Magma fracturing and degassing associated with obsidian formation: The explosive–effusive transition

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ABSTRACT

This paper explores the role of melt fracturing in degassing rhyolitic volcanic systems. The Monte Pilato-Rocche Rosse eruptions in Italy evolved from explosive to effusive in style, and H2O content in quenched glasses changed over time from relatively H2O-rich (~0.90 wt.%) to H2O-poor dense obsidian (~0.10–0.20 wt.%). In addition, healed fractures have been recorded in different eruptive materials, from the glass of early-erupted tube pumice and rinds of brecciated obsidian pyroclasts, to the glass of late-erupted dense obsidian pyroclasts, and throughout the final effusive Rocche Rosse lava flow. These rocks show multiple fault sets, some with intraplated fault planes indicating resumption of viscous flow after faulting, complex obsidian breccias with evidence for post-brecciation folding and stretching, and centimetre- to metre-thick tuffites preserved in pyroclasts and lava, representing collapsed foam due to fracturing of vesicle walls. These microstructural observations indicate that multiple fracturing and healing events occurred during both explosive and effusive eruptions. H2O content in glass decreases by as much as 0.14 wt.% towards healed fractures/faults and decreases in stretched obsidian breccias towards regions of intense brecciation. A drop in pressure and/or increase in temperature along fractures caused diffusive H2O migration through melt towards fracture surfaces. Repetitive and pervasive fracturing and healing thereby create conditions for diffusive H2O loss into fractures and subsequent escape through permeable paths. This type of progressive magma degassing provides a potential mechanism to explain the formation of dense obsidian and the evolution from explosive to effusive eruption style.

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1. Introduction

Deposits from rhyolite volcanoes worldwide indicate that many eruptions evolve from explosive to effusive (e.g., Eichelberger and Westrich, 1981; Cortese et al., 1986; Dellino and La Volpe, 1995; Barclay et al., 1996; Lara, 2009; Watt et al., 2013). Recent observations of rhyolite eruptions at El Chaltén, and Puyehue–Cordón Caulle, Chile, follow the explosive–effusive pattern for rhyolite volcanoes, where Plinian explosive activity transitioned to lava effusion within a week (Castro and Dingwell, 2009; Castro et al., 2012b; Schipper et al., 2013).

The whole-rock major element composition of rhyolites extruded during explosive–effusive rhyolite eruptions is generally constant within the eruptive cycle (e.g., Gioncada et al., 2003; Pallister et al., 2013; Watt et al., 2013). Although there can be a relative increase in crystal content in the products of effusive compared to explosive eruptions (cf. Castro and Dingwell, 2009; Watt et al., 2013), commonly the main change is a decrease in volatile contents (H2O, CO2, F, Cl, Li, Be and S) that is recorded in matrix glasses (Newman et al., 1988; Westrich et al., 1988; Dunbar and Kyle, 1992; Barclay et al., 1996; Lowenstern et al., 2012).

H2O is the dominant volatile phase in volcanic melts, and dissolved H2O contents in erupted rhyolite glasses vary from ~2.2 wt.% in explosive products, to between 0.5 and 0.1 wt.% in effusive deposits (e.g., Eichelberger and Westrich, 1981; Westrich et al., 1988; Castro and Dingwell, 2009). Generally, the juvenile components of explosive rhyolite deposits are dominantly pumice, whereas obsidian (vesicle-poor glass) forms a small proportion of the deposits (e.g., Eichelberger and Westrich, 1981; Schipper et al., 2013). However, H2O-poor obsidian dominates amongst effusive deposits (Fink, 1980; Manley and Fink, 1987; Stevenson et al., 1994; Tuffen and Castro, 2009; Furukawa et al., 2010). Primary magmatic H2O dissolved in glass from pyroclastic obsidian and feeder dikes can vary widely from 2.2 wt.% down to values in equilibrium with atmospheric pressure (0.10 wt.%; e.g., Eichelberger and Westrich, 1981; Westrich et al., 1988; Castro and Dingwell, 2009; Watkins et al., 2012). In contrast, glasses from obsidian lava record a narrower range at low values (0.1–0.5 wt.%; Westrich et al., 1988; Castro et al., 2005; Clay et al., 2012; von Aulock et al., 2013).
The explosivity and style of volcanic eruptions are controlled by the ability of magmatic gas to escape as magma ascends and decompresses. Magma overpressure driving eruption is a function of the balance between gas exsolution and gas loss (e.g., Sparks, 2003). Permeable networks of connected bubbles and shear-induced brittle fractures are thought to allow gas migration and outgassing of the system (Eichelberger et al., 1986; Jaupart and Allègre, 1991; Stasiuk et al., 1996; Gonnernmann and Manga, 2003; Cabrera et al., 2011; Holland et al., 2011; Lavallée et al., 2013; Schipper et al., 2013). The critical vesicularity at which significant gas loss occurs varies due to complex relationships between porosity and permeability (Rust and Cashman, 2004), partly controlled by shear flow (Wright and Weinberg, 2009). Measurements of the permeability of natural, vesicular volcanic samples and laboratory analogs indicate that bubbles form a percolating network at porosities between 30 and 80 vol.%, depending on melt viscosity, crystallinity, magnitude of shear, and bubble expansion rate (e.g., Klug and Cashman, 1996; Saar and Manga, 1999; Mueller et al., 2005).

