

## Experimental phase equilibria constraints on pre-eruptive storage conditions of the Soufriere Hills magma

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**Abstract.** New experimental results are used to constrain the P, T, X(H<sub>2</sub>O) conditions of the Soufriere Hills magma prior to ascent and eruption. The experiments were performed on a powdered andesite erupted in January, 1996, at an fO<sub>2</sub> corresponding to ~NNO+1 with P<sub>H<sub>2</sub>O</sub> and temperatures in the range 50 to 200 MPa and 800 to 940°C. Amphibole is stable at P<sub>H<sub>2</sub>O</sub> >115 MPa and temperatures <875°C. Quartz only becomes stable at low temperatures and after high degrees of crystallization (T <840°C, >72 wt% SiO<sub>2</sub> in residual melt) at P<sub>H<sub>2</sub>O</sub> >115 MPa. Analyses of rhyolitic glass inclusions in quartz and plagioclase from recently erupted samples indicate melt water contents of 4.27±0.54 wt% H<sub>2</sub>O and CO<sub>2</sub> contents <60 ppm. The evolved Soufriere Hills magma would therefore be H<sub>2</sub>O-saturated at pressures <130 MPa.

These results suggest that the Soufriere Hills magma containing the stable assemblage amphibole, quartz, plagioclase, orthopyroxene, magnetite and ilmenite was stored at P<sub>H<sub>2</sub>O</sub> of 115-130 MPa, equivalent to a minimum depth for a water-saturated magma chamber of 5-6 km depth. Magma temperatures were initially low (820-840°C). Quartz is believed to have been destabilised by a heating event involving injection of new basaltic magma. The stability field of hornblende provides a useful upper limit (~880°C) for the extent of this reheating.

### Introduction

Understanding and interpreting the dynamical processes producing the current eruption of the Soufriere Hills volcano requires information about the storage conditions (P, T and X(H<sub>2</sub>O)) of the magma prior to ascent and eruption. The lack of significant seismic signals from the magma storage region, coupled with the problems associated with a small-aperture network [Aspinall *et al.*; this volume] makes determination of the depth of the magma chamber via geophysical methods difficult. Recent phase equilibria studies of subduction-zone magmas [e.g. Rutherford *et al.*, 1985; Moore and Carmichael in press; Martel *et al.*, in press] have proved highly successful in constraining the pre-eruptive P, T and X(H<sub>2</sub>O) conditions of particular magmas via comparison with the naturally occurring mineral assemblages, paying particular attention to phases sensitive to changes in these parameters within the relevant P-T field (e.g. amphibole).

The andesitic lavas erupted from the Soufriere Hills Volcano have ~40 vol% phenocrysts of plagioclase (69%), amphibole (15%), orthopyroxene (12%), magnetite, ilmenite (<5%), and quartz (<2%) in a groundmass composed of microlites and

microphenocrysts of plagioclase, orthopyroxene, clinopyroxene, magnetite, ilmenite, and a silica phase set in a rhyolitic residual glass. Despite the relatively constant bulk composition of the andesites erupted during the current activity, there has been significant textural variation in the mineral assemblage in response to changing dynamical conditions [Devine *et al.* (a), this volume]; both quartz and amphibole show textural evidence of destabilization due to changing P-T conditions prior to and during magmatic ascent and eruption of the magma. Quartz grains commonly show signs of resorption and embayment around their rims and hornblende margins in contact with the melt typically have reaction rims consisting of the assemblage clinopyroxene + orthopyroxene + plagioclase + magnetite + melt. It is the purpose of this paper to quantify the conditions under which the observed phenocryst assemblage is stable using experimentally determined phase equilibria relevant to this system. A second objective is to determine the nature of the heating event that appears to have affected the Soufriere Hills magma just prior to eruption.

