfaces enveloping expanded interiors, associated with Vulcanian explosions. Here, we document pyroclasts from the 2008–2009 eruption of Chaitén (Chile) that are internally as well as externally breadcrusted. The pyroclasts are cut by intersecting µm–mm thick tuffisites with dense glassy walls, which grade into strongly inflated pumiceous material. We find H_2O diffusion gradients proximal to the breadcrusted surfaces, such that H_2O is depleted from far-field magma (0.68 ± 0.04 wt.%) into dense, fractured vein walls (0.2–0.3 wt.%), indicating a spatial association between H_2O mass transfer within the pyroclast interior and both suppressed vesiculation and breadcrusting. We experimentally confirm that diffusive H_2O depletion suppresses bubble growth at shallow conduit conditions. Therefore, we interpret the breadcrust formation to be induced by H_2O diffusion and the associated rise in viscosity, rather than by cooling in the classical breadcrust-formation models. We posit that a 'dehydration quench' is important as degassing continues to very low H_2O contents in shallow conduit magma that continues to vesiculate.

INTRODUCTION

Breadcrust bombs are ballistic pyroclasts ejected explosively during Vulcanian events (e.g. Self et al., 1979), characterized by dense, cracked, and quenched exteriors that grade into comparatively vesicular interiors (Johnston-Lavis, 1888; Wright et al., 2007). The cracking indicates rheological surface stiffening preceding the cessation of interior expansion, conventionally attributed to relatively rapid thermal quenching of outer surfaces in air or water (Walker, 1969; Fisher and Schmincke, 1984) while slower-cooled interiors undergo decompression-triggered, post-fragmentation vesiculation (e.g Wright et al., 2007). Bomb textures and volatile concentrations record pre-blast conduit conditions (Wright et al., 2007; Giachetti et al., 2010): to vesiculate during bomb ejection, magma must exceed an H_2O supersaturation threshold, typically in the range 0.4–0.9 wt.% (Hoblitt and Harmon 1993; Wright et al., 2007; Bain et al., 2019). However, recently identified pyroclast breadcrust textures (Quane and Andrews, 2020) suggests that breadcrust formation may often be more complex than this general model.

Recent rhyolitic eruptions at Chaitén and Cordón Caulle (Chile) involved hybrid explosive–effusive activity from a common vent above a compacting lava plug (Pallister et al., 2013; Schipper et al., 2013). Explosive upper conduit fracturing and tuffisite formation facilitated transient outgassing (Castro et al., 2012, 2014; Saubin et al., 2016; Paisley et al., 2019; Schipper et al., 2021), allowing high-flux effusion of dense rhyolite (e.g. Wadsworth et al., 2020), until welding and healing rendered them impermeable (Farquharson et al., 2017; Heap et al., 2019; Wadsworth et al., 2021). Characterization of tuffisite veins and breadcrust bombs is yet to be integrated, despite common causes (gas overpressure), process implications (explosive plug disruption), and resultant products (partially vesiculated, vein-hosting, glassy pyroclasts). Here, we document pumiceous, tuffisite-bearing rhyolitic pyroclasts from the 2008–2009 Chaitén eruption, which exhibit an interconnected network of internal and external breadcrusted surfaces.

GEOLOGICAL CONTEXT

The 2008–2009 eruption of Chaitén (~42°50'S; Chilean Patagonia: **Fig 1**) began on 1 May 2008 following 24 hr of seismicity (Castro and Dingwell, 2009), with dyke-driven ascent of near-aphyric, high-silica rhyolite from a 5–10 km-deep source region (Pallister et al., 2013). Activity spanned phases 1 (explosive, 10 days), 2 (hybrid activity, 20 days), and 3 (effusive lava emplacement, several months). Column-collapse pyroclastic density currents (PDCs) were emplaced within the caldera late in phase 1; phase 2 involved eruption of rhyolitic lava domes and coulées with synchronous, repeated Vulcanian explosions and intermittent PDC-producing plumes (Pallister et al., 2013), with deposits emplaced on the caldera floor (**Fig 1**).

TEXTURAL DESCRIPTION

Here we describe an irregular pyroclast (15×25×20 cm) (**Fig 2**), which is typical of many deposits emplaced during the latter part of phase 1, including bombs and bomb fragments. The outer surface of the pyroclast is variably coated with centimetric red- or brown-colored plates of sintered fine-grained clastic material, delineated by polygonal fractures in jigsaw-fit organization (**Fig 2**A**, 2**B**, 2E, 2F**) often interpreted as unambiguous evidence of cooling- and vesiculation-induced breadcrusting. These surface textures can be tracked into the pyroclast interior (**Figs 2**C, **2**D), which is characterized by a network of prominent mm-thick tuffisites. Viewed in cross-section, the characteristic breadcrust surface texture and the internal tuffisites are clearly manifestations of the same structures. Cross-cutting tuffisites (**Fig 2**G) indicate two separate fracture events (early- and late-stage).

Late-stage tuffisites comprise curviplanar veins filled by red-brown sintered fragments (as locally observed on the outer clast surface), flanked by ~10 µm-thick, dense obsidian selvages normal to the tuffisite plane. These dense walls grade into pumiceous material that makes up the majority of the clast interior. Early-stage tuffisites are cut by their late-stage counterparts (**Fig 2**G), and themselves cut through variably dense bands (**Fig 2C**). Early-stage tuffisites exhibit thick glassy walls (~2 mm; **Fig 2**G). The incipiently sintered early-stage tuffisite fill consists of angular to sub-rounded, fine-grained (~1–20 µm) glassy clasts (**Fig 2**H). The network of cross-cutting veins delineates discrete angular domains of high-vesicularity material (**Figs 2**C & **2**G). The clastic veneer that coats the pyroclast surface and fills late-stage tuffisites is cut by tensile fractures (**Fig 2**E**, F**)—manifest as variable segmentation in cross-section (**Fig 2G**)—while the underlying pumice has flowed viscously into the resultant voids (**Fig 2**F, **2H**), similar to textures observed in mesoscale boudinage structures in vesicular rhyolite domes (e.g., Panum dome, USA: Castro and Cashman, 1999). Dense tuffisite walls and clastic fill therefore signify distinct rheological responses to the viscous, vesicular material.

H_2O MOBILITY AND DEGASSING

Benchtop and synchrotron-source Fourier-transform infrared spectroscopy (FTIR and sFTIR, respectively) was used to quantify dissolved H_2O concentrations in transects normal to an early-stage tuffisite (**Figs 3A-C**). H_2O content is typically lower within the tuffisite than in the surrounding pyroclast (mean value of 0.31 wt.% H_2O relative to ~0.68 wt.% in the far-field: **Fig 3A**). All transects exhibit systematic H_2O decreases from far-field values ~0.68±0.04 wt.% towards the tuffisite wall (**Fig 3A, C**). Minimum measured values at the wall were 0.16 wt.% for sFTIR (**Fig 3A**) and 0.28 wt.% for FTIR (**Fig 3C**). All transects (**Fig 3**) represent atypical diffusion gradients, inasmuch as vesiculation has overprinted the diffusion process. The approximate pyroclast center porosity ϕ is 0.6, and this post-fragmentation vesiculation has modified measured H_2O concentrations via mass transfer into bubbles. Toward the dense obsidian edge, vesiculation has also dilated the spatial coordinates in the observed H_2O gradient. In the **Data Repository** we provide a quantitative correction for these effects, in order to rectify the diffusion gradient for further analysis (**Fig 3C**). In all transects, the measured H_2O depletion occurs over a lengthscale of L~300 µm, indicating diffusive outgassing into the vein. We define the diffusion timescale as $\lambda_D = L^2/(4D)$, with diffusivity $D \approx 1.3 \times 10^{-12}\ \mathrm{m^2 s^{-1}}$ (Zhang and Ni, 2010) for 825 °C. The diffusion timescale required to produce the observed gradient lengthscales is $\lambda_D = 4.81$ hours. Similarly, we apply a simplified 1D diffusion model (Crank, 1979) $c(x) = c_b + (c_0 - c_b)\mathrm{erfc}[x/(2\sqrt{Dt_D})]$, where erfc is the complementary error function and c_b is the observed value of H_2O concentration c at the wall. Using a least-squares minimization to the observed c_x profile, we compute the time t_D over which diffusion occurred. We find 1.9–6.0 hours (**Fig 3C**), consistent with other estimates for Chaitén tuffisites (Castro et al., 2012; Berlo et al., 2013; Saubin et al., 2016; Heap et al., 2019) and with the scaled timescale λ_D. Assuming far-field H_2O concentration is the pre-ejection solubility value, solubility modeling (Newman and Lowenstern 2002) predicts equilibration at 4 MPa pressure, a magmastatic depth of 170 m (similar to pyroclast depth estimates from Castro et al. (2014)).