For the expected range of permeabilities in vesicular magmas (10^{-15} to 10^{-14} m^2; cf. Rust and Cashman, 2011), calculated degassing times are longer than the estimated timescale of magma ascent through shallow conduits (Gonnernmann and Manga, 2003; Takeuchi et al., 2008; Okumura et al., 2009). Slow degassing implies that another mechanism must explain the gas required to defuse explosive volcanic systems and give rise to effusive lava flows. Shearing experiments and measurements of natural vesicular rhyolite samples indicate that deformation of foams can greatly enhance sample permeability (Okumura et al., 2009; Wright and Weinberg, 2009; Okumura et al., 2010; Caricchi et al., 2011) and give rise to oriented, tube-like, bubble–bubble open paths, thought more typical of the relatively deep parts of conduit based on their presence in highly vesicular pumiceous pyroclasts (Blower, 2001; Wright et al., 2006; Okumura et al., 2009).

Torsional shear deformation experiments on vesicular samples have also shown that foams can sustain brittle fracture and degas (Okumura et al., 2010). However, these experiments also indicate that slip in the fractured interface prevents further brittle failure and shear-induced bubble coalescence. Healing of fractures results in resumption of viscous deformation (Tuffen et al., 2003), thus fracturing will only be a significant degassing mechanism if magma fracturing and healing events are common and pervasive (Okumura et al., 2010).

Different mechanisms have been proposed to explain the lack of preserved open permeable pathways in obsidian and the change from explosive to effusive behaviour in silicic volcanic systems. Vesicle collapse as well as fracturing and healing of melt, both induced by magma shearing in the conduit, can potentially explain: a) degassing, b) low porosity in eruptive products, and c) change in eruptive behaviour from explosive to effusive (Eichelberger et al., 1986; Gonnernmann and Manga, 2003; Castro et al., 2012a; Okumura et al., 2013; Schipper et al., 2013). Cabrera et al. (2011) measured H2O content across a healed fault in pyroclastic obsidian glass and demonstrated that fractures are directly related to degassing by providing low pressure and/or high temperature sites, which lead to diffusion of volatiles from the melt into the fracture, and provide a permeable path for gas escape.

In order to account for considerable degassing of melt and explain explosive to effusive transitions, fracturing and healing must be a pervasive and repetitive process (e.g., Gonnernmann and Manga, 2003; Rust and Cashman, 2007; Cabrera et al., 2011; Castro et al., 2012b). Repeated fracturing and healing of melt during eruptions have been demonstrated in several volcanoes, both in the conduit during ascent and in lavas (Stasiuk et al., 1996; Tuffen et al., 2003; Rust et al., 2004; Tuffen and Dingwell, 2005; Rust and Cashman, 2007; Tuffen and Castro, 2009; Tuffen et al., 2010; Castro et al., 2012b; Schipper et al., 2013). Estimated fracturing and healing times of obsidian glass are compatible with the time between earthquakes recorded during silicic lava eruptions (e.g., Tuffen et al., 2003; Yoshimura and Nakamura, 2010; Cabrera et al., 2011; Castro et al., 2012b).

Castro et al. (2012b) estimated the degree of melt fracture pervasive- ness required to defuse El Chaitén’s 2008 explosive eruption via diffusive water loss into fractures. They measured H2O concentration profiles in the matrix glass of pyroclasts (with evidence of pervasive fracture and healing) and lava. They found that H2O contents drop towards tuffite veins and towards the edges of clasts within tuffite veins. Modelled diffusion times that would account for these H2O drops are ~10^3–10^5 s, similar to modelled times for diffusion of H2O into healed faults in the rind of breadcrumb obsidian bombs on Lipari Island (Cabrera et al., 2011). The transition from explosive to effusive behaviour occurred within 10 days, therefore Castro et al. (2012b) argued that unless the magma at El Chaitén was fragmented into mm-size particles across the conduit, shear-induced fragmentation could only have accounted for enhanced degassing from magma near the conduit margins. However, the hybrid and concomitant explosive–effusive phases at Chaitén and Cordón Caille rhyolite eruptions in Chile, indicate the occurrence of shear-induced degassing processes at the scale of the conduit (Schipper et al., 2013). Schipper et al. (2013) proposed that during the Cordón Caille eruptions, areas of strain localisation and shear-induced magma-fragmentation were not restricted to conduit margins, but rather prevailed within the conduit through a network of branched shallow permeable zones that extended over more than 100 m depth and intersected highly sheared vesicular melt. These zones allowed degassing, where permeable gas escape from a foamed magma dominated over diffusive gas escape into fractures (Schipper et al., 2013). In this paper, we investigate the extent of fracturing and healing in eruptive products; the resultant effects on H2O content of melt; and infer relationships between fracture, degassing, and eruption style during the explosive–effusive Monte Pilato-Rocche Rosse sequences on Lipari Island.

