1	In	situ	granulation	by	thermal	stress	during	subaqueous
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# 2 volcanic eruptions

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#### 15 ABSTRACT

Some of the most complex volcanic thermodynamic processes occur when erupting magma interacts with water. In shallow water, "Surtseyan" eruptions are spectacular, and they efficiently fragment magma into fine ash particles. The aviation hazard from these eruptions depends the amount of transportable fine ash that is generated and whether it is aggregated into particle coatings or accretions. To investigate both mechanisms, we analyzed ash-encased lapilli from the Surtseyan eruptions of

22 Capelinhos (Azores, 1957–1958) and Hunga Tonga–Hunga Ha'apai (Tonga, 2014–2015)

23 using X-ray computed microtomography and electron microscopy. We discovered 24 pyroclasts that were not coated, sensu stricto, but had enveloping ash produced by *in situ* 25 granulation of the particle surface. This was caused by thermal stress as pyroclasts briefly 26 traveled through water and were quenched during eruption. In situ granulation is thus an 27 important secondary disruption process in shallow subaqueous eruptions. Our results 28 imply that ash encasement is not always evidence of particle aggregation and accretion, 29 but it may also result from new ash formation. Shallow-water conditions produce the 30 most efficient ash-generation conditions, leading to the greatest hazard to downwind 31 populations and air traffic.

#### 32 INTRODUCTION

33 Surtseyan eruptions are explosive volcanic events characterized by spectacular 34 explosive jets that burst through ocean or lake water. They were named after the A.D. 35 1963–1967 submarine eruption of Surtsey, Iceland (Thorarinsson, 1967), which followed 36 a similar well-observed eruption at Capelinhos in the Azores (1957–1958). One of the 37 most recent eruptions of this type was at Hunga Tonga–Hunga Ha'apai (Tonga) in 2014– 38 2015, which caused cancellation of international flights due to widespread ash dispersal 39 (Global Volcanism Program, https://volcano.si.edu/; Colombier et al., 2018; Garvin et al., 40 2018). Magma interaction with shallow groundwater or surface water often produces 41 copious volumes of fine volcanic ash, which is particularly hazardous to air transport, 42 such as the subglacial Eyafjallajökull event of 2010 (e.g., Gudmundsson et al., 2012). 43 Water is a far more efficient cooling medium than air, so that shock magma quenching is 44 a primary fragmentation mechanism (Zimanowski et al., 1997), alongside gas-driven 45 magmatic fragmentation (Gonnermann, 2015). Water may also drive secondary magma

46	disruption via steam-driven explosions (Kilgour et al., 2010), thermal granulation
47	(Kokelaar, 1986), and turbulent shedding (Mastin, 2007). The rapid cooling of
48	fragmenting magma also influences eruption dynamics by hindering vesiculation and
49	causing particle aggregation and recycling (Cole et al., 2001; Schipper and White, 2016).
50	Moore (1985) noted that ash-"coated" lapilli were the dominant constituent of the
51	deposits at Surtsey, and aggregation of ash particles was subsequently recognized as a
52	common process during magma-water interaction (e.g., Brown et al., 2012).
53	Aggregation of particles as accretionary lapilli or pyroclast coatings may lead to a
54	lower-than-expected ash dispersal and hazard. To examine the formation and stability of
55	ash haloes, we used three-dimensional (3-D) X-ray computed microtomography (micro-
56	XCT) on particles from Surtseyan eruptions (Capelinhos 1957–1958 and Hunga Tonga–
57	Hunga Ha'apai 2015–2016). We aimed to quantify the effectiveness of the aggregation
58	processes implied in the formation of these particle types (e.g., Mueller, 2013). Instead,
59	we discovered that these reflect additional posteruptive disruption via thermal
60	granulation.

# 61 **METHODS**

Using micro-XCT, we investigated four ash-encased lapilli from the basaltic andesitic to andesitic eruption of Hunga Tonga–Hunga Ha'apai volcano (e.g., Colombier et al., 2018) and three ash-encased lapilli from the basaltic eruption of Capelinhos (Zanon et al., 2013; for a detailed description of the eruption deposits and analytical conditions, see the GSA Data Repository<sup>1</sup>). Two bombs from the magmatic phase of the Capelinhos eruption were analyzed for thermal expansivity and determination of the glass transition temperature. We measured the thermal expansivity using a NETZSCH Dilatometer 402C

