Constraints from Phase Equilibrium Experiments on Pre-eruptive Storage Conditions in Mixed Magma Systems: a Case Study on Crystal-rich Basaltic Andesites from Mount Merapi, Indonesia

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ABSTRACT

Mount Merapi is one of Indonesia’s and the world’s most hazardous volcanoes. Existing constraints on the location and the crystallization conditions of its pre-eruptive magma reservoir based on mineral and glass thermobarometry and geophysical surveys are inconclusive, yet of immediate importance for future hazard mitigation. Here, we use two series of phase equilibrium experiments to quantify the conditions of pre-eruptive magma storage and magma recharge in Merapi’s upper-crustal reservoir: (1) to characterize crystallization at total equilibrium conditions; (2) to characterize crystallization at local equilibrium conditions between crystal rims and host melt. We demonstrate that this experimental approach can constrain pre-eruptive crystallization conditions and their variation in crystal-rich, mixed magma systems, such as that of Merapi, for which standard crystallization experiments and mineral thermobarometry fail. In agreement with geophysical estimates, we infer that Mount Merapi’s pre-eruptive reservoir partially crystallizes at \( P \geq 100–200 \) MPa and thus at relatively shallow depths of \( c. 4–5 \) to \( 9 \) km. Magmas are stored at \( T \geq 925–950 \) \(^\circ\)C with a melt \( H_2O \) content of \( 3–4 \) wt % and a vapour phase with an \( X_{H_2O} \) of \( 0.5–0.6 \). Pre-eruptive recharge magmas, in contrast, have a temperature \( T > 950 \) to \( < 1000 \) \(^\circ\)C, a higher melt \( H_2O \) content of \( 4–5 \) wt % and a vapour phase with a higher \( X_{H_2O} \) of \( 0.8–1 \). We hypothesize that these variations in melt \( H_2O \) content and vapour \( X_{H_2O} \) between resident and pre-eruptive recharge magmas relate to variable degrees of open-system degassing of magma parcels en route into the reservoir rather than to variations in volatile fluxing of the stored magmas.

Key words: \( H_2O; CO_2; \) crystal-rich; experiment; magma mixing; Merapi; volatiles

INTRODUCTION

Constraining pre-eruptive magma storage conditions is of immediate importance for assessing hazard potential and hazard types at active volcanoes (Woods, 1995; Sparks, 2003; Scaillet et al., 2008; Ruprecht & Bachmann, 2012; Cashman & Sparks, 2013), yet this is rarely achieved. Mount Merapi in Indonesia, one of the world’s most hazardous volcanoes, is a prime example, for which pre-eruptive magma storage conditions are poorly constrained. The volcano rises above densely populated countryside and is located \( \sim 30 \) km from the metropolis of Yogyakarta. On average, Merapi erupts...
every few years. Its historical eruptions have been
dominated by periods of lava dome growth and even-
tual gravitational dome collapse that typically produce
VEI1 to VEI3 pyroclastic flows and surges (Voight et al.,
2000; Charbonnier & Gertisser, 2008; Gertisser et al.,
2011). These flows clearly affect the local population,
but proficient hazard management has limited the num-
ber of fatalities. However, explosive VEI4 eruptions that
affect a much larger area and population occasionally
take place, as in 2010, and the rock record shows that
such explosive events were more common in prehis-
toric times (Andreastuti et al., 2000; Camus et al., 2000;
Gertisser et al., 2011). For future hazard management it
is thus imperative to (1) closely monitor the current pre-
eruptive magma reservoir and (2) assess whether
highly explosive eruptions will remain the exception,
or whether they may become more common.

Geophysical surveys—including seismic, ground de-
formation, and magnetotelluric studies—have inferred
shallow magma storage zones at depths of ~1.5–2.5 km
and at >5 km to ~8.5 km below Merapi’s summit; how-
ever, the shallowest zones may represent the presence
of brines rather than magma storage zones (Ratdomopurbo
& Poupinet, 1995, 2000; Beauducel & Cornet, 1999; Müller
& Haak, 2004; Budi-Santoso et al., 2013). Petrological studies and mineral barometry have
suggested a wide range of possible crystallization pres-
sures: (1) 0 to >600 MPa and predominantly ~100–
400 MPa or ~400–500 MPa for clinopyroxene (Gertisser,
2001; Chadwick et al., 2013; Preece et al., 2014); (2)
>300 to ~900 MPa and predominantly ~400–500 MPa
for amphibole (Costa et al., 2013; Nadeau et al., 2013).
Glass inclusion hygrobarometry and the evolved com-
positions of plagioclase and clinopyroxene indicate lim-
ited shallow magma storage at ≤300 MPa (Costa et al.,
2013; Nadeau et al., 2013; Preece et al., 2014). In combi-
nation, these calculated pressures suggest crystalliza-
tion throughout the crust and upper mantle, with major
magma crystallization zones at depths of ~4–18 km.

Estimates for crystallization temperatures in the magma
storage reservoir range widely from ~950°C using
amphibole thermometry (Costa et al., 2013; Erdmann
et al., 2014) to ~1050°C using clinopyroxene thermom-
etry (Preece et al., 2014). The limited data available indi-
cate pre-eruptive melt H₂O contents between ~3.5 and
5.0 wt% (Borisova et al., 2013; Preece et al., 2014).

The aim of our study is to provide experimentally
constrained estimates for the pre-eruptive crystalliza-
tion conditions and processes in Merapi’s current pre-
eruptive magma storage reservoir and, in particular,
constraints on the crystallization pressure and melt
volatile content, which are key hazard factors. We antici-
pat ed that previous estimates of crystallization pressure
and thus the location of the reservoir could be improved
upon, given that they were inferred on the basis of
amphibole and clinopyroxene mineral barometers with
large uncertainties (Fig. 1).

By mimicking the crystallization conditions of natural
magma systems, experimental studies can in principle
provide highly accurate estimates for all intensive crys-
tallization parameters. The challenge is, however, to
constrain the crystallization conditions for mixed
magma systems for which end-member magma com-
positions are not preserved in the rock record
(Pichavant et al., 2007; Blundy & Cashman, 2008).
Merapi’s crystal-rich, compositionally homogeneous
basaltic andesites have been interpreted to represent a
thorough mixture of relatively evolved resident and
more primitive recharge magmas (Nadeau et al., 2010,
2013; Chadwick et al., 2013; Costa et al., 2013), and thus
they provide an excellent case study to address this
challenge. To constrain the conditions in Merapi’s pre-
eruptive reservoir experimentally we needed to identify
the natural equilibrium mineral assemblages and the
crystallization conditions for the resident and the re-
charge magmas, respectively. To achieve this objective,
we have (1) characterized the mixed mineral assem-
blages and their compositions in the eruption products
of the paroxysmal 2010 eruption of Merapi and (2) per-
formed two series of phase equilibrium experiments to
quantify their record of crystallization conditions and
processes during both magma storage and recharge.

SAMPLES, METHODS, AND DATA

The 2010 eruption of Merapi was its largest explosive
event since 1872. For this VEI4 event, eight eruption
stages have been distinguished by Komorowski et al.
(2013): stage 1, precursory unrest; stage 2, volcanic or
phreatomagmatic explosions; stage 3, rapid dome
growth and volcanic explosions; stage 4, paroxysmal,
full dome and upper conduit explosion; stage 5, retro-
gressive old dome collapse; stage 6, convective column
fountain collapse; stage 7, rapid dome growth and ash
venting; stage 8, continuous ash venting. The stage 7
dome has remained largely intact since its emplace-
ment in 2010, although ash plumes and small volumes
of dome clasts were emitted from it in July and
November 2013 and in March and April 2014.

We have studied 25 samples of the 2010 eruptive
dome-forming event including the following: (1)
black, dense and glassy to scoriaceous dome clasts of
stage 4; (2) white pumice of stage 6; (3) light to medium
grey, dense to scoriaceous clasts derived from the
terminal stage 7 dome. The stage 4 + 6 samples were
collected by us in 2010 in Kali Gendol and near the tourist
site at Mariam Bunker. The stage 7 samples were col-
lected by S. Byrdina at Pasar Bubar on 19 November
2013, immediately following their eruption in a small
explosion that occurred on 18 November 2013 through
the dome that was emplaced on 6–8 November 2010
during stage 7. The stage 4 and stage 6 samples were
selected because they represent the main erupted
magma volume and the most explosive eruption stage,
respectively. The stage 7 samples were studied because
they have not been previously characterized; they rep-
resent the final magma emplaced during the 2010 erup-
tion cycle, and we anticipated that they may yield
additional insight into the previously proposed, but cryptic pre-eruptive recharge of Merapi’s upper magma system (e.g. Costa et al., 2013). We are confident that the samples classified as stage 7 are derived from Merapi’s terminal 2010 dome, because they have abundant calcic (≥An60–70) plagioclase microlites that are characteristic for the magmas of the 2010 eruptive event, but are unknown from other recent eruption products (Costa et al., 2013; Preece et al., 2013). Magnetite phenocrysts in these samples show trellis-like exsolution, which is consistent with slow cooling (e.g. Turner et al., 2008) during magma ascent and/or dome residence, but they show no or minor oxidation.

In the following analysis we distinguish between stage 4 + 6 and stage 7 samples, based on variable proportions of evolved and inclusion-rich versus primitive and inclusion-poor phenocrysts. The dome clasts and pumice samples of stage 4 + 6, however, have phenocryst populations with comparable textures and compositions, and they are thus described together.