EXPERIMENTAL VESICULATION

We heated a polished wafer (~112 µm thick) from the early-stage tuffisite margin at 200 °C min^{-1} and ambient pressure to 825 °C in a Linkam TS1500 heated stage attached to a Zeiss Axioscope 1 microscope with Pixelink camera. For full methods see Browning et al. (2020). Images were acquired to characterize the rate and spatial distribution of vesicle nucleation and growth over 2 hr (**Figs 3D–F**). H_2O concentration across the transect from dense glass to pumiceous material was also measured prior to the experiment. As in other transects (**Figs 3A & C**), the pre-experiment sample exhibited increasing H_2O with distance from the tuffisite wall. Vesiculation was entirely suppressed in the dense glassy zone with H_2O <0.5 wt.% (**Fig 3F**), with negligible additional vesiculation in the pumiceous zone (~110 µm from the wall), indicating that it was above the percolation threshold—permeable outgassing occurred (Vasseur et al., 2020). However, the medial zone was sufficiently saturated to allow exsolution and bubble nucleation, manifest in calculated bubble growth rates of ≤3.0 µm min^{-1}, highlighting textural predisposition to thermal vesiculation.

POST-TUFFISITE PYROCLAST VESICULATION AND COOLING

The pyroclast exhibits the characteristic—typical of ballistic ejecta—of highest vesicularity at its center. This reflects slower cooling and extended post-fragmentation vesiculation (e.g. Wright et al., 2007; Giachetti et al., 2010). Pyroclast cooling timescales can be scaled as $\lambda_T = R^2/(4D_T)$, where D_T is thermal diffusivity ($\approx 6 \times 10^{-7}$; Bagdassarov and Dingwell, 1994), and R is the pyroclast radius. Estimating $R = 0.15$ m, we find $\lambda_T = 2.6$ hours, which represents the time available for vesiculation spanning pyroclast decompression, ejection, and quenching. We note that $\lambda_T \approx \lambda_D$, with the lower bound $\lambda_D < \lambda_T$, demonstrating that the diffusive mass transfer of H_2O can occur during the thermal lifetime of the clast. This is consistent with the observation that vesiculation has driven fracturing (**Fig 2F**).

Breadcrust features of the late-stage tuffisite (**Fig 2F, G**) are characterized by displacements between fragments of crust. We argue that the late-stage vein and its walls were more viscous than the far-field melt due to relative volatile depletion, embrittling the magma adjacent to the vein margins. Although the tuffisite fill is clastic, it has undergone some sintering and would be capable of cohesive viscous deformation (e.g. Tuffen and Dingwell, 2005). A range of plausible viscosities η are calculated (Giordano et al. 2008), using the composition of Chaitén melt (Castro and Dingwell, 2009) and the range of measured H_2O concentrations within the sample (**Fig 4**). Assuming the total time for bubble growth $t_B = 60$ s is the time available for vesiculation-derived deformation (Browning et al., 2020), and linear strain γ of 0.25 estimated from the fragment offsets along planar portions of the vein (**Fig 2F**), we estimate a bulk linear strain rate $\dot{\gamma} = \gamma / t_B = 0.0042\ s^{-1}$. Using this $\dot{\gamma}$, we estimate a threshold viscosity for brittle melt failure: $\eta_c = \sigma_c / \dot{\gamma}_c$, where σ_c is the threshold stress for failure ($\sigma_c = 10^8$ Pa; Webb and Dingwell, 1990; Wadsworth et al., 2018). We find that $\eta_c = 2.38 \times 10^{10}$ Pa.s. In **Fig 4**, we show that a viscous deformation response to vesiculation-driven strain is expected for the non-H_2O-depleted far-field regions at ejection temperatures of >750 °C—due to their lower viscosity—allowing clast expansion. By contrast, neighboring volatile-depleted, higher-viscosity melt will fail, despite being at the same temperature, underpinning our model for degassing-driven internal breadcrusting.

CONCEPTUAL MODEL AND CONCLUSIONS

We interpret the sample as recording at least two in-conduit fracture events, periods of sintering, and vesiculation, and ejection-related decompression and cooling. We infer the following sequence (**Fig 4B**): (0) a homogenous, dense aliquot of magma was resident at ~170 m depth; (1) a fracture event occurred, forging pathways for gas/ash flow through the magma; (2) H_2O depletion occurred over the lifespan of these fractures, leaving volatile-poor—rheologically stiffer—magma adjacent to the fracture walls; (3) fracture closure was accompanied or driven by partial sintering of the fine-grained material inside; at this stage or contemporaneous with stage 4, a second generation of fractures was also created (**Fig 2E**); (4) decompression-driven vesiculation occurred with the magma adjacent to the former outgassing pathways remaining non-vesicular due to vesiculation suppression; (5) a brittle response in the mechanically coupled volatile-poor selvages and incipiently sintered tuffisite fill, which resulted in breadcrusting both within the pyroclast interior and on its outer surface. Pyroclast ejection occurred along with (4) or in an explosion shortly afterwards (**Fig 4C**). Stages (0)–(5) all occurred within the thermal lifetime of the pyroclast, pre- and syn-ejection. Both volatile depletion towards the vein and preferential vesiculation distal from tuffisites in this pyroclast led to rheological stiffening associated with degassing that depended primarily on chemical diffusion: an isothermal degassing quench (Ryan et al., 2015) rather than a thermal quench. To date, breadcrust textures in volcanic deposits have been interpreted as unambiguous evidence of a thermal quenching process. Our alternative mechanism for breadcrust formation—a chemo-rheological rather than thermo-rheological process—suggests that similar textures recorded in volcanic deposits worldwide may require reanalysis and reinterpretation. Such textures may occur in explosively ejected fragments of conduit plugs, where they have previously been uniquely interpreted as thermal quench features (i.e. classic breadcrust bombs), and also within silicic lavas, where relatively dense layers—e.g. tuffisites—have been segmented by heterogeneous vesiculation (Castro and Cashman, 1999; Andrews et al., 2021).

ACKNOWLEDGMENTS

We are grateful for financial support from the Royal Society (URF UF140716, to support HT and JF), the EC-FP7 Vuelco project (FW), and the AXA Research Fund (JO). N La Penna and R Wilbraham assisted in the field and with SEM imaging.

REFERENCES CITED

Andrews, G.D.M., Kenderes, S.M., Whittington, A.G., Isom, S.L., Brown, S.R., Pettus, H.D., Cole, B.G., and Gokey, K.J., 2021, The fold illusion: The origins and implications of ogives on silicic lavas: Earth and Planetary Science Letters, v. 553, p. 116643, doi:10.1016/j.epsl.2020.116643.

Bagdassarov, N., and Dingwell, D., 1994, Thermal properties of vesicular rhyolite: Journal of Volcanology and Geothermal Research, v. 60, p. 179–191, doi:10.1016/0377-0273(94)90067-1.

Bain, A.A., Calder, E.S., Cortés, J.A., Cortés, G.P., and Loughlin, S.C., 2019, Textural and geochemical constraints on andesitic plug emplacement prior to the 2004–2010 vulcanian explosions at Galeras volcano, Colombia: Bulletin of Volcanology, v. 81, p. 1, doi:10.1007/s00445-018-1260-y.

Berlo, K., Tuffen, H., Smith, V.C., Castro, J.M., Pyle, D.M., Mather, T.A., and Geraki, K., 2013, Element variations in rhyolitic magma resulting from gas transport: Geochimica et Cosmochimica Acta, v. 121, p. 436–451, doi:10.1016/j.gca.2013.07.032.

Browning, J., Tuffen, H., James, M.R., Owen, J., Castro, J.M., Halliwell, S., and Wehbe, K., 2020, Post-fragmentation vesiculation timescales in hydrous rhyolitic bombs from Chaitén volcano: Journal of South American Earth Sciences, v. 104, p. 102807, doi:10.1016/j.jsames.2020.102807.

Castro, J.M., Bindeman, I.N., Tuffen, H., and Ian Schipper, C., 2014, Explosive origin of silicic lava: Textural and δD–H2O evidence for pyroclastic degassing during rhyolite effusion: Earth and Planetary Science Letters, v. 405, p. 52–61, doi:10.1016/j.epsl.2014.08.012.

Castro, J., and Cashman, K.V., 1999, Constraints on rheology of obsidian lavas based on mesoscopic folds: Journal of Structural Geology, v. 21, p. 807–819, doi:10.1016/S0191-8141(99)00070-X.