### Experimental and analytical techniques

The andesitic samples chosen for the experiments (MVO34) were collected from the growing dome in January 1996. Fe-Ti oxide analyses suggest the magma equilibrated at an fO<sub>2</sub> of NNO+1 (±0.5) log units [Devine *et al.* (b), this volume]. Experiments were performed on both the powdered bulk compositions ('melting' experiments) and on a glass made from this composition by melting it at atmospheric pressure at 1100°C ('crystallization' experiments). Experimental charges consisted of 0.03-0.05g of starting powder and sufficient H<sub>2</sub>O to saturate the melt at the given P and T, both sealed in a 2 or 2.5 mm diameter Ag<sub>70</sub>Pd<sub>30</sub> sample capsule. Lower temperature experiments (800-900°C) were carried out in cold-seal pressure vessels using an Inconel 600 (Ni-rich) filler rod and an H<sub>2</sub>O pressurizing fluid at pressures in the range 50 to 200 MPa and temperatures of 800 to 900°C. Oxygen fugacity in this assembly was buffered at NNO+1 (±0.5) [Geschwind and Rutherford, 1992]. Higher pressure and temperature experiments were carried out in a TZM pressure vessel using a NNO buffer assembly (Brown University) and two melting experiments were carried out in the internally heated pressure vessel (IHPV) at UC Berkeley following the methods of Moore *et al.* (in press). Two experiments were also carried out at 250 MPa in the presence of silver oxalate giving P<sub>H<sub>2</sub>O</sub> = 0.5 P<sub>total</sub>. Experiments were run for time periods spanning 1-10 days dependent on the P, T conditions of the experiment. Similarity in phenocryst proportions and compositions in both the melting and crystallization experiments support a close approach to equilibrium in these experiments.

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The major element compositions of the phenocryst phases and glasses were analyzed using the JEOL 8600SX, Cameca Camebax, and Cameca SX-50 electron microprobes at the University of Bristol, Brown University and UC Berkeley, respectively. Mineral analyses were obtained using an accelerating voltage of 15 kV, with a 15 nA beam current and a focused beam. Analyses were calibrated against well-documented mineral standards and samples show good reproducibility. A defocused beam (20  $\mu\text{m}$ ) and low value beam current were used for glass analyses at Brown University and the University of Bristol in order to minimize alkali mobility [Nielsen and Sigurdsson, 1981]. An on-line volatile element correction procedure [Donovan *et al.*, 1993] was used at UC Berkeley.

Glass inclusion studies used phenocrysts separated from lightly crushed rocks by hand-picking under the binocular microscope. Inclusion-bearing crystals were identified and isolated, mounted in orthodontic resin and then carefully ground using a succession of fine abrasives. An inclusion was exposed and polished on both sides using 3 and 1  $\mu\text{m}$   $\text{Al}_2\text{O}_3$  paper.  $\text{H}_2\text{O}$  and  $\text{CO}_2$  were determined by FTIR spectroscopy at Bristol University using the intensities of OH ( $4500\text{ cm}^{-1}$ ),  $\text{H}_2\text{O}_{\text{mol}}$  ( $5200\text{ cm}^{-1}$ ) and  $\text{CO}_{2\text{mol}}$  ( $2300\text{ cm}^{-1}$ ) absorbance. Glass density was estimated as  $2400\text{ g/cm}^3$  and molar absorptivities of 1.73 l/mole/cm (OH) 1.61 l/mole/cm ( $\text{H}_2\text{O}_{\text{mol}}$ ) and 945 l/mole/cm ( $\text{CO}_{2\text{mol}}$ ) were used [Fine and Stolper, 1985].  $\text{H}_2\text{O}$  was also estimated using electron microprobe analyses and the difference technique of Devine *et al.*, [1995].

### Pre-eruptive volatile contents

Volatile contents measured in quartz- and plagioclase-hosted glass inclusions (Table 1) cover a variety of deposits representing both the time-span of the current eruption and the complete range of eruptive styles encountered so far [Young *et al.*, this volume a].  $\text{H}_2\text{O}$  contents were found to be uniformly high ( $4.27\pm 0.54\text{ wt}\%$ ), with the exception of those from the earliest erupted products where all inclusions have degassed as the result of slow ascent.  $\text{CO}_2$  contents are consistently less than the limits of detection for FTIR (<50 ppm). Although water contents are uniformly high, some variability is present (Table 1). Most of the intact inclusions measured in these samples were relatively thin (<50  $\mu\text{m}$ ) leading to a greater relative degree of uncertainty in the measurement of inclusion thickness. The analytical error on any infrared measurement is considered to be a maximum of 12.5% (as the result of uncertainty due to thickness measurement and background-

fitting procedure). The variability among different samples is therefore not much greater than the uncertainty associated with individual measurements, and the melt water contents are considered to be uniformly high ( $4.27\pm 0.54\text{ wt}\%$ ) in the magma chamber prior to eruption.