Five selected whole-rock samples (one stage 4 dome clast, four stage 7 dome clasts) were geochemically
analyzed at the Regional Analytical Centre at Saint Mary’s University, Canada, and 25 natural samples were studied in thin section. For our experimental study, we used one of the black, glassy, dense basaltic andesite stage 4 dome clasts (sample SEM043b) as the starting material (Table 1; Fig. 2). We have selected this starting material for the following reasons: (1) the sample represents the main clast type that was discharged in the high-volume pyroclastic flows on 5 November 2010 [i.e. it is a Type 1 clast following the classification of Charbonnier et al. (2013), which make up >61 to 84 vol. % and on average 77 vol. % of all stage 4 pyroclasts]; (2) the sample has a whole-rock composition close to the average of all juvenile pyroclasts erupted in 2010 (Fig. 2); (3) the sample has a phenocryst and microphenocryst assemblage of plagioclase–clinopyroxene–orthopyroxene–amphibole–magnetite, which is characteristic of Merapi basaltic andesites (Fig. 3a and b); (4) the sample has mineral rim compositions similar to those of other stage 4 pyroclasts and stage 6 pumice samples investigated (Figs 2 and 4c, g, k; Supplementary Data Electronic Appendix A2; supplementary data are available for downloading at http://www.petrology.oxfordjournals.org).

For one series of experiments, referred to as ‘melting experiments’, we have used a gently crushed powder of the crystal-rich basaltic andesite whole-rock, which was partially melted and partially recrystallized during the experiments. For the second series of experiments, referred to as ‘crystallization experiments’, we have fused the same rock powder to a homogeneous glass at atmospheric conditions, which was then partially crystallized during the experiments. The melting experiments were performed to characterize the local equilibrium compositions of crystal rims and their host melts for Merapi’s crystal-rich basaltic andesites (i.e. accounting for the fact that crystal cores of Merapi’s mixed mineral assemblage are not in equilibrium) (Table 2). The crystallization experiments were performed to provide phase compositions that form at equilibrium for the basaltic andesite composition (Table 2), which guided our interpretation of the mineral rim compositions detected in the melting experiments. For both experimental series, Au capsules were first loaded with variable amounts of ultrafiltered, deionized H2O and silver oxalate (Ag2C2O4 → CO2) to yield equilibrium with a vapour phase with an XH2O [H2O/(H2O + CO2)] between ~0.4 and 1 (Table 3), and then filled with the starting rock or starting glass powders at a fluid/solid ratio of ~1:10 to impose volatile-saturated conditions as in Merapi’s mid- to upper-crustal magma system (see Nadeau et al., 2010, 2013).

The two series of experiments were run simultaneously in internally heated pressure vessels at the Institut des Sciences de la Terre d’Orléans (ISTO), France. The oxygen fugacity (fO2) was controlled by loading ~1 bar of H2 into the vessels, except for one experiment that was performed at the intrinsic fO2 of the vessel at ~NNO + 2 (where NNO is the nickel–nickel oxide buffer) (CE-1, Table 2). The fO2 was monitored by a pair of NiO + Ni-Pd solid sensors (see Pownceby & O’Neill, 1994), which indicate that the fO2 of the experiments with H2 added reached ~NNO + 1.5 to ~NNO + 1.3 in charges with an XH2O = 1, and the fO2 was ~NNO + 0.9 to ~NNO + 0.7 in charges with an XH2O of ~0.6–0.5 (Table 3). The experimental pressure and temperature were then induced by loading Ar into the vessel, followed by heating at a rate of 10–15°C min−1. At the beginning of each experiment, we overstepped the targeted experimental temperature by ~40°C for 2–3 h to promote partial melting and subsequent crystallization of mineral rims that were wide enough for analysis. All experiments were run for ~3 days at 925–1000°C and ~100–400 MPa (Table 3), and ended by drop quench. Temperature and total pressure were continuously monitored; their uncertainties are ± 5°C and ± 2 MPa.

For the natural samples, we distinguish between phenocrysts (>500 μm), microphenocrysts (>100–500 μm) and microlites (>500 μm). For the melting experiments, we distinguish between unreacted crystal cores and newly crystallized inner and outer mineral rims. Mineral proportions in the experimental and natural samples were determined by point counting on backscattered electron images and thin sections, respectively. Mineral, matrix and glass compositions were determined by electron microprobe at ISTO. Average and representative mineral and glass compositional data for the natural samples and the experimental charges are provided in Supplementary Data Electronic Appendices A2 and A3. For the natural samples we determined the phenocryst core and rim compositions for 10 samples and the matrix

| Table 1: Composition of whole-rock (WR) and glass (GI) starting material normalized to 100 wt % |
|---|---|---|---|---|---|---|---|---|---|---|
| St. mat. | SiO2 | TiO2 | Al2O3 | Fe2O3 | MnO | MgO | CaO | Na2O | K2O | P2O5 | Total |
| WR* (100 wt %) | 55.11 | 0.78 | 19.20 | 7.69 | 0.21 | 2.38 | 8.39 | 3.79 | 2.13 | 0.32 | 98.11 |
| GI† (100 wt %) | 57.19 | 0.69 | 18.32 | 6.86 | 0.12 | 2.36 | 8.28 | 3.81 | 2.12 | 0.25 | 98.91 |

St. mat., starting material sample SEM043b.
*Analyzed by XRF at Saint Mary’s University, Canada.
†Pre-normalized totals.
‡Analyzed by electron microprobe at ISTO, France.

The ~11% lower Fe2O3 content determined for the glass as compared with the whole-rock composition may partly reflect Fe loss during glass fusion. Apparent deviations in other element concentrations (e.g. ~4 and 5% for SiO2 and Al2O3) between WR and GI starting materials are considered to reflect analytical and method-related errors.
and glass inclusion compositions for six samples. For the crystallization experiments, we determined mineral and glass compositions for key charges. For the melting experiments, we determined mineral rim and glass compositions whenever they were large enough for analysis. The electron microprobe operating conditions were set at 15 keV and 10 nA with peak and background counting times of 10–20 s. All minerals were analyzed with a
focused beam; the experimental and the natural glasses and matrix compositions were analyzed using defocused 20 μm and 50 μm beam diameters, respectively. We analyzed both glass inclusions and the matrix because we anticipated that the matrix compositions would provide a more complete record of pre-eruptive melt compositions and compositional variation. The matrix analyses intentionally included glass domains and microlites, because they were performed to characterize the melt present in the pre-eruptive reservoir prior to microlite crystallization. Alkali migration in glasses under the electron beam was corrected for relative to the hydrous glass standards of Scaillet & Evans (1999). Glass volatile contents, which primarily represent glass H₂O contents, were estimated relative to the hydrous glass standards and electron microprobe totals with an uncertainty of ±1 wt %.

Glass H₂O and CO₂ contents were also calculated using VolatileCalc (Newman & Lowenstern, 2002) for an average of predicted volatile solubilities in rhyolitic and basaltic melts (to account for the dacitic melt compositions in Merapi’s magma reservoir). The uncertainties on the calculated solubilities are typically better than ±1 wt % for H₂O, but up to ±1000 ppm for CO₂, whereas relative uncertainties on CO₂ solubilities at <500 MPa and in a vapour with an XH₂O of ≤0.5 are <200 ppm (see Baker & Alletti, 2012). Because the calculated glass volatile and glass H₂O contents closely compare (Table 3), and glass volatile contents could not be determined for all experimental charges, we predominantly refer to the calculated melt H₂O contents in subsequent text and figures. The vapour XH₂O contents (XH₂O_v) reported throughout the text and figures are those that prevailed once the experimental melts had reached volatile saturation and the charges had reached equilibrium (XH₂O_v = XH₂O_eq; Table 3).

Reported mineral end-member compositions are percentage anorthite (An = [Ca/(Ca + Na + K)] × 100) and orthoclase (Or = [K/(K + Ca + Na)] × 100) for plagioclase, enstatite (En = [Mg/(Mg + Fe_tot + Ca)] × 100), wollastonite (Wo = [Ca/(Ca + Mg + Fe_tot)] × 100) and ferrosilite (Fs = [Fe_tot/(Ca + Mg + Fe_tot)] × 100) for clinopyroxene and orthopyroxene, and forsterite (Fo = [Mg/(Mg + Fe_tot)] × 100) for olivine. Normal and reverse zoning refer to rimward decreasing or increasing Ca, Al, or Mg contents, respectively. Mineral–melt partition coefficients (K_D) are (Ca/Na)_P/(Ca/Na)_Gl for plagioclase and (Fe/Mg)_P/(Fe/Mg)_Gl for clinopyroxene, which are reported as average values for single charges. The mineral abbreviations used in text and figures follow those of Whitney & Evans (2010).
Fig. 4. Summary of the compositional variation for (a–d) plagioclase, (e–h) clinopyroxene, and (i–l) amphibole core and rim compositions in stage 4+6 and stage 7 samples. The core compositions are bimodal; the rim compositions show a wide range of compositions in which primitive compositions are slightly more common in the stage 7 than in the stage 4+6 samples. *, abundance normalized to 100; n, number of original analyses; #, data from Erdmann et al. (2014). Dark and light grey shaded fields in (c), (g), and (k) show the mode and range of outer mineral rim compositions for our starting material sample SEM043b.