Castro, J.M., Cordonnier, B., Tuffen, H., Tobin, M.J., Puskar, L., Martin, M.C., and Bechtel, H.A., 2012, The role of melt-fracture degassing in defusing explosive rhyolite eruptions at volcán Chaitén: Earth and Planetary Science Letters, v. 333–334, p. 63–69, doi:10.1016/j.epsl.2012.04.024.

Castro, J.M., and Dingwell, D.B., 2009, Rapid ascent of rhyolitic magma at Chaitén volcano, Chile: Nature, v. 461, p. 780–783, doi:10.1038/nature08458.

Crank, J., 1979, The Mathematics of Diffusion: Clarendon Press, 428 p.

Farquharson, J.I., Wadsworth, F.B., Heap, M.J., and Baud, P., 2017, Time-dependent permeability evolution in compacting volcanic fracture systems and implications for gas overpressure: Journal of Volcanology and Geothermal Research, v. 339, p. 81–97.

Fisher, R.V., and Schmincke, H.-U., 1984, Pyroclastic Rocks and Tectonic Environment, *in* Fisher, R.V. and Schmincke, H.-U. eds., Pyroclastic Rocks, Berlin, Heidelberg, Springer, p. 383–409, doi:10.1007/978-3-642-74864-6_14.

Giachetti, T., Druitt, T.H., Burgisser, A., Arbaret, L., and Galven, C., 2010, Bubble nucleation, growth and coalescence during the 1997 Vulcanian explosions of Soufrière Hills Volcano, Montserrat: Journal of Volcanology and Geothermal Research, v. 193, p. 215–231, doi:10.1016/j.jvolgeores.2010.04.001.

Giordano, D., Russell, J.K., and Dingwell, D.B., 2008, Viscosity of magmatic liquids: A model: Earth and Planetary Science Letters, v. 271, p. 123–134, doi:10.1016/j.epsl.2008.03.038.

Heap, M.J., Tuffen, H., Wadsworth, F.B., Reuschlé, T., Castro, J.M., and Schipper, C.I., 2019, The Permeability Evolution of Tuffisites and Implications for Outgassing Through Dense Rhyolitic Magma: Journal of Geophysical Research: Solid Earth, v. 124, p. 8281–8299, doi:https://doi.org/10.1029/2018JB017035.

Hoblitt, R.P., and Harmon, R.S., 1993, Bimodal Density Distribution of Cryptodome Dacite from the 1980 Eruption of Mount St. Helens, Washington: Bulletin of Volcanology, v. 55, p. 421–437, doi:10.1007/BF00302002.

Johnston-Lavis, H.J., 1888, Further Notes on the Late Eruption at Vulcano Island: Nature, v. 39, p. 109–111, doi:10.1038/039109a0.

Newman, S., and Lowenstern, J.B., 2002, VolatileCalc: a silicate melt–H2O–CO2 solution model written in Visual Basic for excel: Computers & Geosciences, v. 28, p. 597–604, doi:10.1016/S0098-3004(01)00081-4.

Paisley, R., Berlo, K., Whattam, J., Schipper, C.I., and Tuffen, H., 2019, Degassing-induced chemical heterogeneity at the 2011-2012 Cordón Caulle eruption: Volcanica, v. 2, p. 211–237, doi:10.30909/vol.02.02.211237.

Pallister, J.S., Diefenbach, A.K., Burton, W.C., Muñoz, J., Griswold, J.P., Lara, L.E., Lowenstern, J.B., and Valenzuela, C.E., 2013, The Chaitén rhyolite lava dome: Eruption sequence, lava dome volumes, rapid effusion rates and source of the rhyolite magma: Andean Geology, v. 40, p. 277–294, doi:10.5027/andgeoV40n2-a06.

Quane, S.L., and Andrews, B.J., 2020, Breadcrust bubbles: Ash particles recording post-fragmentation brittle-ductile deformation: Geology, v. 48, p. 1205–1209, doi:10.1130/G47811.1.

Ryan, A.G., Russell, J.K., Nichols, A.R.L., Hess, K.-U., and Porritt, L.A., 2015, Experiments and models on H2O retrograde solubility in volcanic systems: American Mineralogist, v. 100, p. 774–786, doi:10.2138/am-2015-5030.

Saubin, E., Tuffen, H., Gurioli, L., Owen, J., Castro, J.M., Berlo, K., McGowan, E.M., Schipper, C.I., and Wehbe, K., 2016, Conduit Dynamics in Transitional Rhyolitic Activity Recorded by Tuffisite Vein Textures from the 2008–2009 Chaitén Eruption: Frontiers in Earth Science, v. 4, doi:10.3389/feart.2016.00059.

Schipper, C.I., Castro, J.M., Kennedy, B.M., Tuffen, H., Whattam, J., Wadsworth, F.B., Paisley, R., Fitzgerald, R.H., Rhodes, E., Schaefer, L.N. and Ashwell, P.A., 2021, Silicic conduits as supersized tuffisites: Clastogenic influences on shifting eruption styles at Cordón Caulle volcano (Chile): Bulletin of Volcanology, v. 83, p. 11, doi:10.1007/s00445-020-01432-1.

Schipper, C.I., Castro, J.M., Tuffen, H., James, M.R., and How, P., 2013, Shallow vent architecture during hybrid explosive–effusive activity at Cordón Caulle (Chile, 2011–12): Evidence from direct observations and pyroclast textures: Journal of Volcanology and Geothermal Research, v. 262, p. 25–37, doi:10.1016/j.jvolgeores.2013.06.005.

Self, S., Wilson, L., and Nairn, I.A., 1979, Vulcanian eruption mechanisms: Nature, v. 277, p. 440–443, doi:10.1038/277440a0.

Tuffen, H., and Dingwell, D., 2005, Fault textures in volcanic conduits: evidence for seismic trigger mechanisms during silicic eruptions: Bulletin of Volcanology, v. 67, p. 370–387, doi:10.1007/s00445-004-0383-5.

Vasseur, J., Wadsworth, F.B., and Dingwell, D.B., 2020, Permeability of polydisperse magma foam: Geology, v. 48, p. 536–540, doi:10.1130/G47094.1.

Wadsworth, F.B., Vasseur, J., Llewellin, E.W., Brown, R.J., Tuffen, H., Gardner, J.E., Kendrick, J.E., Lavallée, Y., Dobson, K.J., Heap, M.J. and Dingwell, D.B., 2021, A model for permeability evolution during volcanic welding: Journal of Volcanology and Geothermal Research, v. 409, p. 107118, doi:10.1016/j.jvolgeores.2020.107118.

Wadsworth, F.B., Llewellin, E.W., Vasseur, J., Gardner, J.E., and Tuffen, H., 2020, Explosive-effusive volcanic eruption transitions caused by sintering: Science Advances, v. 6, p. eaba7940, doi:10.1126/sciadv.aba7940.

Wadsworth, F.B., Witcher, T., Vossen, C.E.J., Hess, K.-U., Unwin, H.E., Scheu, B., Castro, J.M., and Dingwell, D.B., 2018, Combined effusive-explosive silicic volcanism straddles the multiphase viscous-to-brittle transition: Nature Communications, v. 9, p. 4696, doi:10.1038/s41467-018-07187-w.

Walker, G.P.L., 1969, The breaking of magma: Geological Magazine, v. 106, p. 166–173, doi:10.1017/S0016756800051979.

Webb, S.L., and Dingwell, D.B., 1990, Non-Newtonian rheology of igneous melts at high stresses and strain rates: Experimental results for rhyolite, andesite, basalt, and nephelinite: Journal of Geophysical Research: Solid Earth, v. 95, p. 15695–15701, doi:10.1029/JB095iB10p15695.

Wright, H.M.N., Cashman, K.V., Rosi, M., and Cioni, R., 2007, Breadcrust bombs as indicators of Vulcanian eruption dynamics at Guagua Pichincha volcano, Ecuador: Bulletin of Volcanology, v. 69, p. 281–300, doi:10.1007/s00445-006-0073-6.

Zhang, Y., and Ni, H., 2010, Diffusion of H, C, and O Components in Silicate Melts: Reviews in Mineralogy and Geochemistry, v. 72, p. 171–225, doi:10.2138/rmg.2010.72.5.

CAPTIONS

Figure 1. Hybrid explosive–effusive activity at Chaitén. Arrow indicates sampling location. *Inset:* Location map of Chaitén (triangle). Photograph from May 26, 2008 (J. N. Marso, USGS).