The major element compositions of glass inclusions in quartz are relatively evolved ( $\text{SiO}_2$  anhydrous >75 wt%, Table 3) and they also reflect the relative inhomogeneity of the residual glass measured in the natural samples. The major element compositions of plagioclase inclusions show considerably less compositional variation [Devine *et al.*, this volume].

### Experimental Phase Equilibria

Plagioclase, orthopyroxene and Ti-magnetite were found to be stable throughout the conditions investigated with the stability of quartz and amphibole dependent on the  $\text{P}_{\text{H}_2\text{O}}$  and T of the experimental run. Amphibole is stable at  $\text{P}_{\text{H}_2\text{O}}$  greater than 115 MPa and temperatures less than  $875^\circ\text{C}$  (Fig. 1). Quartz stability limits are also shown on Figure 1. Clinopyroxene was found in the phase assemblage at the lower pressure and higher temperature conditions where amphibole was not present, but insufficient data exist to satisfactorily constrain the high temperature stability limit of clinopyroxene. The P-T range over which both quartz and amphibole co-exist is relatively limited and confined to temperatures less than  $840^\circ\text{C}$  and  $\text{P}_{\text{H}_2\text{O}}$  between 170 and 115 MPa.

The mixed  $\text{H}_2\text{O}$ - $\text{CO}_2$  experiments, conducted at 250 MPa ( $\text{P}_{\text{H}_2\text{O}} = 125\text{ MPa}$ ) and 850 and  $825^\circ\text{C}$  did not crystallize quartz. Amphibole compositions have significantly greater  $\text{Al}_2\text{O}_3$  contents than the natural amphiboles, and plagioclase is predominantly more anorthitic than that found in the natural assemblage.

### Melt composition and evolution

The compositions and proportions of glass and minerals have been determined when possible, and these analyses are summarized in Tables 2 and 3. Both the experimental samples and the natural samples are relatively phenocryst rich. Plagioclase rim compositions in the experimental products reflect the broad range of run conditions and range from  $\text{An}_{82}$  to  $\text{An}_{44}$ , with more calcic compositions occurring for higher T and higher  $\text{P}_{\text{H}_2\text{O}}$  conditions. A similar distribution of plagioclase composition is found in the natural samples

Table 1. Volatile contents of glass inclusions measured in plagioclase and quartz

Sample	Unit <sup>(6)</sup>	Phase	OH <sup>(1)</sup>	$\text{H}_2\text{O}_{\text{mol}}$	$\text{H}_2\text{O}_{\text{tot}}$	$\sigma_n$ <sup>(2)</sup>	$\text{CO}_{2\text{mol}}$ <sup>(3)</sup>
MonJul.1	July 29th pyroclastic flow	Quartz	1.45	2.49	3.94	0.25	-
MonAug.1	August 11th pyroclastic flow	Quartz	1.56	1.96	3.52	0.29	-
MonAug.3	August 11th pyroclastic flow	Quartz	1.06	3.01	4.07	0.33	<60
MonAug.4	August 11th pyroclastic flow	Quartz	1.13	3.92	5.05	0.45	-
MonSep.1	17th September pumice	Quartz	1.19	3.46	4.65	0.18	<30
Mon.Dec.1	19th December pumiceous pyroclastic flow	Quartz	1.11	3.29	4.40	-	-
JD.1 <sup>(4)</sup>	December 1995 dome	Plagioclase	-	-	0.00	-	-
JD.2 <sup>(4)</sup>	17th September pumice	Plagioclase	-	-	4.7	-	-

(1) All water contents expressed as wt%; - indicates value not determined

(2) Standard deviation of multiple analyses (3-4) on same inclusion; Mon.Dec.1 is single analysis.

(3)  $\text{CO}_2$  contents expressed as ppm.

(4) Water contents measured by difference using electron microprobe; all others measured by FTIR.

(5) All inclusions measured from samples collected in 1996 unless otherwise stated.