Table 2: Possible experimental and natural equilibrium states

<table>
<thead>
<tr>
<th>Equilibrium Type</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total equilibrium</td>
<td>equilibrium for all phases and chemical components of the system</td>
</tr>
<tr>
<td>Local equilibrium</td>
<td>equilibrium for all crystal rims and host melt, but not for crystal cores and host melt</td>
</tr>
<tr>
<td>Partial equilibrium</td>
<td>equilibrium for some, but not all crystal rims and host melt</td>
</tr>
<tr>
<td>Exp.</td>
<td>Charge (vol. %)</td>
</tr>
<tr>
<td>------</td>
<td>-----------------</td>
</tr>
<tr>
<td>Run 6: 195 MPa, 1000°C, $H_2O = 1$ bar, $t = 67$ h</td>
<td></td>
</tr>
<tr>
<td>CE-6</td>
<td>6–11c</td>
</tr>
<tr>
<td>CE-6</td>
<td>6–12c</td>
</tr>
<tr>
<td>CE-6</td>
<td>6–13c</td>
</tr>
<tr>
<td>CE-6</td>
<td>6–14c</td>
</tr>
<tr>
<td>CE-6</td>
<td>6–15c</td>
</tr>
<tr>
<td>ME-6</td>
<td>6–7m</td>
</tr>
<tr>
<td>ME-6</td>
<td>6–8m</td>
</tr>
<tr>
<td>ME-6</td>
<td>6–9m</td>
</tr>
<tr>
<td>Run 5: 295 MPa, 975°C, $H_2O = 1$ bar, $t = 67$ h</td>
<td></td>
</tr>
<tr>
<td>CE-5</td>
<td>5–16c</td>
</tr>
<tr>
<td>CE-5</td>
<td>5–17c</td>
</tr>
<tr>
<td>CE-5</td>
<td>5–18c</td>
</tr>
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<td>ME-5</td>
<td>5–4m</td>
</tr>
<tr>
<td>ME-5</td>
<td>5–5m</td>
</tr>
<tr>
<td>ME-5</td>
<td>5–6m</td>
</tr>
<tr>
<td>Run 9: 202 MPa, 925°C, $H_2O = 1$ bar, $t = 80$ h</td>
<td></td>
</tr>
<tr>
<td>CE-9</td>
<td>9–31c</td>
</tr>
<tr>
<td>ME-9</td>
<td>9–16m</td>
</tr>
<tr>
<td>ME-9</td>
<td>9–17m</td>
</tr>
<tr>
<td>ME-9</td>
<td>9–19m</td>
</tr>
<tr>
<td>ME-9</td>
<td>9–19m</td>
</tr>
<tr>
<td>Run 7: 204 MPa, 950°C, $H_2O = 1$ bar, $t = 70$ h</td>
<td></td>
</tr>
<tr>
<td>CE-7</td>
<td>7–26c</td>
</tr>
<tr>
<td>CE-7</td>
<td>7–27c</td>
</tr>
<tr>
<td>CE-7</td>
<td>7–29c</td>
</tr>
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<td>7–31c</td>
</tr>
<tr>
<td>ME-7</td>
<td>7–13m</td>
</tr>
<tr>
<td>ME-7</td>
<td>7–15m</td>
</tr>
</tbody>
</table>

* $X_{H_2O}$ in = $H_2O/(H_2O + CO_2)$ (mol %) added to the charges.
† $X_{H_2O_{eq}}$ in = final $X_{H_2O}$ of the experiment, calculated from $X_{H_2O_{eq}}$, melt $H_2O$ solubilities, and experimental glass proportions; $X_{H_2O_{eq}}$ reported in text corresponds to $X_{H_2O_{eq}}$.
¶ $\log fO_2$ (experiment) − $\log fO_2$ (NNO), where $fO_2$ in experiments was calculated from solid NiO + Ni-Pd sensors for variable $X_{H_2O}$ of the charges.
§ Melt V is total melt volatile content, estimated by difference from electron microprobe totals and corrected relative to hydrous glass standards.
¶¶ Calculated using VolatileCalc of Newman & Lowensten (2002), where summarized $H_2O$ and $CO_2$ contents are rounded values; estimated uncertainties are high; that is, up to ~1000 ppm (see Baker & Alletti, 2012).
** Intrinsic $fO_2$ of the pressure vessel (see Scaillet & Evans, 1999).
RESULTS

The natural samples

Whole-rock composition and petrology

Maggmas that have erupted at Merapi in prehistoric to historical times are predominantly basaltic andesite in composition, but occasionally include basalts (Abdurachman, 1998; Gertisser & Keller, 2003; Chadwick et al., 2013; Costa et al., 2013; Preece et al., 2013; Fig. 2). Their SiO2 contents correlate poorly with Al2O3 (Fig. 2a), negatively correlate with MgO, Fe2O3, CaO and TiO2 (Fig. 2b–d), and positively correlate with Na2O and K2O (Fig. 2e and f). The stage 4 + 6 samples erupted in 2010 and the stage 7 samples of the final 2010 dome have moderately evolved basaltic andesite compositions with ~53–55 wt % SiO2 and ~18.9–19.6 wt % Al2O3, comparable with those of the majority of magmas discharged at Merapi from 1930 to 2006 (Abdurachman, 1998; Gertisser & Keller, 2003; Costa et al., 2013; Preece et al., 2013; Fig. 2). Basaltic magmas or enclaves, however, did not erupt in 2010 (stage 4 + 6) or in 2013 from the terminal 2010 dome (stage 7). Calc-silicate xenoliths are scarce (<1 vol. %) and none of the samples is enriched in CaO. The compositions of the stage 4 + 6 and 7 samples are broadly comparable, but the stage 7 samples extend on average to slightly more primitive compositions with ~54.7 wt % SiO2 relative to the stage 4 + 6 samples with ~55 wt % SiO2 (Fig. 2a).

The investigated rocks, including dense and scoriaceous dome clasts and pumice samples, are crystal-rich with ~40–60 vol. % phenocrysts and microphenocrysts (Fig. 3). Plagioclase (~25–40 vol. %) and clinopyroxene (~5–15 vol. %) form the following: (1) common phenocrysts with complex zoning patterns, evidence of resorption, and inclusion-rich zones (Fig. 3a–c); (2) microphenocrysts that are largely inclusion-free (Fig. 3a–d); (3) rare phenocrysts that are inclusion-poor and largely unresorbed with core–rim step zoning (Fig. 3b–d). Magnetite (<5 vol. %), amphibole (<5 vol. %), and orthopyroxene (<2 vol. %) are minor constituents that form subhedral to euhedral crystals (Fig. 3a–d). Olivine rarely occurs as inclusions <20 μm in size, most commonly in amphibole phenocrysts. The stage 4 + 6 samples characteristically have few inclusion-poor plagioclase and clinopyroxene phenocrysts with simple core–rim zoning (typically <5 vol. %); they contain abundant subhedral, but few euhedral magnetite crystals (Fig. 3a). Largely unresorbed amphibole microphenocrysts are common, whereas unresorbed amphibole phenocrysts are absent (Fig. 3a). Most of the stage 7 samples, in contrast, have a larger proportion of inclusion-poor plagioclase and clinopyroxene crystals with simple core–rim zoning (typically ~5–15 vol. %), and euhedral magnetite crystals (Fig. 3b). They also have largely unresorbed amphibole phenocrysts in addition to microphenocrysts (Fig. 3d).

Mineral composition and zoning

Plagioclase, clinopyroxene, and amphibole in the basaltic andesites show a large range in composition, in which primitive core and rim compositions slightly increase in abundance from stage 4 + 6 to stage 7 samples (Fig. 4; Supplementary Data Electronic Appendix A2). Plagioclase has ~An40–90 core compositions with compositional maxima at ~An50 and ~An90, where the primitive >An80 compositions are characteristic of inclusion-poor crystal cores (Fig. 4a and b). The crystal rims have predominantly ~An50 to ~An70 composition, but range from ~An40 to ~An90 (Fig. 4c and d). Clinopyroxene is augite and less commonly diopside in composition, where cores (Fig. 4e and f) and rims (Fig. 4g and h) have ~Wo40–50 content with compositional maxima around ~Wo42–43. The core–rim zoning patterns of plagioclase and clinopyroxene include normal and reverse zoning, but zoning is most commonly irregularly oscillatory (Fig. 5). Broad plateau zones with evolved composition (i.e. typically with ~An40–50 and ~Wo42–43) and ~10–100 μm wide zones with intermediate composition (i.e. with ≤An70 and ≤Wo50) alternate. The outer ~100 μm of the crystals most commonly show normal zoning, but reverse zoning is

Fig. 5. Typical zoning profiles for (a) anorthite (An) in plagioclase, (b) wollastonite (Wo) and (c) Al2O3 in clinopyroxene from a stage 4 dome clast [same profile in (b) and (c)]. Characteristic features are (1) the evolved plateau zones with ~An50 plagioclase and ~Wo42–43 and ~2 wt % Al2O3 in clinopyroxene, and (2) the irregular and relatively thin, intermediate zones with <An70 in plagioclase and ≤Wo50 and ≤8 wt % Al2O3 in clinopyroxene.
also developed, particularly for plagioclase and clinopyroxene of the stage 7 samples.

Amphibole microphenocrysts in stage 4 + 6 samples have core compositions with ~11 and ~13 wt % Al₂O₃ (Fig. 4i). The crystals are normally or reversely zoned with rim compositions of ~10–14 wt % Al₂O₃ (Fig. 4k). Amphibole phenocrysts that are characteristic for the stage 7 samples are Al-rich compared with the microphenocrysts of the stage 4 + 6, with compositional maxima at ~12 and 14 wt % Al₂O₃ (Fig. 4j). The cores with ~12 wt % Al₂O₃ often have patchy zoning and small olivine inclusions. The cores of the volumetrically predominant phenocryst type with ~14 wt % Al₂O₃ are largely unzoned except for low-amplitude oscillatory zoning and <200 µm wide normally zoned rims. The rims of the stage 7 phenocrysts and core or rim compositions of the stage 7 microphenocrysts were not analyzed, because they are pervasively oxidized. The near-rim compositions of the phenocrysts are, however, predominantly Al-rich with ~14 wt % Al₂O₃ (Fig. 4i), overlapping with the core and rim compositions of the most primitive stage 4 + 6 microphenocrysts (Fig. 4i and k).

Magnetite with subhedral shape, which is predominant in the stage 4 + 6 and the stage 7 samples, has core compositions with ~77 wt % FeO, ~2–7 wt % Al₂O₃ and ~8–8 wt % TiO₂. The crystals in the stage 4 + 6 samples are reversely zoned with rim compositions of ~76 wt % FeO, ~2–6–4 wt % Al₂O₃, and ~7–0–9 wt % TiO₂, whereas those in the stage 7 samples show trellis-like exsolution that has overprinted any original zoning. The rare euhedral magnetite crystals, which are characteristic for the stage 7 samples, have ~74 wt % FeO, ~6–0 wt % Al₂O₃ and ~5–6 wt % TiO₂ core composition, but their original zoning is also not preserved, owing to exsolution. Orthopyroxene, which predominantly occurs as microphenocrysts, has ~En67 core composition and is largely unzoned. Olivine, which occurs only in the form of inclusions, has an evolved ~Fo60–70, low NiO (<0.25 wt %), and high MnO (~1–0–1.4 wt %) and CaO (~0–1–0.2 wt %) composition.