2. The Monte Pilato-Rocche Rosse sequences

Rhyolite magmas in the Aeolian Islands have been erupting for the last ~55 thousand years (Donato et al., 2006) and over the last ~42 thousand years on Lipari Island. On Lipari, rhyolite eruptive activity is typically cyclic, beginning with an explosive phase and ending with extrusion of obsidian lava flows without a major change in chemical composition (Cortese et al., 1986; Gioncada et al., 2003). The rhyolite explosive and effusive Monte Pilato-Rocche Rosse sequences at the northeastern corner of Lipari Island (Fig. 1) represent the most recent volcanic activity on the island (Cortese et al., 1986; Dellino and La Volpe, 1995) and took place during the sixth century AD (e.g., Dellino and La Volpe, 1995; Lucchi et al., 2010).

The Monte Pilato sequence began with explosive activity forming a large pumice cone (Monte Pilato itself) and the smaller, coeval, parasitic pumice Lami cone on its southern flank (Fig. 1b; Cortese et al., 1986). A small lava body was then erupted in the main crater of Monte Pilato (Tranne et al., 2002). Explosive activity then resumed within the ~1 km wide Monte Pilato crater forming a small tephra cone (Fig. 1b) that corresponds to the Rocche Rosse sequence (Cortese et al., 1986; Dellino and La Volpe, 1995). This later eruption sequence ended with extrusion of the ~2 km long Rocche Rosse obsidian lava flow that covers the northern slope of the Monte Pilato pumice cone (Cortese et al., 1986) and extends into the sea (Gamberi and Marani, 1997). The small lava flow located below the Rocche Rosse tephra and above the Monte Pilato pumice, and an erosional surface located in the crater walls of the Monte Pilato pumice cone (Fig. 1b) separate the Monte Pilato and Rocche Rosse sequences (Dellino and La Volpe, 1995). Petrographic studies of the explosive and effusive products found that these are nearly aphyric containing only rare microlites of K-feldspar and pyroxene (Gimeno, 2003; Gioncada et al., 2003; Davì et al., 2009; Davì et al., 2010; Clay et al., 2012).

2.1. Eruptive products

Pyroclastic deposits from the Monte Pilato-Rocche Rosse sequences contain alternating layers of phreatomagmatic and purely magmatic
tephra (Dellino and La Volpe, 1995, 1996). This study focuses on the magmatic, juvenile products and ignores deposits significantly influenced by external water. The pyroclastic magmatic component is divided into several textural categories: white and grey tube pumice, obsidian clasts including breadcrust bombs, Pele’s tears, dense obsidian lapilli and bombs, and tuffitic obsidian (Fig. 2). Colour differences in tube pumice clasts are attributed to differences in density, where grey tube pumice has a higher density than white (Davì et al., 2011), rather than differences in crystal content or chemistry (Appendix A). The relative proportion of pyroclast types varies with different phases of the eruption. The earlier magmatic deposits related to the Lami cone include white tube pumice pyroclasts, Pele’s tear obsidian pyroclasts, and breadcrusted obsidian bombs, with variably fractured and healed glassy rinds (see section 4.2.1 below). The coeval and main explosive Monte Pilato phase includes white tube pumice, scattered dense obsidian, and highly vesicular obsidian pyroclasts. In contrast dense obsidian and tuffitic obsidian pyroclasts and grey tube pumice were produced by the last explosive phase that formed the Rocche Rosse tephra. Explosive deposits are overlain by the Rocche Rosse lava flow formed by obsidian and variably vesicular and devitrified textural types (e.g., Gimeno, 2003; Clay et al., 2012).

3. Samples and methods

Healed fractures are commonly present in glass forming both explosive and effusive deposits and representative hand samples of a range of textural types were collected. Clasts were cut and polished perpendicular to macroscopic discontinuities and thin sections prepared. Back-scattered electron (BSE) images were obtained on a JEOL 840A Scanning Electron Microscope (SEM) at the Monash Centre for Electron Microscopy, Monash University, Melbourne. The SEM was operated at 20 kV accelerating voltage, 15 mm working distance and a probe current of $1 \times 10^{-9} \text{A}$ (see Appendix A for details). Three samples (L01, RRT and Li07-64) were selected for detailed analyses of $\text{H}_2\text{O}$ content variations in groundmass glass in the vicinity of healed fractures. $\text{H}_2\text{O}$ contents of pristine glass in 23 samples from explosive and effusive deposits (Table 1) were further analysed using Fourier transform infrared (FTIR) spectroscopy (see below). In order to measure $\text{H}_2\text{O}$ contents in glass, chips from each sample were prepared into doubly polished wafers with parallel sides ranging in thicknesses from 77 to 1000 $\mu\text{m}$. The thickness of each wafer was measured with a micrometre with precision of $\pm 5 \mu\text{m}$. $\text{H}_2\text{O}$ contents were measured using benchtop FTIR and synchrotron FTIR (SFTIR) instruments.

