



## Surface tension of hydrous silicate melts: Constraints on the impact of melt composition



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### ABSTRACT

The first step in magma degassing is the nucleation of gas bubbles. The ability of magma to nucleate bubbles is moderated by its surface tension, which is thought to vary with melt composition, temperature, and H<sub>2</sub>O content. Numerous experimental studies of bubble nucleation in silicate melts have quantified surface tension, but those experiments have been run at different temperatures and used melts with different dissolved H<sub>2</sub>O contents. The influence of bulk melt composition may thus be masked. In this study, we decompress hydrous silicate melts that range from phono-tephrite to high-silica rhyolite to investigate conditions needed to trigger homogeneous bubble nucleation. Importantly, dissolved water contents are very similar amongst the melts, and all were decompressed at 1150–1200 °C, isolating the influence of melt composition on bubble nucleation. Despite the 25 wt.% range in SiO<sub>2</sub> content, both the total pressure drop and the degree of supersaturation needed to trigger bubble nucleation vary a little. Because supersaturation varies little,  $\sigma$  for all melts falls to fall within a relatively narrow range of 0.065 to 0.080 N m<sup>-1</sup>. In addition,  $\sigma$  for an even wider range of anhydrous melts is nearly constant, although five times higher. It thus appears that the bulk composition of silicate melt has little impact on  $\sigma$ . It is also known that the variation in  $\sigma$  with temperature is minor, and thus most differences between measured  $\sigma$  values probably result from differences in H<sub>2</sub>O content.

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### 1. Introduction

Volcanic eruptions are driven by the exsolution of volatiles that were dissolved in magmas at depth. In addition, many of the world's significant ore bodies are generated by volatiles released from magmas (Misra, 2000; Seedorff et al., 2005). For magma (i.e., silicate melt) to release volatiles and degas it must become supersaturated with them, of which H<sub>2</sub>O is usually the most abundant (Wallace and Anderson, 2000). Magma can supersaturate with H<sub>2</sub>O through anhydrous crystallization, which concentrates H<sub>2</sub>O in the melt, or release of pressure, because water is less soluble in silicate melt at lower pressure (e.g., Hamilton et al., 1964; Burnham, 1975; Moore et al., 1998).

The first and most critical step in degassing is for gas bubbles to nucleate, with bubbles nucleating faster as supersaturation increases (Hurwitz and Navon, 1994). The ability for hydrous, silicate melt to nucleate bubbles is, however, mitigated by the surface tension ( $\sigma$ ) of the silicate melt (Sparks, 1978). In the absence of crystals, a relatively high  $\sigma$  necessitates significant supersaturations to trigger bubble nucleation, which can be seen from the bubble nucleation rate ( $J$ )

$$J = J_0 \exp\left(\frac{-16\pi\sigma^3}{3kT\Delta P^2}\right) \quad (1)$$

where  $\sigma$  is cubed,  $\Delta P$  is the amount of supersaturation,  $k$  is Boltzman's constant,  $T$  is temperature, and  $J_0$  is the rate at vanishing free energy barrier to nucleus formation (Hurwitz and Navon, 1994). For example,  $\sigma$  of hydrous, rhyolitic melt typically exceeds 0.07 N m<sup>-1</sup> (Mourtada-Bonnefoi and Laporte, 1999; Mangan and Sisson, 2000; Mourtada-Bonnefoi and Laporte, 2002, 2004; Gardner and Ketcham, 2011), and so for even 1 bubble to nucleate in a cubic meter every second (an exceedingly slow rate)  $\Delta P$  must exceed 76 MPa, which equals about 3 km rise in the crust.

It is thus clear that to understand the conditions necessary for magmas to generate bubbles, surface tension must be well known. It is thought that surface tension varies with melt composition, dissolved H<sub>2</sub>O content, and temperature (Walker and Mullins, 1981; Mourtada-Bonnefoi and Laporte, 1999; Bagdassarov et al., 2000; Mangan and Sisson, 2000; Mourtada-Bonnefoi and Laporte, 2002, 2004; Mangan and Sisson, 2005; Gardner and Ketcham, 2011; Gardner, 2012). To fully model variations in surface tension during storage and eruption of magma, how much each of those parameters impacts surface tension must be constrained. Recent experimental work implies that high degrees of supersaturation are needed to trigger homogeneous bubble nucleation in rhyolitic melts (Mourtada-Bonnefoi and Laporte, 1999; Mangan and Sisson, 2000; Mourtada-Bonnefoi and Laporte, 2002; Gardner and Ketcham, 2011). Dacitic and phonolitic melts also appear to require substantial oversaturation, although less than rhyolites (Mangan and Sisson, 2005; Gardner and Ketcham, 2011; Gardner, 2012). Often, however, experimental measurements of surface tension

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are done at different H<sub>2</sub>O contents and temperatures, and so the impact solely of melt composition is masked. In this study, silicate melts of different compositions, ranging from phono-tephrite to high-silica rhyolite (Fig. 1), that have very similar dissolved H<sub>2</sub>O contents were decompressed at similar temperatures (1150–1200 °C) to investigate the impact of melt composition on the ability of bubbles to nucleate.