Matrix and glass inclusion compositions

The matrix (glass plus microlites, analyzed with a 50 µm beam diameter) of the stage 4 + 6 and the stage 7 dome clasts is dacitic with ~66–70 wt % SiO₂, ~15–17 wt % Al₂O₃, and ~2–4 wt % CaO (Fig. 6). Glass inclusions in clinopyroxene phenocrysts have the same range of SiO₂ and Al₂O₃ contents as the matrix (Fig. 6a), but they have lower CaO contents of <2 wt %, indicating that they were modified by post-entrapment crystallization (Fig. 6b). Their volatile contents calculated by difference from electron microprobe totals are ~0–5–2.5 wt % for dome clasts and ~3–4 wt % for pumice samples.

The experimental study

Run products of the crystallization experiments

The seven crystallization experiments were carried out at 925–1000 °C and ~100–400 MPa (Table 3). Six experiments were performed at moderately oxidizing conditions of ~NNO + 1.5 to ~NNO + 0.7; one experiment was performed at >NNO + 2 (CE-1) (Table 3). The crystallization of charges with an XH₂Oᵥ of 1 to ~0.5 produced up to ~90 vol. % of euhedral to subhedral crystals. Crystallinities of ≤40–60 vol. %, which are characteristic for the natural samples, formed in charges crystallized at ≤950 °C with an XH₂Oᵥ of ≥0.7 and in charges crystallized at ≥975 °C with an XH₂Oᵥ of ≤0.8 (Table 3). Plagioclase (7–66 vol. %), clinopyroxene (0–18 vol. %), and magnetite (0–5 vol. %) occur in all run products except for three that crystallized at ≥1000 °C and ≥300 MPa with an XH₂Oᵥ of 1. Orthopyroxene formed in charges crystallized at 950 °C and 100 MPa with an XH₂Oᵥ of ≤0.8. Amphibole and olivine, however, were not detected in any of the crystallization run products.

Matrix: stage 4+6 △ stage 7 Glass inclusions: ▲ stage 4 △ stage 4+6 (Preece et al. 2014)

Fig. 6. Composition of matrix (glass plus microlites, analyzed with a 50 µm beam) and glass inclusions in clinopyroxene phenocrysts of stage 4 + 6 and stage 7 dome clasts. Data shown are from this study and from Preece et al. (2014).
The experimental glasses have a basaltic andesitic to dacitic composition (Fig. 7a). Those that have formed in charges crystallized at 950°C and ~200 MPa with an \( \text{H}_2\text{O}_\text{v} \) of 0.7 and a melt \( \text{H}_2\text{O} \) content of ~3.6 wt % approach some of the least evolved matrix compositions of the natural samples with ~66 wt % \( \text{SiO}_2 \) and ~16.5 wt % \( \text{Al}_2\text{O}_3 \) (Fig. 7a). The experimental plagioclase compositions range from ~An50 to ~An90, compared with the overall more evolved rim compositions of the natural crystals with ~An40 to ~An90 (Fig. 7b). The plagioclase \( K_0 \) (Ca–Na) values for crystal rims formed at 950°C and ~200 MPa (CE-7) increase from 3.6 to 5.9 with increasing melt \( \text{H}_2\text{O} \) and \( \text{X}_\text{H}_2\text{O}_\text{v} \) of the charges (Supplementary Data Electronic Appendix A3). Clinopyroxene Wo and En contents of the experimental and natural crystals largely match (Fig. 7c), but the \( \text{Al}_2\text{O}_3 \) contents of the experimental crystals are high (~3–8 wt %) relative to the natural crystals (~1.2–7 wt %) (Fig. 7d and e). The clinopyroxene \( K_0 \) (Fe–Mg) values for crystal rims formed at 950°C and ~200 MPa (CE-7) vary between 0.19 and 0.30, increasing with melt \( \text{H}_2\text{O} \) and \( \text{X}_\text{H}_2\text{O}_\text{v} \) (Supplementary Data Electronic Appendix A3). The experimental and the natural magnetite crystals are comparable in their \( \text{FeO} \) and \( \text{TiO}_2 \) contents, whereas the \( \text{Al}_2\text{O}_3 \) contents of the experimental crystals are relatively high (Fig. 7f). We note that crystallization at variable oxygen fugacities (i.e. at strongly oxidizing, ~NNO + 2 conditions in CE-1 compared with all other experiments run at ~NNO + 1.5 to NNO + 0.7) affects the \( \text{TiO}_2 \), \( \text{FeO} \), and \( \text{MgO} \) contents of clinopyroxene, magnetite, and glass, but that it does not account for their high-Al contents (Fig. 7).

**Run products of the melting experiments**

The six melting experiments were performed in conjunction with the crystallization experiments at 925–1000°C, ~100–400 MPa, and at ~NNO + 1.5 to ~NNO + 0.7 (Table 3). Partial melting and partial crystallization of the basaltic andesite rock powders in charges with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~0.4–1 resulted in run products with 0 to ~70 vol. % crystals (Table 3). Crystallinities of ~40–60 vol. %, which are characteristic for the natural samples, were obtained in charges recrystallized at ~950°C with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~0.6 and in charges run at ~975°C with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~0.7 (Table 3). Plagioclase (~7–66 vol. %), clinopyroxene (~0–18 vol. %), and magnetite (~0–5 vol. %) are present in all experimental products except for those of three charges run at 1000°C or ~300 MPa with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~1. Amphibole occurs in charges with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~1 and ~0.8 that were run at 925°C and ~200 MPa, forming clusters of randomly oriented crystals. Orthopyroxene is present in charges recrystallized at 950°C and ~100 MPa with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~0.8, forming coarse dendritic crystals. The run products of the ~950°C, ~200 MPa melting experiments thus reproduce the mineral assemblage of natural samples from Merapi. Only olivine, which forms rare inclusions in natural amphibole phenocrysts, was not detected.

The glasses of the melting experiments are basaltic andesitic to dacitic; those that formed in charges run at ~950°C and ~200 MPa with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~0.5–0.6 match the moderately evolved natural matrix and glass inclusion compositions with ~68 and 16 wt % \( \text{SiO}_2 \) and \( \text{Al}_2\text{O}_3 \) (Fig. 8a). They have calculated \( \text{H}_2\text{O} \) and \( \text{CO}_2 \) contents of ~3.3 wt % and ~470 ppm, respectively (Table 3). Glass produced in charges of our ~975°C and ~300 MPa experiments, however, deviate from the natural matrix and glass compositions (Fig. 8a). Their \( \text{SiO}_2 \) and \( \text{Al}_2\text{O}_3 \) contents are ~<2 and ~16.5 wt %, and their calculated melt \( \text{H}_2\text{O} \) contents are ~3–8 wt %. Plagioclase crystal rims produced in the experiments have ~An40–85 composition, and thus compare with the range of rim compositions detected for the natural samples (Fig. 8b). Their average \( K_0 \) (Ca–Na) values vary between 3.0 and 6.7 (Supplementary Data Electronic Appendix A3). The ~An50 composition, which is most common in the natural crystal rims, is reproduced in charges run at ~200 MPa with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~0.5–0.6 and a melt \( \text{H}_2\text{O} \) content of ~<3.3 wt % (Fig. 8b). Crystals with ~An50 to ~An70 rims formed in charges with melt volatile and \( \text{H}_2\text{O} \) contents of ~3–5 wt % that were run at ~200 MPa with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~0.8 or at ~400 MPa with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~<0.4. However, few cores and inner rims (~5 vol. %) of newly crystallized plagioclase (~5–10 vol. %) in the low-pressure, ~<200 MPa experiments with a low \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~<0.7 also have ~An50 to ~An80 compositions, whereas their \( K_0 \) (Ca–Na) values are significantly (~>2) higher than those of the newly crystallized outer rims in the same charges.