Transmission spectra were obtained from obsidian glass in pyroclast interiors, bomb rinds, and from the Rocche Rosse lava flow using a Varian FTS 7000 FTIR Spectrometer at the School of Chemistry, Monash University, and Synchrotron Spectrometer at the Australian Synchrotron. The Monash University bench spectrometer and the Australian Synchrotron FTIR are equipped with an IR microscope and single point and Focal Plane MCT detectors.

In order to determine groundmass glass $\text{H}_2\text{O}$ concentrations of each sample, $\text{H}_2\text{O}_{\text{mol}}$ and $\text{OH}^-$ concentrations were measured using the single point MCT detector interfaced with a KBr beam-splitter. The aperture size was set between 60 and 220 $\mu\text{m}$. Between 5 and 10 spectra of
128 scans each were acquired, on different spots on each wafer using a white light source. Straightline baselines were used to calculate peak heights. Glass density was taken to be 2370 kg/m^3. H_2O concentrations were determined in two different ways: (1) from the intensity of the broad 3570 cm\(^{-1}\) peak at 3570 cm\(^{-1}\) as in Newman et al. (1986) and using an absorption coefficient of 80 L/mol cm\(^{-1}\) (Ihinger et al., 1994), and (2) from the intensities of the 4500 cm\(^{-1}\) and 5200 cm\(^{-1}\) absorption bands using the Beer–Lambert law (Castro et al., 2008; Tuffen et al., 2010) is herein adopted.

Where both the measured H_2O concentrations have standard deviations through different techniques. Average and standard variations of measurements for each analysed area of samples are noted in Table 1. Although measured H_2O concentrations have standard deviations of up to ±0.09 wt.% H_2O, measurements were not repeated in the exact same spot and do not calculate errors related to absorption coefficient and baseline fit; a conservative relative uncertainty value of ±10% (Castro et al., 2008; Tuffen et al., 2010) is herein adopted.

In order to display H_2O concentration variations in glass, 2D raw data array H_2O maps were converted into 2D or 3D images using Bruker Optus version 6.5 and Varian Resolution Pro 4.0 softwares. Each array map varies between one single tile (352 × 352 μm) and up to 5 horizontally or vertically aligned tiles (352 × 1056 μm, sample Li07-64). Furthermore, SFTIR H_2O transects were assembled into rectangular point H_2O maps using multiple single point measurements, within areas up to 80 μm wide and 1840 μm long. For methodology see Cabrera et al. (2011). Transects and measurements are spaced every 10 μm (sample RRT), or 40 and 60 μm (sample LO1). 128 scans for each spectrum were used and the background measured every 5 measurements. Blue and green colours represent areas where IR absorption peak heights for H_2O bands were minimum and yellow to red colours correspond to areas where maximum values were obtained.

### Table 1

H_2O content variations in obsidian glass. Lami and Mt Pilato sequences are the oldest, Rocche Rosse the youngest.

<table>
<thead>
<tr>
<th>Eruptive centre</th>
<th>Facies type</th>
<th>Sample</th>
<th>Detector</th>
<th>Zhang et al. method</th>
<th>σ (±)</th>
<th>3550 peak</th>
<th>σ (±)</th>
<th>n</th>
<th>4520/5230 peak height ratio</th>
</tr>
</thead>
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<td>Li01</td>
<td>Single point</td>
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\(^a\) Cabrera et al., 2011.

\(^b\) Gottsmann and Dingwell, 2001.

\(^c\) Davi et al., 2010.
4. Results

Here we summarize whole-rock chemistry and H2O concentrations of different types of eruptive rocks and then describe typical microstructures of these rocks that suggest multiple ways and times during the eruption in which melt underwent fracturing and healing. The final subsection describes H2O concentrations around healed fractures.

4.1. Whole-rock chemistry and H2O concentration

Whole rock compositions of juvenile material from explosive and effusive phases are within error of each other (Appendix A and Gottsmann and Dingwell, 2001; Gioncada et al., 2003; Davì et al., 2010; Cabrera et al., 2011; Davì et al., 2011; Clay et al., 2012). Samples are all rhyolitic in composition, and the glass contains an average of 74.4 ± 0.3 wt.% SiO2. Furthermore, both explosive and effusive juvenile clasts (excluding devitrified areas of the lava flow) are crystal-free with less than ~1 vol.% microlite needles of pyroxene and feldspar. Significant differences were found for the dissolved H2O concentration in glasses.

Carbon dioxide concentrations, in all cases, were below detection. Fig. 3). Carbon dioxide concentrations, in all cases, were below detection.