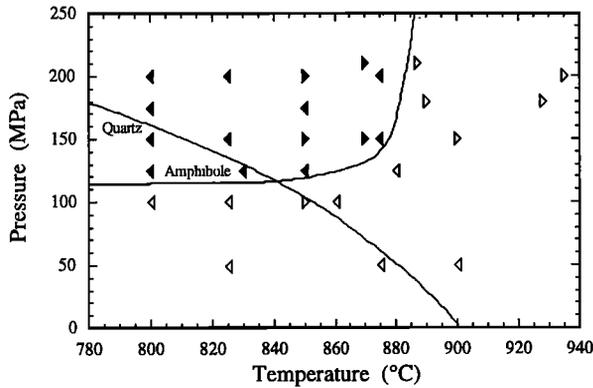


Fig. 1. Stability of quartz and amphibole in Soufriere Hills andesite. Arrows indicate direction of approach to equilibrium; solid = amphibole present. Plagioclase, orthopyroxene and Fe-Ti oxides were also present in every run product.

[Devine *et al.* (a), this volume]. Mafic phase compositions are similar to those found in the naturally occurring assemblage (Tables 2 and 3). Lower pressure run conditions are characterized by amphiboles poorer in aluminum (Table 2).

The relatively small sizes of both crystals and glass pools within many of the experimental samples has made accurate determination of glass compositions difficult. Modal proportions of minerals have been determined by least squares mixing calculations using glass analyses and phase compositions for two experimental runs (Mon6aR45 and M14b, Table 3). The lower pressure experiment (M14b; 130 MPa and 830°C) produced a residual glass composition that closely matches the composition of glass inclusions trapped by plagioclase in the September 17th eruptive products. This glass is less evolved than that found trapped in the quartz crystals and more evolved than the residual glass in the erupted magma.

### Pre-eruptive storage conditions of the Soufriere Hills magma

An indication of the temperature range of the magma prior to eruption is given by geothermometric studies on natural samples. Single pyroxene thermometry [Murphy *et al.*, this volume] has yielded crystallization temperatures ranging from 810°C to 875°C for orthopyroxene cores. Temperature estimates for reverse zoned orthopyroxene rims range between 870 and 990°C, reflecting the varied history of the Soufriere Hills magma chamber. The ubiquitous presence of mafic

inclusions in the eruptive products of the Soufriere Hills volcano provides strong evidence for reheating of the magma chamber via the influx of mafic magma [Murphy *et al.*, this volume; Devine *et al.* (a), this volume]. Fe-Ti oxide geothermometry has also yielded a wide range of temperatures for the Soufriere Hills magma, with equilibration temperatures ranging from 830-940°C [Devine *et al.* (a), this volume].

Measurements of the volatile contents of glass inclusions show that the Qtz-amph-opx-plag-Fe-Ti oxides phenocryst assemblage co-existed with a highly evolved rhyolitic melt containing  $4.27 \pm 0.54$  wt% dissolved  $H_2O$  prior to eruption. Using the solubility model of Moore *et al.* [1998] this is equivalent to  $P_{H_2O}$  of 115-130 MPa.  $CO_2$  concentrations in this melt are relatively low and small amounts of S (<150 ppm; Young *et al.* (b), this issue) have been measured in similar melt inclusions. This corroborates the findings from experimental phase equilibria and supports the point of view that water is the dominant volatile in the Soufriere Hills magma system (both in melt and any co-existing vapor phase). The implied minimum depth of the magma chamber (where  $P_{tot} = P_{H_2O}$ ) is therefore of the order of 5-6 km below the surface. The melt would be vapor saturated at this depth.

Our experimental data show that quartz was part of the crystallizing assemblage at temperatures <840°C and a  $P_{H_2O}$  of 115-130 MPa. Small degrees of heating at these conditions would result in the destabilisation of quartz; e.g. M14b (830°C, 130 MPa) was found not to contain quartz in the crystallizing assemblage, whereas an experiment conducted at 825°C and 130 MPa contained small amounts of quartz. The relatively evolved composition of many quartz-hosted glass inclusions in comparison to the residual melt in experiment M14b suggests that quartz crystallized at lower temperature conditions prior to the eruption of the Soufriere Hills magma. The slightly less evolved glasses trapped in some plagioclase inclusions are compositionally similar to the glass of M14b. This suggests that quartz crystallization took place in a relatively cool, late stage crystal mush with temperatures <840°C at a  $P_{H_2O}$  < 130 MPa. The composition of the residual glass (Mon6aX, Table 3) is also significantly less evolved than that found in the glass inclusions. At a  $P_{H_2O}$  of 115-130 MPa this would suggest re-equilibration at temperatures higher than 840°C, and implies a reheating and resorption of mineral phases following the entrapment of the melt inclusions.