## 2. Methods

### 2.1. Samples and experimental techniques

All experiments used cylinders cored from samples of basaltic andesite, phono-tephrite, trachyte, dacite, or high-silica rhyolite (Fig. 1). Most cylinders were 1.1–1.3 cm long and 2.2 mm wide. Sharp edges were ground with emery cloth to avoid piercing the metal capsule in which the cores were held. The cylinders were cleaned after grinding.

All experiments consisted of a hydration and then decompression. Most hydrations consisted of one cylinder with distilled water inside a precious metal capsule. In most cases, the amount of water was usually ca. 10% by weight of the cylinder, more than enough to ensure that a fluid existed throughout the hydration. Once the capsule was welded shut, it was weighed and then heated on a hotplate for 10–15 min, and then weighed again to check for leaks and water loss. Hydrations were carried out at temperatures and pressures that depended on melt composition (Table 2). Rhyolite cores were hydrated at 875 °C and 160 MPa, following the methods of Gardner and Ketcham (2011). Experimental techniques for the other starting materials are described below.

Phono-tephrite, trachyte, and dacite cores were hydrated at 150 MPa and 1100–1150 °C in 3-mm O.D. Au<sub>80</sub>Pd<sub>20</sub> capsules in titanium–zirconium–molybdenum (TZM) or molybdenum–hafnium carbide (MHC) pressure vessels, pressurized by argon plus one to two bars of methane to slow hydrogen loss from the capsule (Sisson and Grove, 1993). Even with the added methane, numerous runs showed that hydrogen loss became significant after about 48 h, and so each hydration consisted of two runs. First, a capsule ran for two days and was quenched. The sample was extracted from its capsule, placed inside a new one with water, and run for another day, for a total of 64–74 h (Table 2). Pulling the pressure vessel out of the furnace and inverting it quenched the samples by causing them to drop to the water-cooled end of the vessel.

Basaltic andesite cores were hydrated at 1190 °C and 500 MPa in 3-mm O.D. Au<sub>80</sub>Pd<sub>20</sub> capsules, run in a piston cylinder apparatus, following methods similar to those of Moore et al. (2008). All were hydrated for 24 h, and then quenched to room temperature within 1 min by turning off the power to the piston cylinder. The amount of H<sub>2</sub>O added to each capsule equaled ~4.6 wt.% of the core weight, and run conditions were

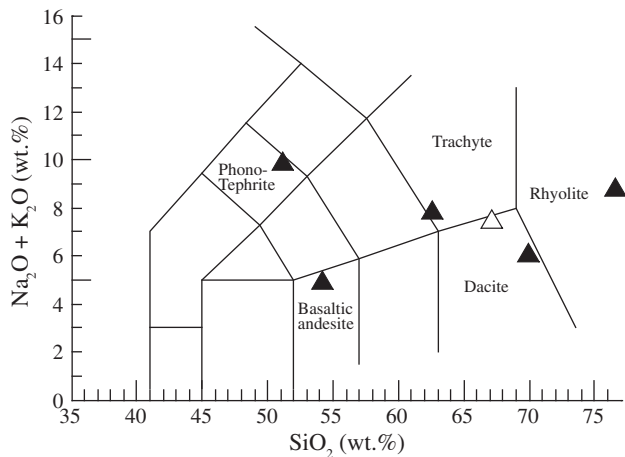


Fig. 1. Compositions of samples used in this study (Table 1). Also shown (open triangle) is the composition of the dacite used in Gardner and Ketcham (2011).

Table 1  
Composition of silicate melts used in this study.