with a heating rate of 10 K min<sup>-1</sup> in argon. The linear expansion coefficient measured 69 between room temperature and T = 550 °C was  $\alpha = 5.5 \times 10^{-6}$  K<sup>-1</sup>. A differential 70 71 scanning calorimeter (DSC) 404C calibrated for temperature and sensitivity was used, and the DSC signal was acquired during heating at 10 K min<sup>-1</sup> in argon. The glass 72 73 transition temperature determined by both expansivity and DSC signal was 74 approximately  $T_{g} \sim 600 \,^{\circ}\text{C}$ . 75 NEW OBSERVATIONS FROM MICROTOMOGRAPHY 76 Ash-encased lapilli are the dominant constituent (>90%) of the 2–5 mm particle 77 size in fall and surge deposits from Capelinhos and Hunga Tonga–Hunga Ha'apai. Their 78 cores consist of scoria with variable vesicularity, vesicle-size distribution, vesicle shape, 79 and crystallinity (Figs. 1A–1C). They are partially to completely surrounded by a  $\leq$ 500-80  $\mu$ m-thick rim of fine ash particles (Figs. 1A–1C), frequently filling the external vesicles 81 at the clast margin (Figs. 1G–1I and 2D). The interface between scoria cores and the ash 82 rim is characterized by abundant cracks, sometimes organized in clusters (Figs. 1D–1I). 83 Particles were not cut or mechanically processed, so the fractures are natural. In partially 84 ash-encased lapilli, fractures occur only where ash rims exist (Figs. 1C, 1F, and 1I). The 85 cracks extend into the scoria and are commonly planar to curviplanar (Fig. 11). 86 In the ash rim, and at the contact to the core, we observed matching jigsaw-fit 87 fracture planes (Figs. 1H, 2A, and 2B), vesicle concavities (Figs. 1H and 2C), and crystal 88 fragments (Figs. 2A and 2B). Many of the jigsaw-textured domains showed varying 89 degrees of internal particle rotation and/or displacement down to the scale of the smallest 90 particles (~20 µm) clearly observable with the micro-XCT resolution. The crack number 91 density decreased from the ash rim toward the scoria core (Figs. 1 and 2).

92	We found that the heterogeneous vesicle and crystal textures strongly influenced
93	the number, density, size, and geometry of the cracks in the particles observed. Vesicles
94	with regular crack spacing demonstrated radial stress concentration and crack activation
95	(Heap et al., 2014; Figs. 2D and 2E). Cracks also commonly diverted at interfaces
96	between phases, showing propagation along crystal-glass boundaries (Fig. DR1 in the
97	Data Repository). These features influenced crack number, density, and size as well as
98	the grain size and morphology of resulting ash particles, similar to that reported
99	elsewhere (Liu et al., 2015).

#### 100 ORIGIN OF THE ASH-ENCASED LAPILLI

101 The jigsaw fit of the ash rims to the host particles indicates that they formed by 102 brittle granulation of the particle margins. That is, these are not "coated" particles, sensu 103 stricto. The clear shape correspondence and packing of neighboring particles in three 104 dimensions show that they were formed in situ by disruption of the porous scoria. 105 The outward increase in the crack number and density from the core to the ash rims 106 implies a continuous transition between cracking and granulation. Therefore, the cracks 107 and jigsaw-fit textures were formed by the same brittle process. 108 The presence of jigsaw textures in subaqueous settings has been reported in 109 studies of hyaloclastites and peperites (Carlisle, 1963; Staudigel and Schmincke, 1984; 110 Hanson and Hargrove, 1999; Doyle, 2000; Skilling et al., 2002) and is generally 111 attributed to *in situ* thermal granulation during water-magma interaction. In this process, 112 and in contrast to explosive fragmentation mechanisms (cf. Carlisle, 1963; van Otterloo

113 et al., 2015), particles remain at their sites of formation. By analogy, we propose that

114	after explosive magma fragmentation, the margins of the lapilli-sized clasts examined
115	here experienced thermal cracking and granulation due to quenching in seawater.
116	During quenching, the margins of particles experience the highest local cooling
117	rates and thermal stress. In the case of direct contact with water, where vapor films are
118	absent or collapse, we can assume that the surface of the particles is instantaneously
119	cooled to the water temperature ( $T_{\rm w} \sim 25$ °C). Therefore, instant thermal stress $\sigma$ can then
120	be calculated using (van Otterloo et al., 2015):
121	$\sigma = (E\alpha\Delta T)/(1-\nu), \qquad (1)$
122	where E is the elastic modulus ( $E = 73$ GPa; Schultz, 1993), $\alpha$ is the thermal expansivity
123	( $\alpha = 5.5 \times 10^{-6} \text{ K}^{-1}$ ), v is Poisson's ratio ( $v = 0.25$ ; Schultz, 1993), and $\Delta T$ is the
124	quenching temperature difference. For the initial melt temperature, we chose a minimum
125	value that corresponds to the glass transition temperature $T_{\rm g}$ (600 °C). The temperature
126	difference $\Delta T$ is therefore $T_g - T_w = 575$ °C. This yields an instant thermal stress at the
127	surface of 308 MPa, which overcomes the tensile strength of basaltic glass ( $\sim 10^8$ Pa;
128	Webb and Dingwell, 1990). This confirms that in situ granulation by thermal stress is
129	plausible at the margins of the lapilli during interaction with seawater. The vesicularity
130	and vesicle size (e.g., Heap et al., 2014), and the permeability and crystallinity of the
131	lapilli all influence the tensile strength of basaltic rocks and therefore affect in situ
132	granulation by thermal stress. Thermal granulation can be a very fast process, as crack
133	propagation velocity is expected to be in the range of hundreds to thousands of meters per
134	second (e.g., van Otterloo et al., 2015). We do not rule out additional granulation induced
135	by clast-to-clast collisions, but we believe it would be localized to isolated impact points