Figure 2. Textures within Chaitén pyroclast. **A** Clast overview, showing red–brown, fractured plates of clastic material on surface. **B** Detail of surface breadcrust texture. **C** Overview; red–brown, fractured plates of clastic material on the surface, connected to tuffisite that cross-cuts flow bands in the clast interior (dashed box). **D** Overview; tuffisite partially exposed at clast surface. **E** Detail of **C**, showing cm-scale polygonal plates of breadcrust on the surface that comprise a clastic red veneer overlying dense grey glass. **F** Exposed tuffisite, showing separation of sintered tuffisite, indicating tensile linear strain of up to ~0.25. **G** Cross-section showing early-stage tuffisite with ~100 µm-thick dense walls, cross-cut by late-stage vein. White triangles mark late-stage tuffisite segmentation. **H** SEM image of late-stage tuffisite: poorly sintered ash adhering to a glassy surface, adjacent to vesiculated pumiceous texture (porosity shown as black).

Figure 3: **A** sFTIR transect s–s' crossing tuffisite (see **B**), with measured wt.% H_2O_T relative to the transect position. **B** Sample wafer, indicating location of FTIR and sFTIR transects (s–s', …, f–f') and wafer used in hotstage experiment. **C** FTIR-derived H_2O_T as a function of corrected distance along transect from tuffisite (see **B**, **Data Repository**) for each transect a–a', …, f–f'. Black dashed line shows best-fit diffusion profile and corresponding diffusion timescale. Grey shaded region shows envelope for timescales of 0.5 to 20 hr. **D** Pre-heating sample wafer. White dashed line indicates tuffisite wall. **E** Post-heating sample wafer. **F** Bubble growth rate as a

function of initial vesicle distance from tuffisite wall. Porosity threshold (see **Data Repository**) indicated by vertical dash line in **C**, **F**.

Figure 4: **A** Viscosity as a function of temperature (Giordano et al., 2008), shown for water concentrations 0.68–0.1 wt.% and Chaitén glass melt composition. Approximate brittle failure/vesiculation regimes are highlighted. Dashed line indicates $\sigma_c/\dot{\gamma}_c$ (brittle failure threshold). **B** Schematic of dehydration quench mechanism. 1: fracture created within dense, avesicular magma aliquot. 2: fracture exploited as gas/ash venting pathway; H_2O diffuses from melt into fracture. 3: diffusion leaves H_2O-depleted selvage; sintering of hot ash particles effectively seals fracture. 4: far-field magma vesiculates, but fracture-adjacent H_2O-poor regions do not. 5: bubble-driven bomb expansion elicits viscous deformation in H_2O-rich melt zones (i.e. low viscosity: **A**), but H_2O-depleted regions undergo a brittle response. **C** Contextual cartoon of in-conduit processes described in **B**, shallowing from 1 to 5.

Figure 1

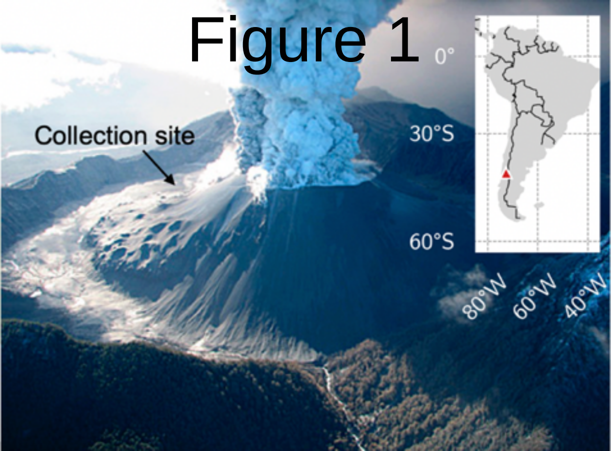


Figure 2

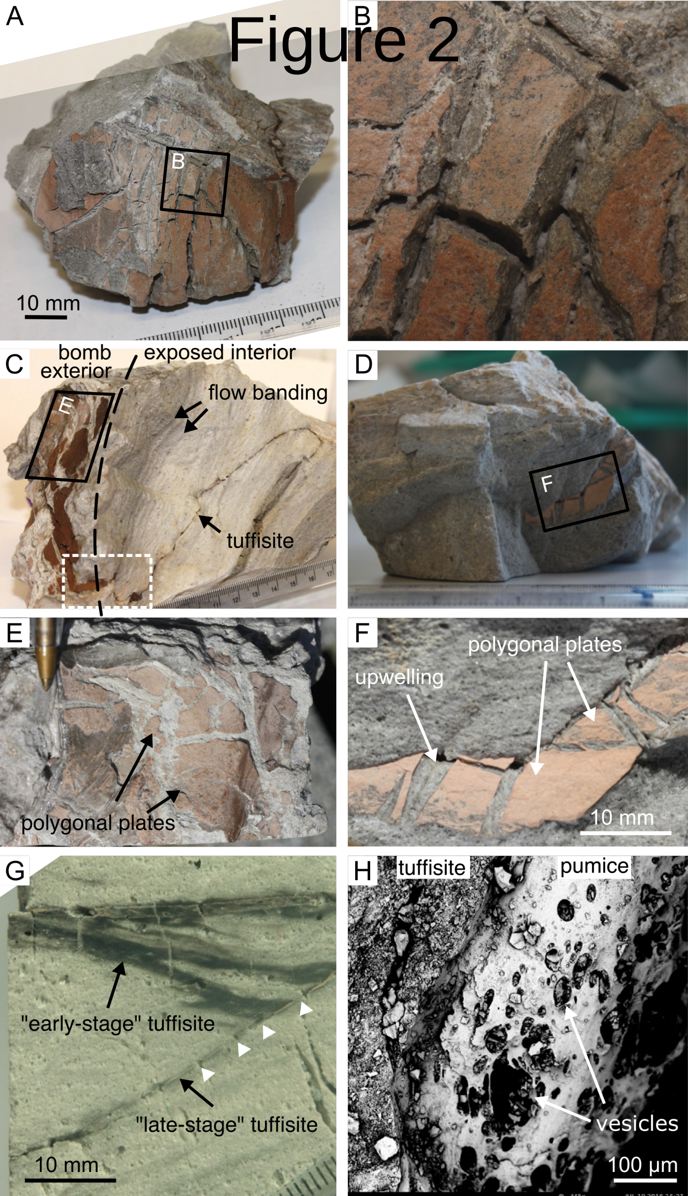

A
B
10 mm
B
C
bomb exterior
exposed interior
flow banding
tuffisite
E
D
F
E
polygonal plates
F
upwelling
polygonal plates
10 mm
G
"early-stage" tuffisite
"late-stage" tuffisite
10 mm
H
tuffisite
pumice
vesicles
100 µm