Clinopyroxene rim compositions have ~En32–42, ~Wo40–50, ~1.8–8 wt % \( \text{Al}_2\text{O}_3 \), ~19–23 wt % \( \text{CaO} \), and ~10–15 wt % \( \text{MgO} \). Their \( K_0 \) (Fe–Mg) values vary between 0.16 and 0.38, decreasing with increasing experimental temperature and increasing with experimental \( \text{H}_2\text{O} \) and \( \text{X}_\text{H}_2\text{O}_\text{v} \). Clinopyroxene with ~Wo42–43, ~En43, and ~2 wt % \( \text{Al}_2\text{O}_3 \) that matches the most common natural rim compositions was approached in charges with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~<0.6 and a melt \( \text{H}_2\text{O} \) content of ~<3.3 wt % at ~<200 MPa (Fig. 8c–e). Primitive natural clinopyroxene rim compositions, and in particular those with ~46 Wo, ~3 wt % \( \text{Al}_2\text{O}_3 \) and ~22 wt % \( \text{CaO} \), are produced at ~<200 MPa in charges with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~<0.8 and at ~300 MPa in charges with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~<0.7 (Fig. 8b–d). Some cores and inner rims of newly crystallized clinopyroxene (~5–10 vol. %) in our ~<950°C, \( \text{X}_\text{H}_2\text{O}_\text{v} \) ~<0.6 experiments, moreover, have high-\( \text{Al}_2\text{O}_3 \) (~>2 to 6 wt %), low-\( \text{CaO} \) (~<20 wt %) compositions that are unknown from the natural samples, but their modal proportion is low (~<5–10 vol. %). Amphibole, which is a minor but characteristic constituent of Merapi basaltic andesites, formed in our melting experiments at 925°C and ~200 MPa, but was not detected in any of the higher temperature runs. Crystals from charges with an \( \text{X}_\text{H}_2\text{O}_\text{v} \) of ~<0.8 and a melt \( \text{H}_2\text{O} \) content of ~4.0 wt %
Fig. 7. Composition of the crystallization experiment (CE) run products compared with the natural phase assemblage: (a) glass compositions; (b–e) plagioclase and clinopyroxene rim compositions; (f) evolved magnetite core (C) and rim (R) compositions. The grey fields outline the range of the natural phase compositions; the red fields mark the most common compositions. All data are reported in Supplementary Data Electronic Appendices A2 and A3. Experiment CE-1 was performed at NNO > 2; all other experiments were performed at ~NNO ± 1 to ~NNO ± 0.7.
Fig. 8. Composition of the melting experiment (ME) glasses and newly crystallized outer mineral rims compared with the natural phase assemblage: (a) glass compositions; (b–e) plagioclase and clinopyroxene rim compositions; (f) evolved magnetite core (C) and rim (R) compositions. The grey fields mark the range of the natural phase compositions; the red fields show the most common compositions. All data are reported in Supplementary Data Electronic Appendices A2 and A3. All experiments were performed at \( \pm NNO + 1.5 \) to \( \pm NNO + 0.7 \).
have compositions comparable with those of the Al
poor (~11 wt % Al2O3) natural microphenocrysts (Fig. 9).
Amphibole from charges with an XH2O = 1 and a
melt H2O content of ~4.9 wt %, in contrast, approach
the composition of the natural Al-rich (~13 wt % Al2O3)
micropocknocrysts cores (Fig. 9). Magnetite compositions
that approach those of the evolved natural crystals form
at 925–950 °C (Fig. 8f). The most evolved compositions,
which are preserved in the natural crystal cores, were
approached in charges with an XH2O of ≤0.6, whereas
the natural magnetite rim compositions are approached
in charges with an XH2O of ≥0.7 at 925 and 950 °C (Fig.
8f). The natural ~En67 orthopyroxene compositions are
reproduced at 950 °C and ~100 MPa in charges with an
XH2O of ≤0.8.

**DISCUSSION**

We first evaluate the conditions of equilibrium in our
crystallization and melting experiments and their potential
for constraining the crystallization conditions in
Merapi’s pre-eruptive magma reservoir. We then
compare the natural and the experimentally produced
mineral assemblages and compositions (1) to constrain the
crystallization conditions of the resident magma, (2) to
characterize the components and conditions of the pre
eruptive magma recharge, and (3) to evaluate the
processes that control the variation in crystallization
conditions. We focus our discussion on interpreting the
evolution of magmas involved in Merapi’s 2010 eruption,
but note that whole-rock compositions, mineral
assemblages, and mineral and glass compositions are
largely invariant in pyroclasts erupted between at least
1930 and 2010 (Berthommier, 1990; Abdurachman, 1998;
Chadwick et al., 2013; Costa et al., 2013; Preece et al.,
2013). We therefore concur with previous studies that
Merapi’s shallow magma plumbing system has under
gone no radical modifications in the last ~100 years,
whereas variations in extensive parameters (e.g. vol-
umes of resident and recharge magmas or the amounts
of carbonate assimilation) may have been important (e.g.
Borisova et al., 2013; Costa et al., 2013; Troll et al., 2013).

**Experimental constraints**

**Results from the crystallization experiments**

The run products of our crystallization experiments show systematic modal and compositional variation as
a function of experimental conditions (Fig. 10a and b;
Table 3; Supplementary Data Electronic Appendix A3).
Plagioclase anorthite content increases with increasing
melt H2O content and with experimental temperature
(Fig. 10a). Average plagioclase K0 (Ca–Na) values also
systematically increase with increasing melt H2O content
from 3.7 to 6.4 for melt H2O contents of ~3.6–5.9 wt %
(CE-7; Supplementary Data Electronic Appendix A3), and
they are comparable with values determined by pre-
vious studies for compositionally comparable systems
(e.g. Sisson & Grove, 1993; Pichavant et al., 2002;
Parman et al., 2011). Clinopyroxene CaO content also
increases with melt H2O content, but decreases with
increasing pressure, whereas it shows little variation as
a function of experimental temperature (Fig. 10b).
Average clinopyroxene K0 (Fe–Mg) values of between
0.20 and 0.30 decrease with temperature and increase
with increasing melt H2O content (Supplementary Data
Electronic Appendix A3), and they also are comparable
with values determined in other studies (e.g. Sisson &
Grove, 1993; Pichavant et al., 2002; Parman et al., 2011).
We suggest that both the systematic compositional vari-
atation of the crystallization experiment run products and
partition coefficients that are comparable with literature
values indicate a close approach to equilibrium in our
experiments as intended (see Table 2). That the run
products failed to reproduce the natural mineral and
glass compositions could indicate that the temperature,
pressure, XH2O, or fO2 conditions did not match those of
the natural system or that our starting material repres-
ents a non-liquid composition. We rule out the inter-
pretation that the explored temperature, pressure, and
XH2O conditions are inadequate, because the crystallin-
ities characteristic for the natural samples and the melt
H2O contents were well reproduced. We also rule out the
interpretation that the moderately oxidizing crystallization
conditions (at an fO2 of ~NNO + 1.5 to ~NNO + 0.7)
are the cause of the compositional mismatch between
the crystallization experiment run products and the nat-
ural samples, because the Fe–Mg–Ti compositions of the
experimental and the natural clinopyroxene, magnetite,
and glasses are well matched. The unmatched high Al
contents of minerals and glasses, in contrast, are not affected by variations in $f_{O_2}$ (Fig. 7). We therefore suggest that our starting material represents a non-liquid composition; that is, we infer that the sample from which we fused our starting glass comprises accumulated cognate, antecrystic and xenocrystic components and mixed liquids, which is consistent with Merapi’s inferred open-system evolution (e.g. Chadwick et al., 2007, 2013; Nadeau et al., 2011, 2013; Borisova et al., 2013; Costa et al., 2013; Van der Zwan et al., 2013; Erdmann et al., 2014). As a result, we cannot use the phase proportions or compositions from our crystallization experiments to constrain the conditions in Merapi’s pre-eruptive reservoir; instead, we use their compositions below to interpret the mineral rim compositional variation detected in our melting experiments.

**Results from the melting experiments**

Compared with the crystallization experiments, melting experiments of the type performed have the advantage that they partially preserve the original crystal assemblage that was present in the magmas stored within the pre-eruptive reservoir, including xenocrysts, antecrysts, and accumulated or mixed phenocryst populations. The experimental conditions closely match the crystallization conditions in the pre-eruptive reservoir if (1) all microlites that crystallized during magma ascent have melted and (2) all phenocrysts in the charges and their rim compositions match those of the natural samples. Melting experiments are thus suitable for characterizing the late-stage crystallization conditions in Merapi’s crystal-rich pre-eruptive reservoir; however, it has to be kept in mind (and is considered further below) that the matrix of the starting rock powders may nevertheless contain components of the resident and the recharge melts. As in the crystallization experiments, the outer mineral rim and glass compositions produced in our melting experiments show systematic modal and compositional variations as a function of experimental conditions and partition coefficients comparable with those determined by previous studies (e.g. Sisson & Grove,
Mineral proportions, because we cannot accurately recharge. We also compare the crystallinities between the resident magma in Merapi’s pre-eruptive reservoir,ing experiments to infer the crystallization conditions of the experiments approached local equilibrium conditions of inner zones of new clinopyroxene, differ by >2σ from the compositions of outer crystal rims (which is unknown from the natural samples). They are interpreted to record early stages of experimental crystallization probably during heating and at high fluid/melt ratios, and they are therefore not considered in the subsequent discussion. Their effect on the composition of the reactive system is nevertheless considered to be limited, given that they have a low proportion of <5–10 vol. % of all newly crystallized plagioclase and clinopyroxene. That the mineral rim and glass compositions of the melting experiments nevertheless follow compositional trends that are comparable with those of the crystallization experiments (Fig. 10c and d versus Fig. 10a and b), that plagioclase–melt and clinopyroxene–melt partition coefficients are comparable with equilibrium values, and that outer rim compositions within single charges have relatively homogeneous compositions (Supplementary Data Electronic Appendix A3) is taken as evidence that the experiments approached local equilibrium conditions as intended.

In the following sections, we therefore use the mineral rim and glass compositions produced in the melting experiments to infer the crystallization conditions of the resident magma in Merapi’s pre-eruptive reservoir, and to approximate the conditions of the pre-eruptive recharge. We also compare the crystallinities between the natural and the experimental samples, but not their mineral proportions, because we cannot accurately determine the volume of, for example, co-crystallizing plagioclase and clinopyroxene in the natural basaltic andesites (this would require extensive compositional mapping). We suggest that the conditions of the resident magma can be closely constrained by this approach, because the compositionally primitive mineral zones (and microlites) that crystallized from the recharge magma remain unmelted. We further suggest that the compositional variation detected in our melting experiments can be used to closely approximate the recharge crystallization conditions in Merapi’s pre-eruptive reservoir, because the compositional difference between the resident melt (inferred to correspond to the evolved matrix and glass inclusion compositions with ~68 wt % SiO₂ and ~15 wt % Al₂O₃; Fig. 6a) and the recharge melt (inferred to approach the primitive matrix and glass inclusion compositions with ≤66 wt % SiO₂ and ≥17 wt % Al₂O₃; Fig. 6a) is limited and the effect on our estimate is thus minor. Previous crystallization experiments with basaltic and basaltic andesitic starting materials with a compositional difference of ≤3 wt % SiO₂ and ≤1 wt % Al₂O₃ but otherwise identical crystallization conditions, for example, yielded equivalent plagioclase and clinopyroxene compositions in charges in which both minerals co-crystallized (e.g. 79–35 g # 12 and 82–66 #5 of Sisson & Grove (1992) or SAR3 and SAR16 of Parman et al. (2011)). We therefore infer that the results of our melting experiments can be used for estimating both the intensive parameters of the resident magma and those of the recharge magma. We acknowledge that estimating the crystallization conditions on the basis of our melting experiments is more uncertain than for estimates based on crystallization experiments (owing to our use of a starting rock powder that introduced some compositional variation between charges), but that uncertainties are lower than the increments at which we have varied the intensive parameters (i.e. up to ±25 °C and ≤75 MPa). Uncertainties in the calculated melt H₂O contents (Table 3) are ±1 wt %, whereas those of the calculated melt CO₂ contents (Table 3) are large, up to ±1000 ppm (see Baker & Alletti, 2012). Fluctuations of H₂O and CO₂ in Merapi’s pre-eruptive reservoir, however, are inferred on the basis of mineral compositional variations, and not on the basis of the calculated CO₂ values.