and is not sufficient to explain the observed textures. We therefore propose that thermalstress is the dominant cause of disruption.

138 Some particles generated by *in situ* granulation may have been spalled off, 139 washed away, or dispersed by winds. Commonly, the observed jigsaw-fit ash particles are 140 slightly rotated or displaced from their site of origin, intruding into the external vesicles 141 of the scoria core and resulting in an ash rim with higher density than the core. Similar 142 densification of jigsaw-fit particles was also observed in peperites and hyaloclastites 143 (Hanson and Hargrove, 1999; Doyle, 2000). Inward displacement of some ash fragments 144 into the external vesicles and tight packing of ash in the rims likely reflect a combination 145 of (1) condensation of internal magmatic gas during quenching, causing suction and 146 absorption of water and ash particles (cf. Allen et al., 2008), (2) compression of 147 noncondensable magmatic gas such as  $SO_2$  or  $CO_2$  by the water column during cooling, 148 and (3) particle collisions during transport. Vesicularity, vesicle connectivity, and 149 permeability partly control the efficiency and depth of ash displacement into the particles 150 during densification. Most of these mechanisms imply densification occurred either 151 below the sea surface or shortly after ejection. After ejection, residual heat causes 152 evaporation of brine and precipitation of salts (e.g., NaCl or MgSO<sub>4</sub>), from both 153 magmatic gases and seawater (Ayris et al., 2014), which may stabilize the "coated lapilli" 154 (cf. Mueller et al., 2017). Our 3-D evidence from micro-XCT provides the first 155 documentation of thermal *in situ* granulation for the microscale production of fine-ash 156 particles in a volcanic eruption. 157 We propose a conceptual model to explain the formation of the ash-encased lapilli

158 in three steps (Fig. 3): Following the initial magma fragmentation, there is (1) direct

159 contact between a primary pyroclast and seawater, causing thermal-stress-induced 160 cracking and granulation. This is followed by (2) inward displacement of ash particles 161 and seawater into the particle, causing densification of the ash rim and release of some 162 outermost fragments generated by *in situ* granulation into the water column. Finally, (3) 163 the ash-encased lapilli are injected into the atmosphere, possibly accompanied by 164 precipitation of salts, which stabilize the ash rims and preserve the jigsaw-fit textures. 165 IMPLICATIONS FOR SUBAQUEOUS ERUPTIONS AND RELATED HAZARDS 166 Ash-encased lapilli are the dominant constituent of the lapilli fraction of deposits 167 from the Surtsey (1963–1967; Moore, 1985), Capelinhos (1957–1958), and Hunga 168 Tonga–Hunga Ha'apai (2014–2015) eruptions. These have always been termed "ash-169 coated" particles, with the inherent assumption of an active process where foreign ash 170 particles are attracted to and adhere to the outside margins of a lapilli particle, i.e., similar to the aggregation process of ash into accretionary lapilli in moist atmospheric eruption 171 172 plumes (Brown et al., 2012). The thermal cracks and jigsaw textures observed in all the 173 lapilli examined in our study indicate that ash rims on scoriaceous lapilli may rather 174 result dominantly from thermal granulation of particle margins. This implies a greater 175 importance of this secondary disruption process than previously considered. Thermal 176 granulation probably contributes to magma disruption during subaqueous eruptions 177 occurring at any water depth and magma composition, producing particles with a broad 178 range of sizes (e.g., the formation of metric pumices during the 2012 Havre eruption; 179 Manga et al., 2018). In contrast to conventional dry and wet aggregation in volcanic ash 180 plumes (Brown et al., 2012), the *in situ* granulation model binds at least some of the ash 181 directly after generation. In this scenario, the ash-encased particles are actually an