Figure 3

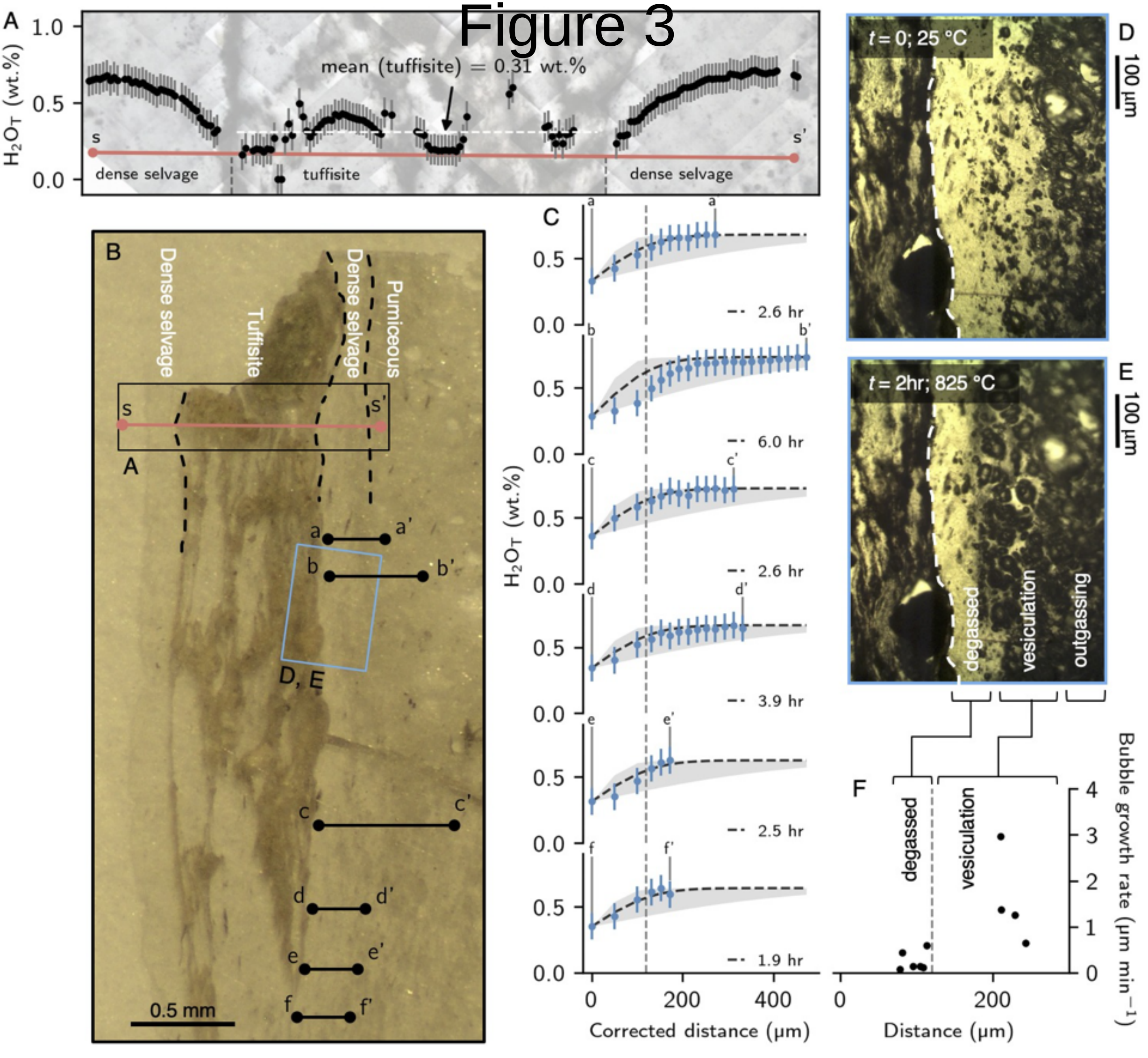
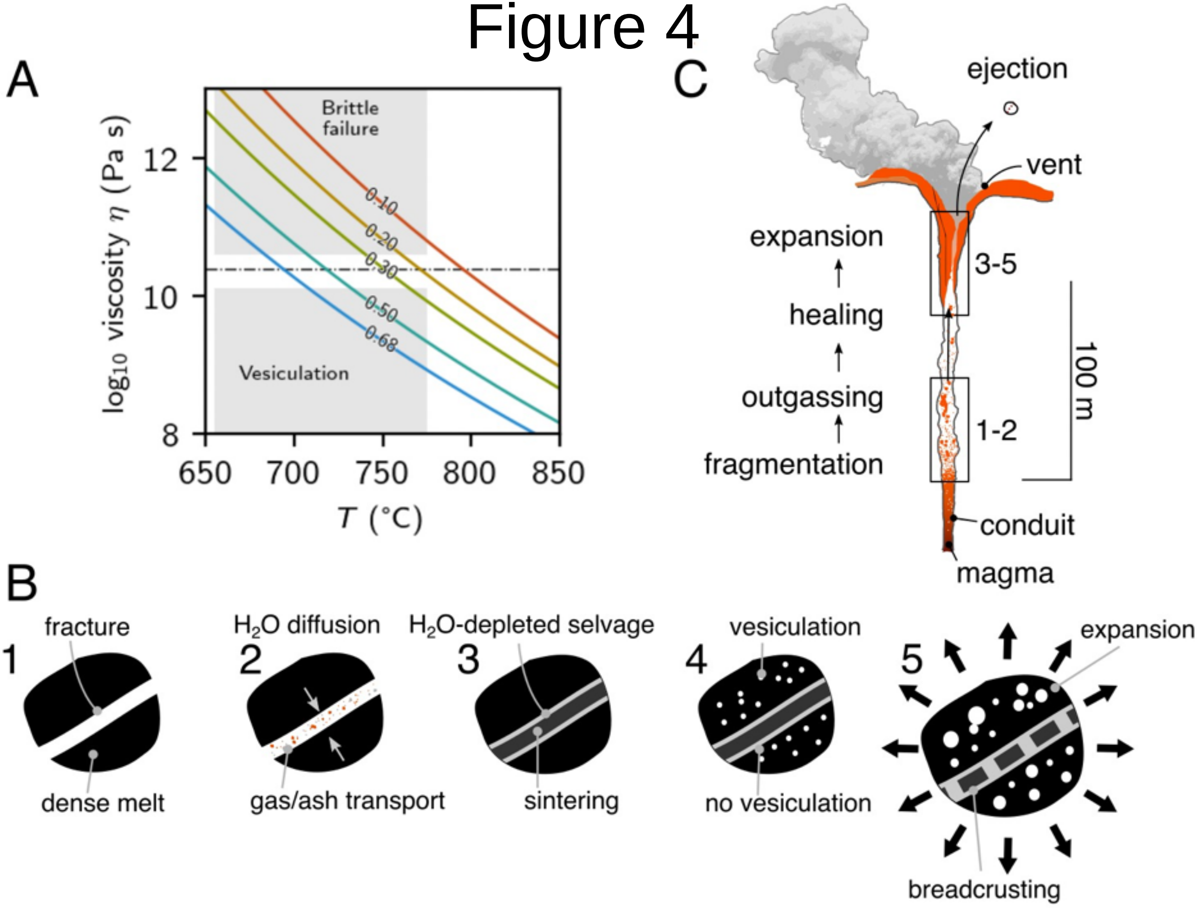

A
1.0
0.5
0.0
H_2O_T (wt.%)
mean (tuffisite) = 0.31 wt.%
s
s'
dense selvage
tuffisite
dense selvage
B
Dense selvage
Tuffisite
Dense selvage
Pumiceous
s
s'
A
a
a'
b
b'
D, E
c
c'
d
d'
e
e'
f
f'
0.5 mm
C
H_2O_T (wt.%)
2.6 hr
6.0 hr
2.6 hr
3.9 hr
2.5 hr
1.9 hr
0
200
400
Corrected distance (μm)
D
t = 0; 25 °C
100 μm
E
t = 2hr; 825 °C
100 μm
degassed
vesiculation
outgassing
F
degassed
vesiculation
0
200
Distance (μm)
Bubble growth rate (μm min^{-1})
0
1
2
3
4

Figure 4


A
log$_{10}$ viscosity η (Pa s)
12
10
8
650
700
750
800
850
T (°C)
Brittle failure
Vesiculation
0.10
0.20
0.30
0.50
0.68
C
ejection
vent
expansion
healing
outgassing
fragmentation
3-5
1-2
100 m
conduit
magma
B
1
fracture
dense melt
2
H_2O diffusion
gas/ash transport
3
H_2O-depleted selvage
sintering
4
vesiculation
no vesiculation
5
expansion
breadcrusting

Dear Editor,

Thank you for your handling of this manuscript. We appreciate the comments from all three reviewers, each of whom highlighted that the research is of broad interest and represents a significant advance in understanding. The reviewer comments are reproduced below, with our responses given in **bold**. Where applicable, changes to the manuscript text are given in **green**. Minor changes to the text have been made for clarity or accuracy following the reviewers' advice. We have also updated Figure 2 with additional images of the clast, and included a section in the final paragraph that links the textures and process described in this manuscript to other volcanic deposits. Minor changes have been made throughout in order that the manuscript does not exceed the character limit after including additions requested by the reviewers. We hope you will consider our improved manuscript suitable for publication in *Geology*.

Yours sincerely,
Jamie Farquharson, on behalf of the co-authors.

Reviewer Comments:

Reviewer #1 Evaluations:

Recommendation: Minor Revisions
Provocative: Yes
Of Broad Interest: Yes
Represent Significant Advance in Understanding: Yes
Innovative: Yes
Objects and Rationales Presented Clearly: Rationale and objectives are clear
Methods and Data Adequate: Methods and data support the objective and hypothesis
Conclusions Clear and Supported: The summary of results is concise and accurate
Figures Pertinent: Figures and tables support the data and conclusions and are legible
Supplemental Information Appropriate: Supplemental material is supplemental and supports the objectives of the paper

Reviewer #1 (Identity):

Amelia Bain

Reviewer #1 (Comments to the Author):