4.2. Microstructures in fractured and healed obsidian

Fracturing of obsidian is documented in the different types of pyroclasts and lava flow. Healing tends to erase evidence for fracturing, but displacement of pre-existing features across the fracture plane, together with dusty particles on the plane, makes fractures obvious.

4.2.1. Breadcrusted and tuffitic obsidian pyroclasts

A breadcrusted obsidian bomb from the Lami cone shows a single planar micro-fault in its rind. The fault truncates folded micro-vesicle and microlite-rich bands (~1% needles of very thin and <100 μm long pyroxene) and pristine glassy bands (Fig. 4a, b; Cabrera et al., 2011).

Single planar faults are also present in dense tuffite (clastic material) glass from pyroclasts in the Rocche Rosse tephra. For example, sample RRT contains a healed fault that offsets tuffite bands (Fig. 4c-e). The fault trace is highlighted by a dusty brown colour due to a concentration of small <10 μm opaque microlites (Fig. 4e). The fault trace is crenulated and crenulations are asymmetric, which combined with drag folding of banding on the glass on either side of the fault, indicates sense of movement and that the glass resumed its viscous behaviour after faulting (Fig. 4e).

Two types of tuffite are present within obsidian pyroclastic samples from the Rocche Rosse tephra deposits: vesicular tuffite and banded tuffite. Vesicular tuffite contains narrow tuffite bands in pyroclasts with both dense obsidian and pumiceous domains. In the sample in Fig. 5, vesicular tuffite bands cut across a block-sized pyroclast. The bands are up to 1 cm wide and filled with grey, clastic glassy matrix <1 mm in particle size surrounding elongated black obsidian clasts that vary in length from millimetres to 1 cm. In some cases, obsidian clasts are broken up into fragments with a jig-saw fit pattern (Fig. 5c). These dense clasts are oriented parallel to tuffite band margins and have asymmetric shapes indicative of shearing, marked by curved foliation in the fine matrix wrapping around the clasts (Fig. 5a). Both dense and pumiceous textural domains surround the tuffite bands. However, dense veneers around pumiceous domains and separating pumice from tuffite suggest that vesiculation in pumiceous domains occurred after tuffite formation (Fig. 5a,b).

In contrast, banded tuffite (Fig. 6) forms disconnected lenses or irregular and connected grey bands in hand specimen (Figs. 4c and 6a) separated by lenses or bands of clean, microlite-free obsidian (black in hand specimen) or jigsaw fit angular clasts (Fig. 6d). Banded tuffite lacks vesicular domains. Tuffite in these clasts is comprised of brown glass, rich in opaque microlites and small clasts of transparent glass.

![Fig. 3. H2O content of samples from the different eruptive centres corresponding to the Monte Pilato-Rocche Rosse sequences and related textural product types. Letters and numbers on horizontal axis are sample labels, vertical lines separate eruptive centres and tephra vs. lava from the same eruption. *= FTIR H2O values from Cabrera et al. (2011); ** = FTIR H2O values from Gottsmann and Dingwell (2001); and *** = FTIR H2O data from Davì et al. (2010).](image-url)
The edges of each transparent glass lens are well-defined by concentrations of opaques, whereas interiors are clean. Both glass types form lenses with their longest axes varying in length from ~50 μm to ~3 mm. The distribution of lenses in Fig. 6 is complex in detail. In some areas, lenses of brown dusty glass are surrounded by transparent glass (Fig. 6b). In other areas, the opposite is true (Fig. 6c). In Fig. 6d, angular transparent clasts are hosted by brown glass matrix, which in turn forms a separate clast within transparent obsidian. Commonly, the two types of glass are found as long, thin bands that show complex folding structures (Fig. 6e). As exemplified by the sample in Fig. 6, the definition of matrix and clast in banded tuffsite depends on scale. The two glass types are mingled at various scales, fractured, folded, and stretched into long lenses (see Section 5).

4.2.2. Obsidian lava flows

Obsidian from the Rocche Rosse lava flow commonly exhibits multiple micro-fault sets showing both planar and crenulated faults (Fig. 7). The northwestern edge of the Rocche Rosse lava flow front is also cut by subplanar breccia zones (Fig. 8). Road cut exposures demonstrate that the breccia bands persist for at least ~20 m into the lava body and maintain a consistent ~N350/30E orientation. Breccia bands are ~1 m wide and continuous for up to 6 m (Fig. 8a) near the flow margin.
Towards the interior of the lava, bands narrow, varying from ~3 cm to ~40 cm wide, and are continuous for up to 2 m. The bands consist of angular to elongated and welded clasts in a finer clastic glassy matrix bordered by localised bands of dark blue to brown banded obsidian concentrated near the breccia zone walls. Brecia contents are similar in composition and texture to tuffites in pyroclasts, although clasts reach larger sizes in breccia bands than in tuffites. Clasts within breccia bands are generally oriented parallel to fracture walls and vary in size from lapilli to block-size (Fig. 8b). Clasts are variably vesicular and variably devitrified. Abrasion of breccia margins during fracturing of the lava is evidenced by preservation of different stages of break-up of wall clasts (Fig. 8b). Doubly polished wafers of the obsidian matrix reveal high aspect ratio vesicles (>100 μm long and <10 μm across, Fig. 8c) defining foliations that are truncated by sharp fault surfaces (Fig. 8d) and flow folds (Fig. 8e).