182	indicator of ash production, rather than sequestration of free-ash particles into coatings.
183	Additional wet aggregation of ash particles, or alternatively loss of ash from the rims,
184	may occur during transport above sea level. Wet particle aggregation initiates at relative
185	atmospheric humidity levels of 15%-20% or higher (Mueller et al., 2016), which is
186	highly likely in Surtseyan eruption plumes. Interpreting ash-encased lapilli solely as the
187	result of aggregation following primary fragmentation might cause an error in the
188	inferred aggregation rate and total grain-size distribution, which are two essential
189	eruption source parameters in models of tephra dispersal (e.g., Folch et al., 2010). In the
190	future, understanding the conditions that alter the relative balance between in-plume
191	aggregation (decreasing free ash) and the subaqueous production of ash by in situ
192	granulation (possibly increasing free ash) will be a key for better assessment of potential
193	hazard of ash particles in the atmosphere impacting human populations and air traffic.
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#### 308 FIGURE CAPTIONS

- 309 Figure 1. Two- and three-dimensional (2-D and 3-D) textural properties of ash-encased
- 310 lapilli. Top, medium, and bottom rows correspond to samples CAP370–3–1, HH47–2,
- 311 and HH28–3, respectively. A–C: 2-D slices through X-ray computed microtomography
- 312 (XCT) data showing internal textures of lapilli. White line corresponds to boundary
- 313 between core and ash rim. Ash particles filling external vesicles of cores are common.
- 314 Note highly variable internal textures (vesicle size, elongation, size distribution, and
- 315 crystallinity of cores) in lapilli. D–F: 3-D XCT volume renderings illustrating cores
- 316 (blue) and presence of large cracks at their margins (red). Dashed circles in E serve to
- 317 highlight locations of smaller cracks in this sample. G: 2-D slice through XCT data of

318	sample CAP370-3-1 showing margins of core (pink outline) and associated ash rim with
319	jigsaw-fit particles (outlined in blue solid lines). Smaller particles (<20 $\mu m$ ) could not be
320	identified as jigsaw-fit due to voxel resolution. Particles 1 and 2 represent two fitting
321	particles with vesicle concavity in common, and their fit with part of core (labeled 3 and
322	visible as part of core in 3-D). H: Higher-magnification view of margin of sample HH47-
323	2 showing jigsaw-fit texture in ash rim at margins of vesicular core (highlighted in pink
324	solid line). Orange dashed line corresponds to vesicle concavity in common with several
325	ash particles as well as with core. Note high number and density and small size of cracks
326	in this area. I: Close-up of margins of sample HH28–3 showing cracks (red), ash rim, and
327	fillings.
328	
329	Figure 2. Jigsaw-fit textures. A–B: Backscattered-electron–scanning electron microscope
330	images of sample HH37–3 showing jigsaw-fit textures at margin of dense core, with (B)
331	four jigsaw-fit particles separated by cracks and sharing matching crystal fragments. C:
332	X-ray computed microtomography (XCT) volume of jigsaw-fit particles 1 and 2 in Figure
333	1G viewed from two different angles (left and right). Matching vesicle concavities are
334	highlighted in orange dashed circles. D–E: Two- (D) and three-dimensional (E)
335	visualizations of XCT data showing vesicle with radial crack (red) distribution in samples
336	HH47–2 and CAP372–2–3.
337	
338	Figure 3. Conceptual model of formation of ash-encased lapilli. A: At time t1, a
339	magmatic particle is ejected into water column and is in direct contact with water (in
340	blue). High cooling rate induces a high thermal gradient at margins of particle;

341	subsequent quenching and high levels of thermal stress trigger cracking at margins of
342	particle. Crack number and density are much higher at margins due to higher temperature
343	contrast between particle and coolant, causing in situ granulation and formation of ash
344	particles at outer parts of margins, showing jigsaw-fit textures. Thermal cracks are
345	represented in red. B: At time t2, inward displacement and rotation of ash particles
346	toward core induce ash filling of external vesicles and densification of rim. Some
347	outermost ash particles might also be spalled off after granulation and are released into
348	water column. Arrows represent both spalling of some external particles released to
349	plume and inward displacement of ash toward core during densification. C: At time t3,
350	once particle is well above water, residual heat in particle core leads to evaporation at
351	margins and subsequent salt precipitation, enhancing stability of ash rims when deposited
352	on land. Ash-encased particles can be easily identified with jigsaw-fit (highlighted in
353	blue), whereas margins of the core are outlined in pink. Green rectangles represent salts
354	binding ash particles in coating.
355	
356	<sup>1</sup> GSA Data Repository item 2019xxx, description of geological setting of study,
357	description of methods for thermal stress analysis, and discussion on effect of crystals on
358	crack propagation, and Table DR1 (scan conditions for ash-encased lapilli analyzed by

- 359 XCT), is available online at http://www.geosociety.org/datarepository/2019/, or on
- 360 request from editing@geosociety.org.