	Trachyte	Dacite	Rhyolite	Phono-tephrite	Basaltic andesite	D(G&K)
SiO <sub>2</sub>	62.57	69.85	76.53	51.13	54.12	66.93
TiO <sub>2</sub>	0.96	0.83	0.06	0.71	2.33	1.02
Al <sub>2</sub> O <sub>3</sub>	16.63	16.04	13.01	19.76	16.23	17.13
FeO*	4.99	1.05	0.79	6.55	9.45	1.15
MnO	0.1	0.12	0.08	0.13	0.21	0.09
MgO	2.26	1.32	0.02	2.83	4.08	1.62
CaO	4.35	4.74	0.74	8.51	8.22	4.21
Na <sub>2</sub> O	5.49	5.18	3.87	2.11	4.02	4.97
K <sub>2</sub> O	2.3	0.86	4.91	7.74	0.87	2.58
P <sub>2</sub> O <sub>5</sub>	0.35			0.53	0.47	0.32

Composition of samples used in this study by electron microprobe (normalized to 100%), with all Fe reported as FeO; oxides are in wt.%. D(G&K) = dacite from Gardner and Ketcham (2011).

undersaturated in H<sub>2</sub>O, and all H<sub>2</sub>O dissolved into the liquid. Weighing the capsule before and after running it in the piston cylinder indicates that none lost weight. One sample was removed from its capsule and its dissolved H<sub>2</sub>O content was found to agree well with the amount of H<sub>2</sub>O added to the capsule (Table 2).

Except for basaltic andesites, most hydrated samples were removed from their capsules and split, with one piece being used to measure dissolved H<sub>2</sub>O contents by Fourier-transform infrared spectroscopy (see below) and the other used in a decompression experiment (Table 3). Four samples were split into three and two were used in decompressions. Basaltic andesite samples were simply removed from the piston cylinder, cleaned, and then used in decompression experiments, without removing them from their capsules.

Decompressions were run at 1150 °C, except for basaltic andesites, which were run at 1200 °C (Table 3). Most consisted of a hydrated cylinder and distilled water inside 3-mm O.D. Au<sub>80</sub>Pd<sub>20</sub> capsules run in TZM or MHC pressure vessels, pressurized by argon only. Each was heated for ca. 45 min to ensure thermal equilibrium at high pressure

Table 2  
Experimental hydration conditions.

Run	Starting material	P <sub>H</sub> (MPa)	T (°C)	Time	[H <sub>2</sub> O] (wt.%)
G-1037	Trachyte	150	1150	72	–
G-1039	Trachyte	150	1150	72	–
G-1060	Trachyte	150	1150	69	4.44 ± 0.04
G-1047	Trachyte	150	1150	70	4.77 ± 0.13 [4.62 ± 0.62]
G-1056	Trachyte	150	1150	70	4.87 ± 0.06 [4.24 ± 0.50]
G-1074	Trachyte	150	1150	64	4.13 ± 0.04
G-1141	Trachyte	150	1150	70	4.31 ± 0.08
G-753	Dacite	150	1100	72	4.59 ± 0.04
G-770	Dacite	150	1100	72	4.84 ± 0.02
G-771	Dacite	150	1100	74	4.61 ± 0.04
G-1038	Dacite	150	1150	72	4.80 ± 0.04
G-1135	Dacite	150	1150	72	4.74 ± 0.03
G-1090	Phono-tephrite	150	1150	64	4.36 ± 0.07
G-1093	Phono-tephrite	150	1150	70	4.49 ± 0.10 [4.62 ± 0.05]
G-1100	Phono-tephrite	150	1150	66	–
G-1108	Phono-tephrite	150	1150	66	4.40 ± 0.10 [4.80 ± 0.55]
G-1127	Phono-tephrite	150	1150	64	4.37 ± 0.06
G-1130	Phono-tephrite	150	1150	64	4.37 ± 0.05
G-1155	Phono-tephrite	150	1150	63	4.43 ± 0.06
G-1169	Phono-tephrite	150	1150	63	4.59 ± 0.25
NBC-2-2	Basaltic andesite	500	1190	24	4.63
NBC-2-4	Basaltic andesite	500	1190	24	4.44
NBC-2-6	Basaltic andesite	500	1190	24	4.64
NBC-2-7	Basaltic andesite	500	1190	24	4.85
NBC-2-8	Basaltic andesite	500	1190	24	4.57
G-1010	Rhyolite	160	875	120	5.12 ± 0.01
G-1011	Rhyolite	160	875	120	4.99 ± 0.09

P<sub>H</sub> = hydration pressure of the experiment. Times are hours at hydration pressure. [H<sub>2</sub>O] is the average total water content (±1σ) measured by FTIR; water for NBC-2 samples by weight of water added to experiment. Numbers in brackets are water contents measured by SIMS.

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