4.3. Microstructures in fractured and healed pumice

Pumice pyroclasts show distinct domains characterized by vesicles of different orientation or aspect ratio in 2D sections (Fig. 9). Boundaries between these domains may be more or less sharp (compare differences in Fig. 9). Fig. 9c shows three bands of strongly elongated vesicles forming tube pumice (top, middle and bottom) interlayered with two bands of moderately to weakly elongated vesicles with stair-stepping foliation, containing kernels where vesicles are roughly circular. Banded microstructures are interpreted to represent zones of high strain interlayered with zones of low strain. The asymmetry of the foliation in Fig. 9c indicates shear in a top-to-the-left direction.

4.4. H2O distribution around fractures

SFTIR and FTIR 3D maps of H2O concentrations across healed fractures in pyroclastic and effusive obsidian define V-shaped troughs of low H2O content coincident with fracture planes (Fig. 10). In pyroclastic obsidian sample L01-Lami, H2O varies from 0.66 and 0.73 wt.% on either side of the fault trace, to a low of 0.61 wt.% at its trace (Fig. 10a and b). Single point FTIR H2O content measurements on either side of healed fractures in different areas of the same sample show a broadly similar pattern. Similarly, the trace of healed faults in the glass from the late Rocche Rosse lava flow sample Li07-64 in Fig. 7 defines H2O content troughs (Fig. 10c–f). Interestingly, H2O profiles in the vicinity of the two faults are similar and decrease to a low of 0.12 wt.% in the fault plane (Fig. 10d–e).

Fig. 11 shows a SFTIR H2O transect that crosses both the brown, dusty tuffite and the clean, microlite-free obsidian bands in a banded tuffite. As is common, the brown glass hosts smaller and elongated clasts of transparent obsidian (Fig. 6). H2O content decreases gradually from 0.34 and 0.29 wt.% in the transparent and clast-free obsidian lenses, to 0.26 wt.% at the centre of the clast-rich brown obsidian breccia.

5. Discussion

The Monte Pilato-Rocche Rosse sequence evolved from explosive to effusive eruptions and the products of eruption include obsidian with variable dissolved H2O contents reaching very low values, and include healed fractures at many scales (e.g., Tuffen and Dingwell, 2005). In this section, we discuss evidence for fracture and healing processes and degassing during fracturing. Our results support models suggesting that fracturing of magmas in conduits can explain both the formation of volatile-poor, vesicle-free obsidian (Yoshimura and Nakamura, 2010; Cabrera et al., 2011) and the decrease in explosivity during a typical eruptive cycle (e.g., Gonnermann and Manga, 2005).

5.1. General features of obsidian in the Monte Pilato-Rocche Rosse sequences

5.1.1. Repeated fracturing and healing

Like high-silica rhyolite glasses erupted elsewhere, obsidian from the Monte Pilato-Rocche Rosse explosive eruptions is richer in H2O than the effusive counterpart (Fig. 3). Given similar whole-rock major element compositions of explosive and effusive products (Appendix A), it is most likely that magmatic volatile content controlled the change in eruption style (e.g., Newman et al., 1988; Villemant and Boudon, 1998). Microstructural features documented above indicate that melts forming the eruptive Monte Pilato-Rocche Rosse products underwent multiple brittle–ductile transitions during emplacement and that degassing due to fracturing may have played an important role in the decreasing explosivity of the eruptive sequence.