**Shallow, H₂O-poor pre-eruptive magma storage**

Consistent with previous studies, we suggest that the pre-eruptive magma storage conditions in Merapi are recorded by the compositionally evolved mineral zones and by the evolved matrix (glass plus microlites) and glass inclusion compositions in the erupted, crystal-rich basaltic andesites (Table 4; see also Costa et al., 2013; Nadeau et al., 2013; Erdmann et al., 2014). Crystallinities of ~40 to ~60 vol. % and the predominant, ~An50 natural plagioclase and Al-poor clinopyroxene compositions are simultaneously reproduced in our melting experiments performed at 925–950 °C, ~100–200 MPa,
Table 4: Components and inferred crystallization conditions for Merapi’s magma plumbing system

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<th>Pre-eruptive Recharge</th>
<th>Deep, unconstrained Recharge</th>
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Fig. 11. Glass inclusion compositions, estimated resident and recharge melt H2O–CO2 contents, and inferred volatile evolution. (a) Glass inclusion compositions from Nadeau et al. (2013) and Preece et al. (2014); redrawn from Preece et al. (2014) with our estimates for resident and recharge melt compositions and vapour isopleths at an XH2OV of ~0–6 and ~0–8. (b–d) Insets shown in (a). (b, c) Scenario 1: Merapi’s pre-eruptive reservoir is filled by magmas with an XH2OV of ~0–8 (b, time 1) that become relatively H2O-poor as a result of CO2-rich volatile fluxing during magma storage (c, time 1) and by magmas with an XH2OV of ~0–6 during pre-eruptive periods (d, time 2). We favour the interpretation of Scenario 2, proposing that magmas ascending during inter-eruptive periods (at time 1) approach a vapour-buffered degassing path, whereas magmas ascending during pre-eruptive periods (at time 2) approach a closed-system degassing path.

with an XH2OV of ~0.5–0.6, and at a melt H2O content of ~3–4 wt % (Table 3; Fig. 8b, d and e). The evolved matrix and glass compositions of the natural samples with ~68–69 wt % SiO2 and ~15–15 wt % Al2O3 are also closely approached at these experimental conditions (Fig. 8a). Their calculated melt H2O and CO2 contents of ≤3–3 wt % and ~240–590 ppm, moreover, match the volatile contents of some of the natural glass inclusions analyzed by Preece et al. (2014) (Fig. 11). Amphibole with compositions approaching those of the evolved microphenocrysts of our stage 4 + 6 samples formed in charges run at 925°C with an XH2OV of ~0.8 and an estimated melt H2O content of ~4.0 wt %, but not at 950°C. Magnetite comparable with the evolved core composition of the natural samples also formed in charges run at 925°C (Fig. 8f). In combination, the mineral and glass compositions and crystallinities indicate that Merapi’s pre-eruptive reservoir crystallizes at predominantly ≤200 MPa, at a temperature of ~950°C and ≥925°C, and with a melt H2O content of ~3–4 wt % (Table 4; Fig. 12a
and b). Experimental orthopyroxene of composition \( \sim \text{En68} \), which closely corresponds to the natural \( \sim \text{En67} \) crystal compositions, crystallized at \( 950 \, ^\circ \text{C} \) and \( \sim 100 \, \text{MPa} \) in equilibrium with an \( \text{X}_{\text{H}_2\text{O}} \) of \( \sim 0.6 \) and a melt \( \text{H}_2\text{O} \) content of \( \sim 2.4 \, \text{wt\%} \), further indicating that Merapi’s pre-eruptive reservoir extends to levels shallower than a depth equivalent to \( 200 \, \text{MPa} \) (Tables 3 and 4; Fig. 12). Consistent with the experimental assemblage, we further propose that the natural plagioclase crystals with compositions of \( \sim \text{An40–50} \) and clinopyroxene crystals with \( \leq \text{Wo42} \) and \( \leq 19.5 \, \text{wt\% CaO} \) have formed in this shallow reservoir region, in agreement with the interpretations of Costa et al. (2013). The melt \( \text{H}_2\text{O} \) and \( \text{CO}_2 \) contents of \( \sim 2.4 \, \text{wt\%} \) and \( \sim 240 \, \text{ppm} \) calculated for these conditions also match those detected in several natural glass inclusions (see Preece et al., 2014). That the Fe, Mg, and Ti contents of glass, clinopyroxene, orthopyroxene, and magnetite produced in charges run at an \( f_{\text{O}_2} \) of \( \sim \text{NNO} + 1.0 \) and an \( \text{X}_{\text{H}_2\text{O}} \) of \( \leq 0.6 \) replicate the natural mineral and glass compositions indicates moderately oxidizing crystallization conditions (Table 3).

We emphasize that the evolved mineral and glass inclusion compositions characteristic of Merapi basaltic andesites were in equilibrium and that they all record crystallization in the pre-eruptive reservoir. Previous studies, in contrast, have suggested that crystals and glass inclusions record magma evolution at deep and shallow levels, respectively, and thus that their record of crystallization conditions is largely decoupled (Nadeau et al., 2013; Preece et al., 2014). Our estimated magma storage temperature of \( \geq 925–950 \, ^\circ \text{C} \) is consistent with the crystallization temperatures calculated using evolved amphibole compositions and the Ridolfi et al. (2010) or the Ridolfi & Renzulli (2012) calibrations (i.e. \( \sim 920–970 \, ^\circ \text{C} \); Costa et al., 2013; Nadeau et al., 2013; Erdmann et al., 2014). Calculated clinopyroxene crystallization temperatures of \( \sim 1025 \) to \( 1060 \, ^\circ \text{C} \) (Preece et al., 2014) using the thermometer of Putirka et al. (2003) are comparatively high, but the lower estimated crystallization temperatures are within the error of our estimate. The inferred shallow pre-eruptive storage at \( \geq 100–200 \, \text{MPa} \) indicates that Merapi’s pre-eruptive reservoir is located at a depth of \( \geq 4.5 \, \text{km} \), and thus within the carbonate-dominated upper crust (see Smyth et al., 2005), if we assume an average overlying crustal density of 2242 kg m\(^{-3}\) (see Tiede et al., 2005) and lithostatic pressure conditions. If the reservoir was under significant overpressure (see Costa et al., 2013; Genareau et al., 2014), then we infer

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**Fig. 12.** Sketch summarizing the estimated crystallization conditions for Merapi’s pre-eruptive magma storage reservoir, its location and the main processes proposed for its evolution prior to recent historical eruptions, showing a timeline leading up to the 2010 eruption. (a, b) During inter-eruptive storage, Merapi’s basaltic andesitic magmas crystallize an evolved mineral assemblage. (c, d) During pre-eruptive magma recharge, the reservoir is replenished by magma with an \( \text{H}_2\text{O} \)-rich melt and vapour phase, crystallizing mineral zones of intermediate composition. (e, f) The intensive parameters of magma parcels of the original resident and the recharge magma equilibrate. (g, h) Inter-eruptive recharge by magma with an \( \text{H}_2\text{O} \)-poor melt and vapour phase begins. The inferred time scales of this evolution need to be assessed by detailed mineral composition diffusion modelling. We further note that the geometry of the magma storage zone is unconstrained by our data, except that we infer that the erupted magma and its phenocrysts must have collected over a pressure range of \( \sim 100 \, \text{MPa}, \) equivalent to a depth range of \( \sim 4.5 \, \text{km} \). BA = basaltic andesite.
that it is located at even shallower levels (e.g. at $\geq 2.5$ to 7 $\pm$ 3 km for an overpressure of $\leq 50$ MPa). A shallow, ephemeral magma storage zone at $\sim 1.5$–2.5 km below Merapi’s summit, which has been proposed (Ratdomopurbo & Poupinet, 1995, 2000; Saepuloh et al., 2010; Nadeau et al., 2013) but also contested on the basis of geophysical data and petrological data (Beauducel & Cornet, 1999; Müller & Haak, 2004; Costa et al., 2013), is not evident from our results. Our pressure estimates, however, are consistent with seismic and ground deformation data, which suggest magma storage at depths of $\geq 5.5$ to $\sim 8.5$ km below Merapi’s summit, from at least 1992 to 2010 (Ratdomopurbo & Poupinet, 1995, 2000; Beauducel & Cornet, 1999; Budi-Santoso et al., 2013). Our results are also consistent with glass inclusion barometry, which has been used to infer re-equilibration of magmas at $\leq 265$ MPa and depths of $\leq 10$ km (Preece et al., 2014), but they differ significantly from previous estimates based on amphibole and clinopyroxene barometry, which have suggested a main zone of pre-eruptive magma storage at $>100$–400 or at $\sim 400$–500 MPa (Gertisser, 2001; Chadwick et al., 2013; Costa et al., 2013; Preece et al., 2014). This result is not surprising, considering the uncertainties of the barometric methods employed (e.g. Fig. 1; Nimis, 1999; Putirka, 2008; Erdmann et al., 2014).

