



Density of phonolitic magmas and time scales of crystal fractionation in magma chambers



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ABSTRACT

Understanding magmatic processes and their evolution in the Earth's interior requires a better knowledge of silicate melts and their physical properties. Among them, density is one of the most important to constrain the residence or ascent time of magma in the upper mantle and crust. However, the volumetric properties of volatile-bearing and highly polymerized silicate melts are still poorly constrained due to scarce experimental data. In this study, the density of dry and hydrous (4.35 wt% H₂O) phonolitic melt was measured *in situ* using the X-ray absorption method in a Paris–Edinburgh press at 0.73–3.1 GPa and 1484–1855 K. Calculated melt densities range from 2.49 ± 0.02 to 2.66 ± 0.03 g/cm³ and from 2.31 ± 0.02 to 2.52 ± 0.02 g/cm³ for the dry and hydrous compositions at depths of 30 to 100 km. The results are used to calibrate the equation of state (EoS) of phonolitic liquids for crustal and upper mantle conditions and to derive the partial molar volume of water: a least-squares fit of the *P–T–ρ* data to a third-order Birch–Murnaghan EoS yields: $V_0 = 28.10^{+0.10}_{-0.74}$ cm³/mol, $K_T = 14.6^{+3.2}_{-3.0}$ GPa, $K' = 8.9^{+3.3}_{-2.6}$ and $\alpha = 47^{+28}_{-30} \times 10^{-6}$ /K for the dry melts and $V_0 = 20.6^{+7.5}_{-0.8}$ cm³/mol, $K_T = 4.6^{+2.3}_{-3.0}$ GPa, $K' = 5.5^{+6.1}_{-0.5}$ and $\alpha = 135^{+114}_{-115} \times 10^{-6}$ /K for the hydrous component at 1673 K. Combined with literature data, our EoS for water indicates that $\bar{V}_{\text{H}_2\text{O}}$ does not depend strongly on the silicate liquid composition at the investigated conditions.

Crystal settling velocities in phonolitic liquids were calculated from the EoS to constrain the time scales of crystal fractionation in magma chambers. The high compressibility of phonolitic melts results in a small density contrast between nepheline and sanidine crystals and melt ($\Delta\rho_{\text{neph-phon}} = 0.13\text{--}0.27$ g/cm³ and $\Delta\rho_{\text{san-phon}} = 0.05\text{--}0.21$ g/cm³) with corresponding settling times of about 380 and 510 yr/km, respectively, for crystal sizes of 2 cm. Because of their higher density, similar-sized magnetite and augite only require about 31 and 100 yr/km, respectively. The observed phenocryst populations in phonolitic magmas indicate that the residence time was sufficiently long to fractionate augite and magnetite, but not sanidine and nepheline. Thus, our density data, combined with the phenocryst populations of the erupted lavas point to a residence time in the crust between a few tens to hundreds of years, consistent with estimates from U–Th–Ra isotope compositions.

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1. Introduction

Silicate melts play a key role in igneous and differentiation processes within the Earth and their physical properties are essential parameters for a better understanding of the Earth's interior. Together with viscosity, magma density affects magma mixing, volcanic activity, melt segregation, fractionation, crystallization and partial melting processes (Bagdassarov et al., 1999; Ohtani, 1983; Ridgen et al., 1984; Stolper et al., 1981).

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Phonolitic magmas occur worldwide in various geological locations (i.e. Tambora, Vesuvius, Eifel Laacher See, Tenerife – Canary Islands, Mount Erebus in Antarctica and Kenya rift zone in Africa) and are commonly characterized by small volume eruptions compared to other compositions. Nevertheless, there have been some events, which show tremendous volume of erupted phonolite (i.e. the Kenya Plateau phonolites or Tambora 1815 eruption). Indeed, the Plateau phonolites from the Kenya rift have an estimated volume of 50,000 km³, exceeding the total combined volume of phonolitic lava found elsewhere in the world by several orders of magnitude (Lippard, 1973; Williams, 1972). Therefore, they have attracted significant attention as a model setting to understand the mechanism of phonolitic magma generation. These alkali-rich magmas are thought to form: (i) as fractionation products from low-silica melts, with primary compositions such as