Evidence for multiple brittle–ductile transitions in a single sample is demonstrated by healed and crenulated fault planes in pyroclasts (e.g., Fig. 4). This indicates that the melt resumed viscous flow after faulting, leading to crenulation (Fig. 4e). Repeated crossing of the glass transition in this manner implies a strain-rate dependent rheological transition (as opposed to strictly temperature-dependent transition). We infer that strain rate variations thereby led to in-conduit brecciation, as recorded by vesicular and banded tuffites (Figs. 5 and 6). Vesicular tuffite (Fig. 5) marks brecciation and local vesiculation interrupted by ejection. Banded tuffite (Fig. 6) records multiple brecciation events followed by resumption of viscous flow, giving rise to a “stretched and folded” and rather cryptic breccia in the conduit. Similarity to microstructures from other rhyolite deposits suggests that multiple events of silicic magma fracturing and healing might be a general process (Rust and Cashman, 2007) and that these could result in typical flow banding characteristic of obsidian (Tuffen et al., 2003; Tuffen and Dingwell, 2005) as supported by models (Gonnermann and Manga, 2005).
Brecciation is not restricted to non-vesicular regions of melt; vesicular material (foams) also experienced multiple fracture events. Pumice pyroclasts have line-sharp planes that separate distinct micro-structural domains, where domains vary in vesicle elongation direction and aspect ratio (tube pumice in Fig. 9a–b and d) or in degree of strain (banded pumice with asymmetric vesicles, Fig. 9c; Wright and Weinberg, 2009). These microstructures could have formed in two ways: a) fragmentation of foam into individual clasts and subsequent healing in the conduit (Fig. 9a–b and d) (Tuffen et al., 2003; Tuffen and Dingwell, 2005), or b) strain localization into narrow shear zones where stresses exceed the yield strength for foams (Fig. 9c; yield behaviour is characteristic of foams, e.g., Weaire, 2008). These microstructures in tube pumice pyroclasts on Lipari (see also Wright and Weinberg, 2009) are comparable to those in pumice clasts from the Cordón Cauquil, Chile 2011–2012 eruptions (Schipper et al., 2013), Vesuvius 79 A.D. (Shea et al., 2012), and to those experimentally reproduced from sheared silicic foams (Okumura et al., 2010). Brecciation and healing or strain localization in silicic foams with yield strength may therefore be general features of eruptive activity (Okumura et al., 2010; Castro et al., 2012b; Shea et al., 2012; Schipper et al., 2013). In-conduit brecciation of pumice may represent events where the fragmentation threshold of the vesicular magma was exceeded locally but not globally, and thereby was insufficient to produce an explosive eruption (cf. Spieler et al., 2004).

Evidence for fracturing and healing is also present in the Rocche Rosse lava flow. Here too, obsidian from the lava flow records early crenulated and healed micro-fractures displaced by sets of later faults (Fig. 7) indicating a crossing of the glass transition at least twice. At
the macro-scale, parts of the lava fractured into metre-long clasts that were included in the flowing magma within breccia planes (Fig. 8b). The obsidian matrix in these subplanar breccia zones has microstructures suggestive of rapid emplacement, such as the strongly elongated vesicles (Fig. 8c), folding of obsidian bands (Fig. 8e), as well as fracturing and healing (Fig. 8d).

5.1.2. H2O loss associated to melt fracturing

In the previous section, we discussed microstructural evidence for multiple fracturing and healing events during conduit flow and extrusion, and of both highly vesicular (Figs. 5 and 9) and dense (Figs. 4 and 6) magma. Melt fracturing is a general feature. In this section we discuss evidence that fracturing is accompanied by degassing.

The link between faulting of glass in the conduit and H2O loss has been documented here and in other studies (Cabrera et al., 2011; Castro et al., 2012b; Shea et al., 2012; Watkins et al., 2012). Profiles presented here show H2O loss of up to 0.14 wt.% towards single fractures in pyroclasts and lava samples (Fig. 10) as well as tuffsite bands in the stretched breccias formed in the conduit (Fig. 11). H2O troughs across healed faults indicate that H2O escaped while fractures were still open and hot (Fig. 10a–b). The drop in water content indicates that fracturing gave rise to low pressure or local increase in temperature, possibly due to friction, causing H2O diffusion from the glass into the fracture (Cabrera et al., 2011). Pressure or temperature inside the fracture cannot be gauged accurately from the H2O content curve, because P–T could fluctuate over time scales too short to allow H2O content equilibration, and because H2O content changes during fracturing will be at least partly homogenized by post-healing H2O diffusion (Yoshimura and Nakamura, 2010; Cabrera et al., 2011). Fractures remain open for an undetermined time, which depends upon relative pressure within the fracture, translation and rotation of fragments, and timescale to heal juxtaposed surfaces, which is itself dependent upon temperature and roughness of the fracture surface (Yoshimura and Nakamura, 2010). However long the fractures remained open, the

Fig. 8. a) Subplanar breccia zones marked by dashed lines in the flow front of the Rocche Rosse lava flow (2 km away from vent, in the vicinity of Aquacalda). Breccias are composed of welded clastic material derived from abrasion of the walls. Clasts are elongate and irregular in shape. Rectangle indicates location of (b). b) Large (~1.5 m long) devitrified obsidian clast at the wall of the breccia zone broken in situ into smaller clasts. c–e) Photomicrographs (plane polarized light) of obsidian matrix. White (transparent) is glass, black is oxides. Arrow in (c) points to an elongated vesicle (in 2D). Small dark ellipses are high aspect ratio micro-vesicles and thin brown lines are concentrations of elongate micro-vesicles in 2D. Note truncation planes in (d) and tight folds in (e) indicated by white dashed line.
presence of crenulated healed faults (Figs. 4e and 7) suggests that the glass was sufficiently hot to flow after fracturing.