Crystalization pressure calculated for clinopyroxene using the Nimis (1999) barometer, which is not recommended for calc-alkaline compositions, yields moderate to high calculated pressures of $\sim 400$–500 MPa for Merapi’s main clinopyroxene crystal population if the crystallization temperature is moderately overestimated (i.e. at 1000 $^\circ$C in estimates of Chadwick et al. (2013)], and lower calculated pressures of $\sim 200$–400 MPa if the crystallization temperature is largely overestimated (i.e. at 1050 $^\circ$C in estimates of Preece et al. (2014)). For clinopyroxene that crystallized from more evolved magmas than the Merapi basaltic andesites, even higher pressure overestimates would result (Fig. 1a). That a large range in crystallization pressure is calculated with the Nimis (1999) calibration (e.g. Chadwick et al., 2013) is inevitable if the natural clinopyroxene crystals formed over a range of temperatures, but average clinopyroxene crystallization temperatures are assumed (e.g. over- and underestimates of crystallization temperatures by 20 $^\circ$C propagate into a 100 MPa decrease or increase in the calculated pressure; Nimis, 1999). The clinopyroxene crystallization pressure calculated for Merapi’s magma system using the geobarometer equations (32b) and (32c) of Putirka (2008) yields pressure estimates of predominantly $\sim 200$–300 and $\sim 100$–200 MPa (Preece et al., 2014). These agree well with our estimate, reflecting the fact that the barometers yield values that approach known crystallization pressures for clinopyroxene crystallized from mafic–intermediate magmas at moderately low temperatures (Fig. 1b and c). We point out, however, that the values calculated for Merapi are (1) based on largely overestimated clinopyroxene crystallization temperatures of 1025–1058 $^\circ$C, which increases the calculated pressure values, and (2) based on estimated primitive melt composition that corresponds to Merapi basaltic andesite whole-rock compositions (instead of dacitic compositions as recorded by glass inclusions), which decreases the calculated pressure values. The pressure estimates for the Merapi system based on the amphibole barometers of Ridolfi et al. (2010) and Ridolfi & Renzulli (2012) yield minimum values of $\sim 250$–400 MPa, reflecting that the method calculates intermediate pressures for amphibole crystallized in equilibrium with dacitic melts (Fig. 1d; Erdmann et al., 2014).

We emphasize that using single mineral thermometers or barometers to estimate the crystallization conditions of mixed magma systems has no advantage over experimental constraints, because any compositional variations in the magma and melt that complicate the experimental estimates equally affect the thermobarometric estimates. It is, moreover, important to keep in mind that errors on the calculated conditions may significantly exceed the standard errors of the methods, if the employed mineral thermometers or barometers were calibrated based on compositions or intensive parameters (e.g. $P$, $T$, $fO_2$, or $X_{H_2O}$) unlike those of the investigated system.

The geometry of Merapi’s shallow magma system cannot be constrained on the basis of our results or previous barometric estimates, but we emphasize that the erupted magma and its phenocryst cargo was assembled over a significant pressure and depth range of $\sim 100$ MPa and $\sim 4.5$ km. The pre-eruptive magma reservoir outlined in Fig. 12 highlights this depth range, but we cannot resolve if the magma storage zone is a single reservoir or a number of shallow sub-reservoirs connected by dykes as proposed by Chadwick et al. (2013) and Nadeau et al. (2013). An elliptical, subhorizontal magma source at a depth of $\sim 8.5$ km was predicted for the 1996–1997 eruption on the basis of global positioning system (GPS) deformation and tilt data (Beauducel & Cornet, 1999); this is more consistent with a plumbing system consisting of several sub-reservoirs, but remains to be confirmed. In the future, currently installed continuous GPS and tiltmeters (CVGHM/BPPTK and DOMERAPI projects) will provide tighter constraints.

**Pre-eruptive, $H_2O$-rich magma recharge**

The recharge of Merapi’s magma storage reservoir by a larger than average volume of volatile-rich mafic magma has been suggested in several previous studies to have played a key role in triggering and driving its paroxysmal 2010 eruption (e.g. Budi-Santoso et al., 2013; Costa et al., 2013; Jousset et al., 2013; Preece et al., 2013, 2014). Petrological studies have considered phenocrysts with primitive compositions and An-rich microlites as evidence for this recharge, but have
pointed out that the juvenile materials erupted in 2010 all have a homogeneous basaltic andesite composition (Costa et al., 2013; Preece et al., 2014; Fig. 2). More important is that the stage 7 samples, which have a higher proportion of pristine phenocrysts with primitive core and rim compositions that record the pre-eruptive recharge (Figs 3 and 4), also are closely comparable in composition with the stage 4 + 6 samples. We therefore suggest that the recharge magma of the 2010 event was basaltic andesitic and close in composition to the resident magma, at least in terms of major elements. This is important, because it indicates that magmas crystalize to a crystal-rich mush and mix in Merapi’s shallow pre-eruptive reservoir, but that they do not significantly fractionate at this level. If any significant differentiation takes place, it must be at deeper levels of the magma plumbing system.

The most direct evidence for magma recharge and mixing within Merapi’s pre-eruptive reservoir is the assemblage of normally zoned plagioclase, clinopyroxene, amphibole and magnetite phenocrysts with primitive core compositions [i.e. ~An90, ~Wo48, ~14 wt % Al2O3 amphibole, and ~6 wt % Al2O3 magnetite; Table 4; see also Costa et al. (2013) and Nadeau et al. (2013)]. The evolved ~Fo60–70 composition, low NiO (<0.25 wt %) and high CaO (>0.1 wt %) and MnO (>1 wt %) contents of rare olivine inclusions are characteristic for crystallization from basaltic andesitic magmas at <1000 °C (e.g. Sisson & Grove, 1992; Moore & Carmichael, 1998), and we thus interpret them as records of magma evolution in Merapi’s magma storage system and not as mantle-derived xenocrysts. We further consider the Al-poor (~12 wt % Al2O3) amphibole phenocrysts of the stage 7 samples as part of the primitive recharge assemblage, suggesting that they crystallized by replacing early stage olivine phenocrysts; this explains their common olivine inclusions, patchy zoning, and their high SiO2/Al2O3 contents as compared with the predominant population of Al-rich amphibole phenocrysts. Similar textures have been reported, for example, from andesites formed by mixing of evolved and primitive magmas (Foley et al., 2013; Kiss et al., 2014). The characteristic ~An80 plagioclase and ~Wo48 clinopyroxene core compositions of Merapi’s phenocrysts are reproduced in our H2O-rich charges crystallized at 400 MPa and 1000 °C, whereas amphibole, magnetite, or olivine that are part of the recharge assemblage did not form in the experiments. Whether this indicates that they crystallized from a magma that differed in composition from our starting material (e.g. from a basaltic magma or from one that was uncontaminated by calc-silicate components), or whether they crystallized at pressure and temperature conditions that were not explored by us (e.g. at >400 MPa or at ≤400 MPa and <1000 °C) needs to be established in future studies. The low proportion (~15 % of all phenocrysts) of the primitive, normally zoned recharge phenocrysts in Merapi’s basaltic andesites is notable, but is comparable with the proportion of recharge phenocrysts observed in other compositionally homogeneous, crystal-rich magmas such as those that have erupted in recent decades at Mount St Helens (Berlo et al., 2007; Streck et al., 2008; Cashman & Blundy, 2013).

We consider it possible that some of the An-rich plagioclase and Ca-rich clinopyroxene crystals in the Merapi magmas may be derived from the disaggregation or the assimilation of calc-silicate xenoliths, as has been inferred on the basis of plagioclase 87Sr/86Sr compositions (Chadwick et al., 2007), plagioclase and pyroxene δ18O compositions (Troll et al., 2013), calcic glass inclusions in some phenocrysts (Borisova et al., 2013; see also Deegan et al., 2010), and whole-rock trace element, 87Sr/86Sr, and δ18O compositions (Chadwick et al., 2007; Borisova et al., 2013; Troll et al., 2013). We note, however, that calc-silicate xenoliths are rare in the Merapi basaltic andesites erupted in 2010 (~<1 vol. %) and that clinopyroxene approaching wollastonite in composition has not been detected in any significant proportion (Borisova et al., 2013; Costa et al., 2013; Preece et al., 2014; our data), whereas it forms one of the main constituents of locally occurring calc-silicate xenoliths (e.g. Chadwick et al., 2007; Troll et al., 2013; our unpublished data). More important is that Merapi’s basaltic andesites have ‘typical’ CaO contents (e.g. <10 wt % at ~52 wt % SiO2; Fig. 2d) compared with those of other calc-alkaline basaltic andesites (e.g. Ambrym, Picard et al., 1994; Soufrière Hills, Murphy et al., 2000; Tatar San Pedro volcanic complex, Dungan et al., 2001; Mont Pelée, Pichavant et al., 2002). Small amounts (<2 wt %) of carbonate assimilation, as inferred by Chadwick et al. (2007), are consistent with the CaO contents and other compositional and mineralogical features of the Merapi magmas. Large amounts of inferred carbonate assimilation of up to ~20 or 15–40 wt % (see Borisova et al., 2013; Troll et al., 2013) should, however, clearly result in strongly CaO-enriched whole-rock compositions, as Costa et al. (2013) have previously shown.

Volumetrically more abundant than the primitive phenocrysts are intermediate zones and rims on phenocrysts of the resident magma and rims on the recharge magma phenocrysts with >An50 to ~An70 and >Wo43 to ~Wo50 composition, and amphibole microphenocrysts with ~13 wt % Al2O3 cores (Fig. 6). We infer that they crystallized in equilibrium with a melt that was close in composition to the least evolved matrix and glass inclusions in Merapi basaltic andesites with <67 and probably >65 wt % SiO2 (Fig. 7a). We rebut the interpretation that the intermediate plagioclase and clinopyroxene compositions and the Al-rich amphibole microphenocrysts represent crystallization at elevated pressures, involving large-scale convection and down-cycling, because this process is unlikely to produce the following features: (1) the commonly observed saw-tooth-like zoning patterns in plagioclase and the characteristic compositional plateau zones of plagioclase and clinopyroxene; (2) the texturally and compositionally...
bimodal amphibole population; (3) the mineral rims with intermediate compositions (they should have developed evolved overgrowths during their return flow to shallower levels of the reservoir). The crystallization of the ≲An70 plagioclase, ≲Wo50 clinopyroxene, and amphibole microphenocrysts with ~13 wt % Al2O3 may thus record crystallization in the pre-eruptive reservoir following magma recharge at an elevated temperature and/or at an elevated melt H2O content.