Our phase equilibria and glass inclusion data therefore also strongly suggest that the Soufriere Hills magma has been reheated prior to ascent. Our findings have shown that the original phenocryst assemblage can only have been stable at temperatures <840°C, and that the composition of the residual

Table 2. Representative compositions of mafic phases in natural sample and experimental run products

Experiment	MVO34	MVO34	Mon6aR.11	M14b	Mon6aR.30	Mon6aR.38	Mon6aR.30
Type	Natural	Natural	Crystallisation	Melting	Crystallisation	Crystallisation	Crystallisation
T (°C)			875	830	875	900	830
P (MPa)			150	130	210	150	125
Phase	Amph	Opx	Amph	Amph	Amph	Opx	Opx
SiO <sub>2</sub>	48.33	52.87	47.80	48.11	47.45	53.04	51.30
TiO <sub>2</sub>	1.44	0.14	1.72	1.09	1.37	0.18	0.15
Al <sub>2</sub> O <sub>3</sub>	6.46	0.61	7.55	6.53	6.92	0.63	0.62
FeO	13.61	23.91	15.15	14.50	14.58	23.75	23.78
MnO	0.47	n.d.	0.42	n.d.	1.37	1.45	1.56
MgO	14.73	20.46	14.12	15.86	13.75	19.67	20.99
CaO	11.35	0.99	9.56	9.27	10.86	0.07	1.05
Na <sub>2</sub> O	1.30	0.15	1.27	1.09	1.32	0.01	0.00
K <sub>2</sub> O	0.13	0.05	0.17	0.12	0.16	0.01	0.00
Total	97.82	100.83	97.75	97.08	97.03	99.95	99.45

Table 3. Glass compositions and phase proportions of Natural Sample and experimental run products

Sample	MVO34 <sup>(3)</sup>	MVO34 <sup>(3)</sup>	Mon6aX <sup>(5)</sup>	jd.m153	Monaug.3	Mon6ar45	M14b
P (MPa)	n/a	n/a	n/a	n/a	n/a	180	130
T (°C)						900	830
Glass Composition <sup>(1)</sup>				plag. inclusion	quartz inclusion		
SiO <sub>2</sub>	59.15	59.46	71.41	76.27	79.13	68.13	77.00
TiO <sub>2</sub>	0.63	0.66	0.28	0.20	0.12	0.39	0.23
Al <sub>2</sub> O <sub>3</sub>	18.29	17.90	13.58	12.88	10.87	16.87	12.90
FeO	6.75	6.89	2.78	1.52	1.27	3.24	1.42
MnO	0.19	0.23	0.13	0.10	0.00	0.18	0.10
MgO	2.91	2.77	1.64	0.29	0.26	1.05	0.36
CaO	7.57	7.53	4.86	2.23	1.52	5.01	2.30
Na <sub>2</sub> O	3.59	3.64	3.73	4.43	4.99	3.83	3.73
K <sub>2</sub> O	0.77	0.81	1.6	2.07	1.86	1.32	1.95
H <sub>2</sub> O <sup>(2)</sup>	100.00	99.82	-	4.7	4.07	6.10	4.30
% glass	60	60	n/a	n/a	n/a	68	45
% phenocrysts <sup>(4)</sup>	40	40	n/a	n/a	n/a	32	55

(1) Glass data normalized to anhydrous composition, for ease of comparison.

(2) H<sub>2</sub>O determined from P for Mon6ar45 and M14b using solubility model of Moore et al., (1998) Monaug.3 determined by FTIR (Table 1).

(3) Col. 1 and 2 are XRF analyses of fused MVO34 samples at Bristol and Brown respectively; % glass does not include microlite growth.

(4) Phase proportions in MVO34 are Opx:Cpx:Amph:Fe-Ti oxides:Plag = 12:0:15:4:69 [Devine et al., this volume]; proportions in Mon6ar45 determined by least squares mixing are 16:11:0:11:62 and in M14b are 10:0:15:9:66.

(5) Average composition of groundmass determined from rastered electron microprobe analysis

melt found in the erupted products must have been produced as the result of reheating to temperatures greater than 840°C. This supports the petrological evidence for magma mixing and reheating [Murphy et al., this volume]. It is likely that localized reheating occurs at temperatures significantly greater than this lower bound. The stability field of hornblende provides a useful upper limit (~880°C) for the extent of this reheating, as no signs of pervasive heating-induced hornblende breakdown have yet to be found in the eruptive products.

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