Decrease in H$_2$O content of glass across banded tuffsite from the Rocche Rosse tephra (Fig. 11) is continuous from the centre of a nonvesicular obsidian domain to the most intensely brecciated clastic band (from left to right in Fig. 11). We suggest that this H$_2$O profile is the result of H$_2$O loss during syn-eruptive brecciation in the conduit, with maximum loss where fracturing was most intense, as marked by fine clastic material (see also Rust and Cashman, 2007; Stasiuk et al., 1996; Tuffen et al., 2010). The H$_2$O profile has been smoothed after healing by diffusion while the material was still hot (cf. Castro et al., 2005; Yoshimura and Nakamura, 2008 and 2010). Castro et al.
(2012b) found that glass of vesicular tuffsite pyroclasts from Chaiten also has maximum H$_2$O loss linked to fractured areas (e.g., Fig. 3b in Castro et al., 2012b). Similar to our results, Rust and Cashman (2007) found H$_2$O variations of up to 0.25 wt.% amongst healed clasts in welded breccia from pyroclastic deposits at Newberry Volcano. They interpreted that shearing and flowing of rising magma could have brought together clasts from different areas of the conduit.

5.2. Melt fracturing, degassing and decreasing explosivity of rhyolite eruptions

Degassing through fractures inhibits explosive fragmentation by inhibiting the formation and growth of bubbles (Yoshimura and Nakamura, 2010), increasing the pressure difference necessary for explosive fragmentation (Spieler et al., 2004).
chemical signatures described here indicate that degassing due to fracturing is pervasive during silicic eruptions and has the potential to be a defusing mechanism for rhyolite volcanoes (e.g., Gonnermann and Manga, 2003).

A similar scenario to Cordón Caulle in Chile, may have occurred in the Monte Pilato-Rocche Rosse conduit, whereby non-explosive magma fragmentation develops branched fractures that intersect variably vesicular magma in the conduit and leads to efficient degassing (Schipper et al., 2013). In other words, in this case fracturing events will not only release water dissolved in melt but also have the potential to release gas trapped in bubbles, leading to vesicle collapse and forming tuffites such as those documented above.

The evolution of rhyolitic eruptions from explosive to effusive may be summarized by focusing on the difference between the total volatile content of the magma, including H2O in vesicles and dissolved in the melt ($X_{\text{tot}}$), and the H2O content that the melt can dissolve at equilibrium ($X_{\text{eq}}$) for given P-T conditions. This can be considered a measure of the magma potential to erupt explosively ($W = X_{\text{tot}} - X_{\text{eq}}$). For a volume of melt that ascends and vesiculates, the total H2O content will remain approximately constant if outgassing is inefficient, increasing its potential to decompress explosively ($W$ increases). High-permeability fracturing events (e.g., Gonnermann and Manga, 2003) resulting from magma shearing against conduit walls or shock waves due to local pressure differences exceeding the threshold for fragmentation of vesicular melt, allow for a step-wise drop in that potential, and equilibration of the magma, decreasing explosivity ($W$ decreases).

Extensive fracturing documented in Lipari samples, including the pervasive fragmentation in obsidian breccias and tuffites (Figs. 5 and 6) and pumice breccias (Fig. 9), combined with development of H2O-poor obsidian, suggest that fracture degassing may have played a significant role in defusing the intensity of the rhyolite eruption.

6. Conclusions

Monte Pilato–Rocche Rosse eruptions ejected rhyolite pyroclasts with relatively high and variable dissolved H2O contents followed by

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**Fig. 11.** H2O variations in banded tuffite obsidian from the Rocche Rosse pyroclasts: a) Wafer 92 μm thick from sample RRT. The rectangle highlights the area of two parallel synchrotron FTIR H2O transects. Each transect (A to B) and measurement points along the transects are spaced at 10 μm. White is obsidian glass, grey is welded tuffite. b) H2O profile for transect A–B with calculated H2O content values (average between the two parallel transects). H2O content decreases gradually from A to B. The H2O content in the clast-free obsidian band at A is ~0.08 wt.% higher than the clast-rich band near B. Standard deviation for H2O content at each point is <0.002 wt.%. Dashed vertical lines separate boundaries between bands free of clastic material and bands rich in clasts.
effusion of H₂O-poor obsidian lava. During these eruptions, fracturing and healing of the high viscosity magmas is responsible for a number of different microstructures, ranging from simple planar faults, multiple cross-cutting fault sets, crenulated fault planes, and stretched out and complex obsidian and pumice breccias. These features indicate that fracturing was a common, repetitive and pervasive process in the conduit. This study also demonstrates that fracturing leads to diffusive H₂O loss from the melt, and that repeated fracturing events lead to high permeability gas loss events. Fractures not only capture newly exsolved volatiles, but also provide escape paths for volatiles trapped in vesicles. Thus, syn-eruptive degassing at depth through a combination of vesiculation and extensive fracturing could explain decreasing H₂O content from early explosive pyroclasts to late effusive lava flow. This process may be critical in defusing explosive potential of rhyolitic magmas.

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Appendix A. Supplementary data

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References