nephelinite and melilite (Edgar, 1987) or (ii) by direct melting or fractionating of the mantle (Bailey, 1987; Price and Green, 1972; Thompson et al., 2001). Based on experimental and geochemical data it has been suggested that these Plateau-type phonolites were derived from partial melting of plume-derived alkaline basaltic parents in the lower crust, although some samples show evidence for fractional crystallization (Hay and Wendlandt, 1995; Hay et al., 1995; Kaszuba and Wendlandt, 2000). Although the density contrast between melt and crystal has an important effect on crystal fractionation and magma differentiation rates, density data for alkaline magmas typically found in continental rift zone setting are not available, precluding the assessment of the efficiency of crystal fractionation processes as a mechanism to generate phonolitic melts. In this perspective, the time scale of crystallization and differentiation plays a major role and is mainly dominated by the crystallization rate and crystal settling in the magma chambers during cooling. U–Th–Ra isotope data suggest differentiation ages from a basanite to a phonolite composition in the order of $\sim 10^5$ yr, whereas the residence time before eruption (i.e. differentiation of a more evolved magma) is dated to be within few hundred years (Hawkesworth et al., 2000; Reagan et al., 1992). Knowledge of the densities of melts and crystals will help to derive time scales for magma storage, which holds the link between fractionation and volcanic eruption.

Despite the importance of magma density, direct measurements at high pressures and temperatures relevant for geological processes are scarce due to experimental challenges. Although different methods have been applied to determine the density of magmas at high pressures, including the sink/float method (Agee, 2008b; Ghosh et al., 2007; Jing and Karato, 2012; Matsukage et al., 2005; Sakamaki et al., 2006), shock compression (Akins et al., 2004; Ridgen et al. 1984, 1988), internally heated pressure vessel techniques (Burnham and Davis, 1971), X-ray microtomography (Leshner et al., 2009; Xiao et al., 2010) and most recently the X-ray absorption method (Katayama et al., 1993; Sakamaki et al., 2009, 2010; Sakamaki et al., 2011; Sanloup et al., 2000; van Kan Parker et al., 2012), the density of magmas that typically reach the Earth's surface, such as basaltic, phonolitic, andesitic, or granitic compositions, remain unknown for the conditions at which they are generated. Moreover, water is the most important volatile in the Earth's interior and strongly affects the physical and chemical properties of silicate melts. The density and compressibility of hydrous silicate melts may be affected by changes in the partial molar volume of dissolved water in the melt that are not well constrained for a broad range of magmatic compositions, including alkaline silicate melts.

In this study, we report *in situ* density measurements of dry and hydrous phonolitic melts at crustal and upper mantle conditions (1.0–3.1 GPa, 1585–1855 K and 0.73–2.95 GPa, 1484–1618 K, respectively) in a Paris–Edinburgh press (PEP) using synchrotron X-ray absorption. This method allows *in situ*, contact-free liquid density measurements at elevated pressures and temperatures. The equation of state (EoS) was calibrated up to 3.1 GPa and allows to predict the evolution of the partial molar volume of water in alkaline silicate liquids. The EoS is used to calculate crystal settling velocities, constraining the storage time of phonolitic magma and the mechanism that generates phonolitic magmas.