This manuscript describes and analyses for the first time the occurrence of breadcrusted curviplanar features cross-cutting the interior of rhyolitic bombs from Chaitén volcano. These brittle features appear to have formed along volatile-depleted tuffisite veins formed prior to the ejection of the bomb from the volcanic conduit, whereas adjacent vesicular material clearly behaved in a viscous manner contemporaneously. In addition, these features contrast with more commonly described breadcrusted surfaces on bomb exteriors, which are typically interpreted to have formed due to cooling-induced quenching upon ejection from the conduit.
The authors analyse the H2O content of glass in texturally contrasting zones across a cross-section through these features, and calculate the recorded timescale for H2O diffusion from the bomb interior into the tuffisite vein. They also calculate the timescale for cooling of the bomb, giving its 'thermal lifetime', and demonstrate that the diffusion timescale is shorter than the cooling timescale of the bomb, showing that the diffusion gradient recorded could have formed before the bomb cooled. They also demonstrate, via a heating experiment, that certain textural domains still have the potential to thermally vesiculate, whereas other domains are so volatile depleted that vesiculation is suppressed. The authors interpret their findings as evidence of chemo-rheological quenching, where volatile depletion induces brittle behaviour of the tuffisite vein and adjacent glass upon deformation, producing internal breadcrusting in a volcanic bomb.
The paper is very well written, clear, and well-argumented. The findings are novel, and the narrative is compelling. The figures illustrate well the main points, arguments and results, and are of high-quality. The supplemental information is consistent with the Geology policy statement. I believe that the paper is of broad interest to the readership

of the journal, and written in a manner that is accessible to a wide range of backgrounds. In addition, the documentation of a chemo-rheological quenching mechanism producing breadcrusting contrasts with the generally accepted model of thermo-rheological quenching, and is therefore paradigm-shifting and does not merely represent an incremental scientific advance. I agree with the authors' conclusion that "similar deposits recorded at other volcanoes worldwide may require reanalysis and reinterpretation", based on their findings. Finally, the manuscript demonstrates for the first time that breadcrusting in volcanic bombs can form due to more than one mechanism, as the paper does not invalidate the concept of thermo-rheological breadcrusting in other settings. This is a finding that will be of broad interest, will impact field interpretations of breadcrusting observations, and will foment further discussions and investigations of conduit processes.
I therefore recommend this article for publication in 'Geology' and provide some minor comments below that may improve the text.

We thank the reviewer for their kind remarks on the manuscript, in particular noting that it is “paradigm-shifting” and “well written.” We have responded to their detailed comments below.

Minor comments

Lines 47-49: Bain et al. (2019) also found a threshold of >0.4 wt% H2O for vesiculation to occur in andesitic bombs from Galeras volcano (Bull. Volc. 81:1, 2019).

We thank the reviewer for bringing this reference to our attention, and have now included it:
“Threshold values—associated with variable magma outgassing—are typically in the range 0.4–0.9 wt.% (Hoblitt and Harmon 1993; Bain et al., 2019; Wright et al., 2007).”

Line 112: It seems that "is" might be missing from the first part of the sentence to make it flow better, please revisit for clarity.

We have now reworded: “The approximate bomb core porosity ϕ is 0.6, and…”

The following sentence has now also been reorganized for clarity:
“Toward the dense obsidian edge, vesiculation has also dilated the spatial coordinates in the observed H_2O gradient.”

Line 118: Clarity could be enhanced for readers with a range of backgrounds by defining lambda_D and D as part of the sentence, e.g. "Defining the diffusion timescale as lambda_D" and "with the diffusivity D", as has been done for L.

Good suggestion. We now state:

“We define the diffusion timescale as $\lambda_D = L^2/(4D)$, with diffusivity $D \approx 1.3 \times 10^{-12}\ \mathrm{m^2 s^{-1}}$ (Zhang and Ni, 2010) for 825 ℃. The diffusion timescale required to

produce the observed diffusion gradient lengthscales is $\lambda_D = 4.81$ hours."

Line 121: Similarly, consider defining c and c0 within this sentence, for clarity.

We now state: "…the observed value of H_2O concentration c at the wall"

Line 123: I suggest using "we compute the time t", for readability.

We have made the suggested change.

Line 155: I suggest using "R is the pyroclast radius".

We have made the suggested change.

On line 123, the authors use t to define the time over which diffusion occurred, and on line 169, t is defined as a timescale for bubble growth/timescale for vesiculation-derived vesiculation to occur. I suggest using different subscripts for each of these for clarity, in the text and within the corresponding equations.

We have now modified t to read t_D (diffusion time) or t_B (bubble growth time), as appropriate, throughout the manuscript.

Line 194: I suggest to use "resulted in breadcrusting within the interior and on the exterior to the bomb", or something similar, for clarity/readability.

We now state:
"a brittle response in the mechanically coupled volatile-poor selvages and incipiently sintered tuffisite vein-fill, which resulted in breadcrusting within the interior and on the exterior of the bomb."

Lines 195/196: I suggest to replace "All of (0)-(5) occurred on the thermal lifetime of the bomb pre- and syn-ejection." by "Stages (0)-(5) all occurred within the thermal lifetime of the bomb, pre- and syn-ejection", for readability.

We have made this change as suggested.

Lines 197/198: I suggest to use the past tense in this sentence - "led" and "depended".

We have made this change as suggested.

Line 347: I suggest replacing the semi-colon by a comma.

We have made this change as suggested.

References/Citations:

The following reference is not currently cited within the text:
Berlo, K., Tuffen, H., Smith, V.C., Castro, J.M., Pyle, D.M., Mather, T.A., and Geraki, K., 2013, Element variations in rhyolitic magma resulting from gas transport: Geochimica et Cosmochimica Acta, v. 121, p. 436-451, doi:10.1016/j.gca.2013.07.032.

This has been added to the text:
"…consistent with other constraints on Chaitén tuffisite wall diffusion times (e.g. Castro et al., 2012; Berlo et al., 2013; Saubin et al., 2016; Heap et al., 2019)…"

The list of authors is incomplete for the following references:

Schipper, C.I. et al., 2021, Silicic conduits as supersized tuffisites: Clastogenic influences on shifting eruption styles at Cordón Caulle volcano (Chile): Bulletin of Volcanology, v. 83, p. 11, doi:10.1007/s00445-020-01432-1.

Wadsworth, F.B. et al., 2021, A model for permeability evolution during volcanic welding: Journal of Volcanology and Geothermal Research, v. 409, p. 107118, doi:10.1016/j.jvolgeores.2020.107118.

Well noticed. We have been using the Geology style with the Zotero citation manager, but it seems to have truncated some of the author lists. These have now been amended.

Citations without a corresponding reference:
Schipper et al. 2013 (line 53)

The reference

Schipper, C.I., Castro, J.M., Tuffen, H., James, M.R., and How, P., 2013, Shallow vent architecture during hybrid explosive–effusive activity at Cordón Caulle (Chile, 2011–12): Evidence from direct observations and pyroclast textures: Journal of Volcanology and Geothermal Research, v. 262, p. 25–37, doi:10.1016/j.jvolgeores.2013.06.005.

has been added.

Reviewer #2 Evaluations:

Recommendation: Accept
Provocative: Yes
Of Broad Interest: Yes
Represent Significant Advance in Understanding: Yes
Innovative: Yes

Objects and Rationales Presented Clearly: Rationale and objectives are clear
Methods and Data Adequate: Methods and data support the objective and hypothesis
Methodology is up to date
Conclusions Clear and Supported: The summary of results is concise and accurate
Figures Pertinent: Figures and tables support the data and conclusions and are legible
Supplemental Information Appropriate: Supplemental material is supplemental and supports the objectives of the paper

Reviewer #2 (Identity):

Benjamin Andrews

Reviewer #2 (Comments to the Author):

Review of Mid-loaf Crisis: Internal breadcrust surfaces...
Authors: Hugh Tuffen, Jamie Farquharson, Fabian Wadsworth, Cameron Webb, Jacqueline Owen, Jonathan Castro, Kim Berlo, C. Schipper, and Katia Wehbe
Reviewer: Benjamin Andrews, Smithsonian Global Volcanism Program

Summary
The authors present detailed textural and FTIR analysis of a bomb from Chaiten volcano that exhibits internal and internal breadcrusted surfaces or features. A pyroclast will develop a breadcrust surface when its exterior (or, as shown in this paper, even interior) surfaces are more viscous than the vesiculating and expanding interior, this rheological difference results in brittle deformation of the more viscous surface and the formation of "breadcrust" textures. The work here shows that breadcrusted surfaces can exist on bomb interiors, not just the exterior, and that both the interior and exterior breadcrusted surfaces are commonly jacketed with, or comprise, tuffisite veins. The authors show that the tuffisite veins and their glassy walls are more degassed than the interior, vesiculated portions of the bomb. The authors explain the textures as recording at least two episodes of fracture formation, tuffisite production, and partial degassing of the tuffisite and nearby melt. That degassing increases the viscosity of the vein and walls. When the bomb is erupted (likely through another episode of fracturing and ash production), its interior vesiculates and expands. The strain induced by the expansion occurs too quickly for the tuffisite vein to flow viscously, and the vein instead deforms brittly, resulting in a breadcrusted interior surface. The authors refer to this as a "chemical" or "degassing" quench.