Plagioclase with a characteristic ≲An70 composition, which matches the compositions common in intermediate zones and rims of the natural crystals, is produced in our melting experiments at 1000°C, and at the same 200 MPa pressure, XH2O of ~0.5–0.6, and a melt H2O content of ~3–4 wt % that we infer for the pre-eruptive crystallization conditions of the resident magma (Table 3; Fig. 8b). Magnetite with ≥13 wt % Al2O3, which matches the rim composition of the natural crystals, also forms at these conditions (Fig. 8f). However, the Wo and Al2O3-rich compositions of clinopyroxene or amphibole that are characteristic of the recharge magma do not crystallize at these conditions (Fig. 8c and d). That the ≲An70 plagioclase and ≲Wo50 clinopyroxene, Al-rich amphibole microphenocrysts, and Al-rich magnetite rims record magma recharge with an increase in temperature alone therefore seems unlikely. In contrast, the assemblage with ≲An70 plagioclase, clinopyroxene with ≲Wo50 and ≥4 wt % Al2O3, amphibole with ~13 wt % Al2O3, and magnetite with ≥13 wt % Al2O3 is produced in our melting experiments at 925–950°C, ~200 MPa, an XH2O of ≥0.8, and with a melt H2O content of ~4–5 wt % (Fig. 8b–f). The most primitive natural matrix and glass inclusion compositions with ~66 wt % SiO2 and ~17 wt % Al2O3 are also closely approached at these conditions (Figs 6 and 8a). In combination, we therefore take the intermediate plagioclase, clinopyroxene, amphibole, and magnetite compositions to estimate that the XH2O and melt H2O content of the recharge magma were ≥0.8 and ~4–5 wt %, and we infer that its temperature was >950°C and <1000°C (Fig. 12c and d).

It is notable in this regard that H2O-rich glass inclusions in natural samples from Merapi, which are inferred to represent, or closely approach, entrainment compositions, have up to ~5 wt % H2O and ≤800 ppm CO2 (Preece et al., 2014). These inclusions with ≥2.5 wt % H2O have compositions indicating equilibration with a vapour phase characterized by an XH2O of between ~0.6 and ~0.8 (Fig. 11a; data from Preece et al., 2014). This variation in glass H2O and CO2 contents and XH2O could reflect the following: (1) analytical uncertainties; (2) analysis of compositionally heterogeneous glass domains and/or microcr or submicron volatile inclusions [as proposed by Preece et al. (2014) for their CO2-rich, but not for their H2O-rich glass inclusion data]; (3) melt inclusions (glass) that variably re-equilibrated (leaked) during magma ascent. However, the compositional variation may also record the primary variations in melt composition and XH2O that we infer on the basis of plagioclase and clinopyroxene compositions; that is, melt entrapment during inter-eruptive magma residence in equilibrium with an XH2O of ~0.6 and melt entrapment during pre-eruptive magma recharge in equilibrium with an XH2O of ~0.8 (Fig. 12).

Origin of the inferred volatile variation in Merapi's pre-eruptive magma reservoir

Our experimental study indicates that the Merapi pre-eruptive magma reservoir is contained within carbonate-dominated strata; previous experimental studies have characterized the release of CO2 by carbonate assimilation in Merapi magmas (Deegan et al., 2010; Blythe et al., 2015). High δ13C (CO2) gas emissions from summit fumaroles during Merapi’s eruption in 2006 have been interpreted as evidence for carbonate assimilation (Troll et al., 2012), which at that time may have contributed significant amounts of CO2 to Merapi’s volatile budget. High CO2 emissions prior to the 2010 eruption may also reflect CO2 liberated by carbonate assimilation (Borisova et al., 2013; Blythe et al., 2015), but they may equally reflect CO2 released from the recharge magmas (Surono et al., 2012; Costa et al., 2013). Phenocryst glass inclusions with high CO2 contents may furthermore record CO2 fluxing related to carbonate assimilation (Deegan et al., 2010; Nadeau et al., 2013; Blythe et al., 2015), but the locally high CO2 contents of glass inclusions trapped in Merapi’s phenocrysts are more likely to represent heterogeneous post-entrapment CO2 distribution (Preece et al., 2014). In agreement with others, we therefore propose that Merapi’s volatile phase comprises contributions from the carbonate-dominated upper crust, but that its largely uniform composition predominantly derives from a steady-state magma source comprising mantle and subducted sediment components (Allard, 1983, 2013; Allard et al., 2011; Troll et al., 2012).

That gas fluxing or degassing within Merapi’s magma plumbing system accounts for most of the variations in XH2O in the pre-eruptive reservoir is hence the most likely scenario. One possibility is that magmas reach Merapi’s pre-eruptive reservoir with an XH2O of ≥0.8 (Fig. 12). During storage they are then fluxed by CO2-rich volatiles from deeper levels of the magma system, decreasing the reservoir’s XH2O to ~0.5–0.6 (Fig. 12c). Such CO2-rich volatile fluxing of shallow reservoirs has been inferred for a number of arc volcanoes, including Mount St Helens and Soufrière Hills (Blundy et al., 2010; Cashman & Blundy, 2013; Cassidy et al., 2015; Riker et al., 2015). For the Merapi system this process would require that CO2 fluxing of the pre-eruptive reservoir is spatially fairly homogeneous, given that the largest phenocryst and microphenocryst volume crystallizes at these conditions. The characteristic plateau zones of clinopyroxene and plagioclase phenocrysts would further require that the reservoir’s XH2O reaches a steady state at ~0.5–0.6, which could indicate that the inflowing vapour is close to this composition or
that the influx of CO₂-rich vapour diminishes or ceases once the volatile phase of the pre-eruptive reservoir has reached this composition. In this scenario, Merapi’s deeper magma plumbing system behaves as an open system, releasing volatiles, whereas the pre-eruptive reservoir behaves as a closed or semi-closed system during inter-eruptive periods, trapping some (or the majority) of the volatiles derived from deeper levels (Fig. 12c).

Another possible scenario is that magmas ascending through Merapi’s magma supply system follow different degassing paths—open, vapour-buffered and closed-system degassing paths—during inter-eruptive and pre-eruptive recharge of the reservoir. During inter-eruptive periods, magmas reach the pre-eruptive reservoir with an XH₂Ov of ~0.5–0.6 (Fig. 12), following or approaching a vapour-buffered degassing path, along which both H₂O and CO₂ partially exsolve. During pre-eruptive recharge, in contrast, magmas ascend into Merapi’s pre-eruptive reservoir following or approaching an open-system degassing path, releasing a comparatively large amount of CO₂, and thus reaching the reservoir at a higher XH₂Ov of ~0.8 (Fig. 12d). A comparable, although longer-term switch from closed-system to vapour-buffered degassing paths has been inferred for magmas that sourced the 1980 Mount St Helens’ plinian eruption and its subsequent effusive eruptions on the basis of both mineral assemblages and glass inclusion CO₂ and H₂O compositions (Blundy et al., 2010; Riker et al., 2015). That the supply system of Merapi’s pre-eruptive reservoir has switched between degassing trends in such a fashion prior to 2010 and other recent eruptions (as recorded by equivalent mineral compositions and zoning patterns) could relate to changes in the magma ascent and supply rate, which ultimately trigger Merapi’s small-volume eruptions. We consider this scenario plausible, because it is consistent with all the petrological and compositional data available, and because it can also account for the strong degassing of CO₂ observed prior to the onset of the 2010 eruption (e.g. Surono et al., 2012). However, additional glass and fluid inclusion data and analyses of inter-eruptive and pre-eruptive gas emission data are urgently needed to further evaluate the inferred variation of volatile compositions and the proposed scenarios for volatile evolution.

CONCLUSIONS

The key findings of our study on pyroclasts erupted at Merapi, Indonesia, in 2010, and our experiments on one of its crystal-rich basaltic andesites are as follows.

1. Experiments that partially melt and partially recrystallize coarse-grained powders can constrain the crystallization conditions for both resident and recharge magmas in mixed systems by reconstructing local equilibrium conditions between mineral rim and host melt composition, if both magmas and their melt fractions are closely comparable in composition. The performance of standard crystallization experiments that reconstruct mineral and melt compositions at total equilibrium for the same starting material, and thus for a system with a similar composition, can be used to ascertain that the phase compositions produced in the melting experiments indeed represent local equilibrium.

2. Mount Merapi’s current pre-eruptive reservoir hosts crystal-rich magma that is repeatedly recharged by basaltic andesite magma with a closely corresponding composition. The magmas partially crystallize at ≥100–200 ± 75 MPa and thus at a depth between c. 4.5 and 9 km. They are stored at 925–950 ± 25 °C with melt H₂O contents of ~3–4 wt %, and an XH₂Ov of ~0.5–0.6. Recharge magmas that mix with the resident magma of the reservoir have higher melt H₂O contents of ~4–5 wt %, a higher XH₂Ov of ~0.8, and a higher temperature of >950 to <1000 °C. These variations in melt H₂O and XH₂Ov of the resident and recharge magmas may reflect variable degrees of open-system degassing of the magmas en route to the pre-eruptive magma storage reservoir, which may in turn record variations in magma ascent and supply rates from the deep plumbing system.

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SUPPLEMENTARY DATA

Supplementary data for this paper are available at Journal of Petrology online.

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