2. Experimental methods

2.1. Sample description and characterization

The starting materials are synthetic haplo-phonolitic glasses with a composition similar to the Plateau flood phonolites from

the Kenya rift (Hay and Wendlandt, 1995). To avoid uncertainties in the density determinations due to iron loss to the assembly material during the experiments, the FeO and Fe₂O₃ contents in natural phonolites [$\text{Fe}^{2+}/(\text{Fe}^{2+} + \text{Fe}^{3+}) = 0.72$] (Lippard, 1973) were replaced by MgO and Al₂O₃, respectively. A haplo-phonolitic glass was first prepared at ambient pressure by grinding SiO₂, TiO₂, MgO, CaCO₃, Na₂CO₃, K₂CO₃ and AlOOH powders in an agate mortar. This mixture was placed in a furnace and quenched from 1673 K to ensure dehydration/decarbonization. The water-bearing glass was prepared by adding 4.5 wt% of liquid H₂O to the dry glass powder. The mixtures were sealed in platinum (Pt) capsules and compressed in an end-loaded piston-cylinder apparatus (Boyd and England, 1960) using a talc–silica–MgO assembly and a graphite heater to synthesize bubble-free glasses that permit accurate density measurements. Both compositions were equilibrated for 3 h at 1673 K and 2 GPa and quenched with an estimated quench rate of 100–200 K/s by switching off the furnace power. The densities of the resulting glasses were determined with the sink/float method in a diiodomethane–acetone mixture. The reproducibility for repeated measurements is better than 0.1% and the density of a glass standard was reproduced within 0.5%. The measurements yielded 2.470 ± 0.013 g/cm³ and 2.428 ± 0.012 g/cm³ for the dry and water-bearing glasses, respectively (Table 1).

The major element composition of the starting glasses and the recovered glass samples from *in situ* density measurements were analyzed using a Jeol JXA-8200 electron probe microanalyzer (EPMA) at ETH Zurich (Table 1). Analyses were performed with peak counting times of 40 s, using a voltage of 15 kV, 4 nA beam current and a spot size of 30 μm . Alkali counting rates were stable under these conditions. The volatile content was analyzed by Fourier transform infrared spectroscopy (FTIR) on double-side polished sections (Table 1) using molar absorption coefficients from the literature (Behrens et al., 2009; Carroll and Blank, 1997; Mandeville et al., 2002; Morizet et al., 2002; Stolper, 1982). The infrared absorption spectra were collected with a Bruker Hyperion 3000 microscope connected to a Vertex 70 interferometer. The observed 10% relative uncertainty mostly arises from uncertainty on the molar absorption coefficients (Behrens et al., 2009; Carroll and Blank, 1997; Mandeville et al., 2002; Morizet et al., 2002; Stolper, 1982). The water content for the dry starting glass was lower than 200 ppm while CO₂ and carbonate were below the limit of detection (± 70 ppm); the water-bearing starting glass has an initially water-content of 4.35 wt%.

The recovered run products are homogeneous in composition (Fig. 1 and Fig. S1a–b), CO₂ free and agree with the starting composition within analytical uncertainty for the major oxide components (Table 1). The water content of the dry runs run2 and run5 increased to 0.13 and 1.05 wt% respectively, probably due to diffusion into the diamond capsule through the Pt lids during the 5–11 h long experiments (Fig. S2). The water contents of quench products from the water-bearing experiments agree with that of the starting glass within 0.35 wt% (Table 1). Secondary Ion Mass Spectrometry (SIMS) analyses indicate that carbon and B₂O₃ contamination, from the diamond capsule and the hexagonal boron nitride (hBN) pressure medium, did not exceed 0.2 and 0.8 wt%, respectively for the two dry runs (Fig. S3a–d). The B₂O₃ contents in the hydrous runs were lower than 0.08 wt% for short runs (~ 2 h) and did not exceed 0.4 wt% for longer runs (~ 4 h) as determined from LA-ICPMS analyses (Table 1). While the measured elemental carbon content should have a negligible effect on the phonolitic melt density, the measured water and B₂O₃ contents in the dry runs will decrease the density by at most 1.5 and 0.4% respectively, based on the effect reported for other melt compositions (Knoche et al., 1995). Because the effect is within the level of uncertainty of the density measurements and it is unclear if the H₂O and B₂O₃ contamination occurred before or after the den-

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