Importance or Significance
The authors make a compelling argument for dehydration quenching as an important process in at least some volcanic eruptions (certainly in Vulcanian eruptions, and probably in eruptions of silicic lavas). The authors show that the diffusion timescales indicated by FTIR profiles of water concentration in the melt are consistent with several hours between fracture formation and quenching, and those timescales are consistent with previous estimates of the timescales of tuffisite formation at Chaiten. Although this is a case study (for Chaiten), this approach could be applied to samples from other

volcanoes to better understand the timescales of shallow processes (that is, processes in the final few hundred meters). In addition, the authors present a regime diagram that shows the conditions over which dehydration quenching should be expected to produce breadcrusting, thus providing a mechanism to at least coarsely apply these findings to other volcanoes.

Areas for improvement
The only revisions that I suggest are to refer to the quench as "dehydration quench" or "degassing quench", and to consider making the analogy between the interior breadcrusted surface and boudins that appear in metamorphic rock - this could help structural and metamorphic petrologist folk relate to the textures discussed in the paper. The paper is clearly written, with good figures, and a clear organization.

We thank the reviewer for these suggestions. We have changed "chemical quench" to "dehydration quench" throughout the manuscript as appropriate. We have also updated the title to explicitly mention this process: "*Mid-loaf crisis: Internal breadcrust surfaces in rhyolitic pyroclasts reveal dehydration quenching*." We appreciate the suggestion to include a mention of boudinage. While we acknowledge that there are some interesting parallels between certain boudinage mechanisms in metamorphic rocks and the expansion-extension-fracture model presented in our article, they are not directly analogous—due to metamorphism taking place entirely through solid state crystallization, whereas our materials are mostly amorphous melts and glass in the throes of brittle-viscous behaviour—and thus, we think this might invite confusion. Given the space constraints of Geology articles, we are reticent to include a cursory comparison to a metamorphic process that isn't directly relevant to the key features under discussion. That said, there are natural examples of boudinage in rhyolite domes (Panum, California) that for all intents and purposes formed in similar ways to those in our study, particularly as pertains to the juxtaposition of rheologically contrasting materials that in turn show brittle (degassed) and viscous (bubbly) behaviour. We have added a reference (Castro and Cashman, 1999) that describes rhyolite boudinage in the revised manuscript and have inserted a new sentence (we think the inclusion of rhyolite boudinage may help stimulate further investigaton into the applicability of our dehydration quenching to other structures in rhyolite lavas):

"Comparable textures have been previously observed in mesoscale boudinage structures in vesicular rhyolite domes (e.g., Panum dome, USA: Castro and Cashman, 1999)."

Recommendation
I recommend acceptance. The paper is clearly written, and it addresses an important topic in volcanology. The content should also be of interest to the broader community as it discusses a "popular" type of pyroclast (lots of geology departments have breadcrust bombs on display or in their teaching collections), and it shows how quantitative and

otherwise inaccessible information on timescales can be accessed through relatively straightforward observations and application of simple/elegant equations.

Sincerely,
Benjamin Andrews
Smithsonian Global Volcanism Program

We thank the reviewer for their kind comments, and appreciate the additional detailed comments, which we have responded to below.

Line-by-line comments
24 - suggest changing from "to the breadcrusting" to "to the breadcrusted surfaces"

We have made this change as suggested: "We find H_2O diffusion gradients proximal to the breadcrusted surfaces, such that…"

30 - I like the concept of the "chemical quench," but I wonder if it might be better phrased as a "dehydration quench" - my reasoning is that this is the process that is occurring, and so you can be a bit more specific.

We thank the reviewer for this suggestion, and have amended "chemical quench" to "dehydration quench" where appropriate.

99 - the description of the bomb in this paragraph is generally good and clear. I would suggest, however, that you use a different phrase than "adhering pumice" - maybe "underlying pumice" for clarity or disambiguation. I know this is a minor issue (especially as the fine clasts adhere to the pumice and the pumice technically adheres to the clasts) but generally I think of the little guys as adhering to the big fellow (and not the other way around).

A fair point—we have made this change as suggested: "…whereas the underlying pumice has flowed viscously into the resultant voids…"

111 - by "atypical" do you mean that these are the "best of" transects and represent the ideal or best case transects rather than being representative? Just checking.

The transects are representative of the bomb itself, but atypical in the sense that most published diffusion transects are not overprinted by a secondary mechanism (in this case, vesiculation). This is why a correction is applied to the data. We now clarify in the text: "atypical diffusion gradients, inasmuch as vesiculation has overprinted the diffusion process."

133 - how thick was the glass wafer used in the experiment?

The median thickness is 112 µm. There is some variability (101–115 µm) given the pre-existing sample vesicularity) We have now added this information to the manuscript:
“We heated a polished wafer (~112 µm thick) from the early-stage tuffisite vein margin at 200 °C min^{-1} and ambient pressure to 825 °C…”

156 - given that this paper talks about different types of "quenching," what type of quenching occurs over the ~2.6 hours? Is this thermal quenching, dehydration quenching? Does it matter?

Presumably both quench mechanisms are operative during this timeframe, although—as the reviewer hints at—decoupling the contributions of the mechanisms at this stage and for this specific bomb does not particularly matter. The important observation is the evidence for a dehydration quench occurring at all, which is the novel aspect of our contribution.

162 - I suggest making this sentence begin with "We argue that the late-stage vein and its walls..." I think that you make a good point and you back it up in the rest of the paragraph, but it needs to be clear that you are arguing or postulating that the viscosity of the vein/walls is greater than the far-field melt, not that it is a given.

We agree with the reviewer, and have amended the text accordingly:
“We argue that the late-stage vein and its walls must have a higher viscosity than the far-field melt due to relative volatile depletion, embrittling the magma adjacent to the vein margins.”

Reviewer #3 Evaluations:

Recommendation: Minor Revisions
Provocative: Yes
Of Broad Interest: Yes
Represent Significant Advance in Understanding: Yes
Innovative: Yes
Objects and Rationales Presented Clearly: Objective is not supported by background
Methods and Data Adequate: Methodology is up to date
Analyses are inappropriate
Needs further analyses
Conclusions Clear and Supported: The summary of results is concise and accurate
Paper is missing discussion of potential limitations
Figures Pertinent: Figures and tables support the data and conclusions and are legible
Supplemental Information Appropriate: Supplemental material is supplemental and supports the objectives of the paper

Reviewer #3 (Identity):

Steve Quane

General Comments: This manuscript comprises a comprehensive treatment of textures in breadcrusted bombs from Volcan Chaiten in Chile. The analyses are of high quality and are thorough. I appreciate the detailed H2O analysis and accompanying experimental replication of the processes involved. The coupling of the H2O analysis with rheology and viscosity calculations allow the authors to place a timescale on the processes involved and that adds depth and meaning to the results.

Critical Issue: I have one main issue with the paper, however, that has potential implications of its overall meaning. Are these actually breadcrusted bombs? I understand that they are discrete pyroclasts that show breadcrust texture, but what is the evidence that any of the breadcrusting happened post fragmentation? In other words, did the breadcrusting actually take place as a bomb, or could all of these processes have occurred when the material was part of the dome and the fragmentation could have emplaced these materials as already deformed bombs. It is stated that these bombs were emplaced during the hybrid phase 2 of the eruption (transitional explosive–effusive activity, 20 days in length).

Looking at Figure 1, it appears that the bombs were collected proximal to the eruption conduit. Presumably, during the phase 2 a plug of glassy dome-like material was extruding to the surface and was fragmenting. The proximity of the bombs to the vent and the relatively large size ofteh bombs, indicates that the gas pressures causing fragmentation of these bombs were not sufficient to pulverize all of the magma into ash (as seen in the plume of ash exiting the vent; Figure 1).

So, I ask this question, what evidence do you have that these bombs are not fractured pieces of the lava dome that were ejected? What evidence exists that the textures found in the pieces happened as a result of it being a bomb? In the manuscript, you state: Bomb ejection occurred along with (4) or in an explosion shortly afterwards (**Fig. 4C**). All of (0)–(5) occurred on the thermal lifetime of the bomb pre- and syn-ejection. How is this different from the deformation (brittle and ductile) that happens in an obsidian lava flow?

Looking at photos from a visit to Obsidian Dome near Mammoth, California, I see similar textures to what you are using to explain the chemo-rheological process occurring at Chaiten.

See pictures here:

[…]

While these photos are not showing the exact textures you show in Figure 2, they are similar-polygonal plates of material on the surface; flow banding in the interior; variably

vesiculated material adhered to dense materials. In addition, looking at the photos below from Andrews et al., 2020 (Figure 6), many of the textures you mentioned as evidence for this process in a bomb exist in deformed lava flows:
This is not to discount the importance of chemo-rheological changes in the materials that you have found. The profiles of H2O indicate clearly a viscosity difference will occur in the different materials causing rheological differences.

When I look at your Figure 2, I am not convinced that you are showing a breadcrust bomb. The pieces you have are discrete and have been ejected from the vent, so, yes, they should be classified as bombs, however, the photos you are showing do not show convincing breadcrust texture. You compare your bombs to Wright et al., 2007; Giachetti et al., 2010, but from the photos in Figure 2, I am not seeing a convincing breadcrust surface that differentiates these bombs as breadcrusted pyroclasts and not severed pieces of a deforming lava dome.

The distinction is important for a two reasons. A) Without showing convincing evidence of these being breadcrust bombs and indicating that the deformation processes forming the textures found are happening pre and syn ejection, you are not differentiating these from deformation happening in lava flows or conduits. B) The reason this is important is that you are limiting yourself to breadcrusting in bombs and perhaps the chemo-rheological process you are describing has larger implications for in-conduit processes that you could explore.

Resolution: I think you need to show better evidence for breadcust BOMB features in Figure 2. This might comprise a better photo of the exterior surface of the bomb to show the classic breadcrust feature, if it exists. You might also reconsider broadening your application of this chemo-rheological process to other processes. If you claim that this all happened in conduit or syn-ejection, doesn’t that have larger implications for the behavior of this type of eruption? Isn’t that fundamentally different bomb behavior than those described by Wright et al., 2007 and Giachetti et al., 2010? Those bombs clearly deformed post-ejection.

I feel that these issues need to be clearly addressed in order for the impact of these findings to be clear.

The third reviewer highlights a single concern with the manuscript, the main points of which are reproduced here:

Are these actually breadcrusted bombs?

[W]hat evidence do you have that these bombs are not fractured pieces of the lava dome that were ejected?

We do not have photos of the clasts *in situ*, but they were collected from a pock-marked (i.e., cratered) bomb field that was strewn only with ballistics and not blocks of collapse flow-front lava. There is also evidence within the clast of substantial vesiculation, to a far greater extent

than observed within the dome material at Chaitén. We infer that this reflects a decompression-triggered vesiculation event (similar to the bomb described in Saubin et al. [2016]). Notably, the measured far-field H_2O concentrations in this sample are 0.68 ± 0.04 wt.%. With the reasonable assumption that this represents a lower bound on pre-ejection solubility, equilibration pressure is ~4 MPa (Newman and Lowenstern 2002): corresponding to a magmastatic depth of <u>at least</u> 170 m. The dome thickness as of 26 May 2008 was estimated at 120 m (Pallister et al 2013, Andean Geology, Fig. 6), and the sample emplacement predated the extrusion of the dome, probably coming from close to the end of the purely explosive phase of 1–11 May. At this stage—towards the end of the purely explosive phase—the conduit is thought to have started to block up with debris just prior to the first lava effusion. As such, the sample was initially ejected from deeper than the dome, and potentially during a period where there was no lava dome at all. Together with its size (>64 mm diameter) and form (very approximately spherical), these facts suggest that this particular clast is indeed a bomb, by any definition. Nevertheless, to avoid any potential confusion related to the clast genesis, we now refer instead to "pyroclast" throughout, as this is a non-genetic descriptor.

What evidence exists that the textures found in the pieces happened as a result of it being a bomb?

The reviewer's comment is perceptive, in that the key process outlined here—dehydration quenching—is not limited to breadcrust bombs. It is simply that in the case of the clast in question, the driving force for (post-fracture) vesiculation was decompression that most probably accompanied ejection as a bomb. However, *any* process that results in vesiculation after diffusive degassing will be subject to rheological gradients and therefore prone to "breadcrusting", even without thermal gradients.

How is this different from the deformation (brittle and ductile) that happens in an obsidian lava flow?

The reviewer references images from Andrews et al. 2021 (EPSL, https://doi.org/10.1016/j.epsl.2020.116643). In that case, we infer that a secondary vesiculation event in the lava has driven segmentation of a pre-existing tuffisite layer. The reviewer hints at the similarity of these features, and we agree that the processes are related (although not identical). To reflect this (and the comments above), we have made a series of changes that reflect the broader applicability of our model.

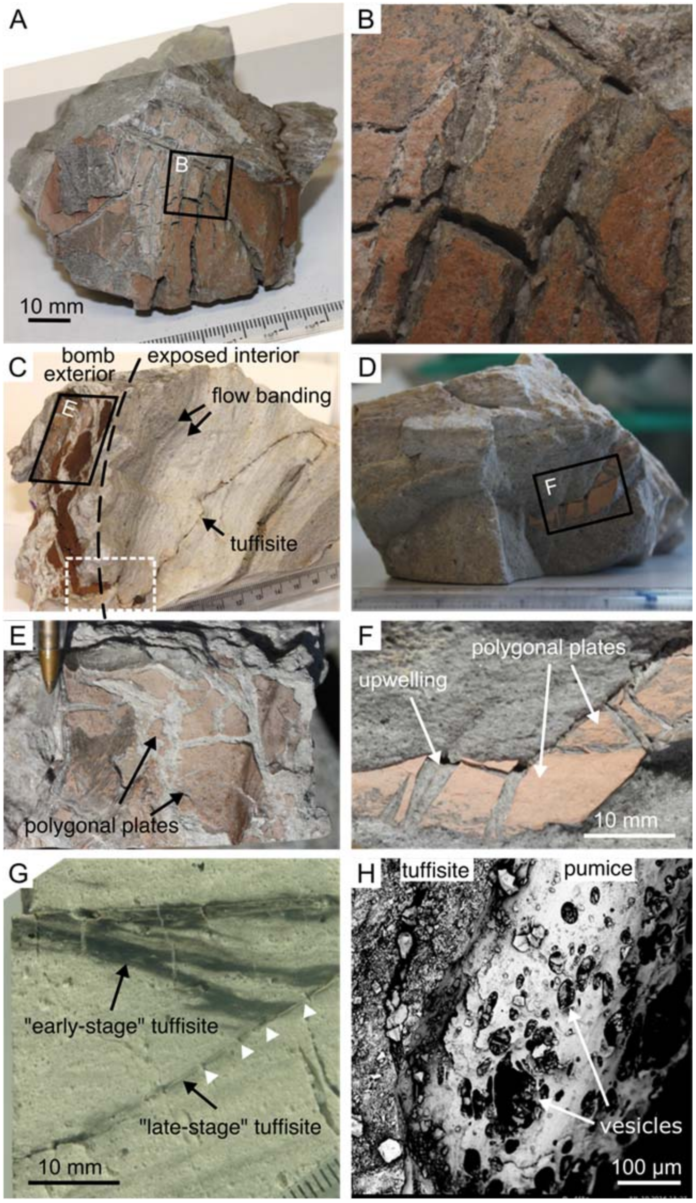
- **We have updated Figure 2 with two extra panels that better illustrate the "typical" breadcrust textures (shown later in this document), including the segmentation of "late-stage" tuffisites;**
- **We now refer to "pyroclasts" rather than "bombs" throughout the text;**
- **We have modified the title to spotlight the process rather than the sample type: "Mid-loaf crisis: Internal breadcrust surfaces in rhyolitic pyroclasts reveal dehydration quenching";**
- **We are more explicit about the emplacement conditions: "Column-collapse pyroclastic density currents (PDCs) were emplaced within the caldera late in phase 1; hybrid**

phase 2 involved eruption of rhyolitic lava domes and coulées with synchronous, repeated Vulcanian explosions and intermittent PDC-producing plumes (Pallister et al., 2013), with deposits emplaced on the caldera floor (Fig 1). " … "Here we describe an irregular pyroclast (15 × 25 × 20 cm) (Fig. 2), which is typical of many deposits—including bombs and bomb fragments—within the study area, emplaced during the latter part of the explosive phase I."

- We conclude the manuscript with explicit reference to Andrews et al., and highlight that textures which have been attributed uniquely to thermal processes may indeed be manifestations of dehydration quenching:

"To date, breadcrust textures in volcanic deposits have been interpreted as unambiguous evidence of a thermal quenching process. Our alternative mechanism for breadcrust formation—a chemo-rheological rather than thermo-rheological process—suggests that similar textures recorded in volcanic deposits worldwide may require reanalysis and reinterpretation. Such textures may occur in explosively ejected fragments of conduit plugs, where they have previously been uniquely interpreted as thermal quench features (i.e. classic breadcrust bombs), and also within silicic lavas, where relatively dense layers—e.g. tuffisites—can become segmented by heterogeneous vesiculation (Castro and Cashman, 1999; Andrews et al., 2021)."

We thank the reviewer for their insightful comments, which serve to highlight the broader applicability of our results.



Updated Figure 2