



Water diffusion in potassium-rich phonolitic and trachytic melts



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ABSTRACT

Water diffusivity was investigated in phonolitic and trachytic melts containing up to 6 wt.% of dissolved water at temperatures between 1373 K and 1673 K for running time of 108 to 1186 s using the diffusion couple technique. The experiments were performed in an internally heated gas pressure vessel (IHPV) at pressures of 0.2 or 0.3 GPa and in a piston cylinder apparatus (PCA) at pressures between 0.5 and 2.5 GPa. A newly developed rapid heating and rapid quench device was used for short term experiments in the IHPV. Concentration profiles of hydrous species (OH groups and H₂O molecules) and total water (bulk water concentration as sum of OH and H₂O molecules) were measured along the cylindrical axis of the diffusion sample using IR micro-spectroscopy. The IR spectroscopic technique was calibrated using a set of samples with bulk water contents measured by Karl–Fischer titration. Electron microprobe traverses show no significant change in relative proportions of anhydrous components along H₂O profiles, indicating that our data can be treated as effective binary diffusion between H₂O and the rest of the silicate melt.

Bulk water diffusivity was derived from profiles of total water using a modified Boltzmann–Matano method as well as using fittings assuming a functional relationship between the total water diffusivity ($D_{\text{H}_2\text{O}_t}$) and the total water concentration ($C_{\text{H}_2\text{O}_t}$). The fitting of the profiles indicates that for phonolitic melt the water diffusivity is roughly proportional to water content. The following formulation was derived for the prediction of total water diffusivity (m^2/s) as a function of T (K) in the T -range of 1373 to 1673 K and $C_{\text{H}_2\text{O}_t}$:

$$\log D_{\text{H}_2\text{O}_t} = -7.11 - 2.07 \log C_{\text{H}_2\text{O}_t} - \frac{(4827 - 4620 \log C_{\text{H}_2\text{O}_t})}{T}$$

The experimental data are reproduced by this relationship with a standard deviation of 0.07 log units. Water diffusivity in trachytic melts is similar at the same conditions. A pressure effect on water diffusivity could not be resolved in the range 0.2 to 2.5 GPa for phonolitic or trachytic melts.

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1. Introduction

Water is the most abundant volatile component in natural magmas. The water content strongly affects the physical and chemical properties of silicate magmas, such as viscosity, density and diffusivity, and, hence, it is a crucial parameter controlling magmatic processes from fluid–melt interaction to magma mingling and mixing and volcanic eruption (Shaw, 1963; Watson, 1981). The eruptive style of a volcano is controlled by the kinetics of volatile release and its effect on rheological properties of the magma. Fundamental processes are bubble nucleation and growth, which are strongly affected by water diffusion in the melt (Kerr, 1995; Toramaru, 1995; Navon et al., 1998; Proussevitch and Sahagian, 1998; Zhang, 1999; Liu and Zhang, 2000; Martel et al., 2000; Zhang and Xu, 2003, 2008). It is therefore essential for modeling of various volcanic and magmatic

processes to broaden our knowledge on water diffusion in silicate melts.

The diffusion of H₂O in rhyolitic, basaltic, andesitic and dacitic melts has been extensively studied in the past (see review of Zhang and Ni, 2010). Diffusion of water in silicate melts is some orders of magnitude faster than Si and Al diffusion but it is slower than diffusion of fast elements such as sodium and lithium (Watson, 1994). For volatile components, H₂O diffusion is faster than CO₂, SO₂, Ar, Kr and Xe but slower than He, H₂, and Ne (Behrens, 2010; Zhang and Ni, 2010; Zhang et al., 2010). So, for a given time, H₂O can migrate deeper into a contiguous melt than most other volatile components except for He, Ne and H₂. At the same time, dissolved water reduces the melt viscosity and enhances diffusion of slow elements.

In silicate melts and glasses the total water is dissolved at least in two hydrous species: OH[−] groups and H₂O molecules (H₂O_m). It is important to consider the role of speciation in diffusion because H₂O_m and OH[−] possess different diffusivities. For polymerized melts such as rhyolite it is meanwhile accepted that molecular H₂O is the

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mobile species while OH⁻ groups are immobile (Zhang and Ni, 2010). For depolymerized melts such as dacite and possibly basalt, there is indication that hydroxyl groups may also contribute significantly to the diffusive transport of water (Behrens et al., 2004). In order to get deeper insight into the mechanisms of water diffusion, it is helpful to measure water species concentrations after diffusion experiments. This may allow constraining individual diffusivities of H₂O_m and OH⁻ groups (Zhang et al., 1991). However, unless the experimental temperature is low enough (e.g., ~773 K but depending on the water content and melt composition), concentrations of H₂O_m and OH⁻ groups in quenched melts do not represent the experimental conditions but speciation was frozen in at the glass transition (Dingwell and Webb, 1990; Zhang et al., 1995). Hence, information about the speciation at experimental conditions is often not available.

The aim of this work is to study water diffusion in phonolitic and trachytic melts at high temperature and pressures. Phonolite and trachyte are silica rich effusive rocks presenting high viscosity magmas with high degree of fragmentation during eruption, therefore producing some of the most powerful and destructive volcanic eruptions. Our experiments aim to provide data to quantify bubble growth and degassing in these eruptive systems. By combining our new data with previous results, we also attempt to discuss the mechanism of water diffusion and to describe its dependence on temperature and pressure.

2. Experimental and analytical procedures

2.1. Starting materials

As starting materials, synthetic analogs of potassium-rich phonolite and trachyte were made to match the phonolite whole rock composition of the Upper Lacher See Tephra (Smincke, 1997; Wörner and Smincke, 1984; Behrens and Hahn, 2009) and the composition of a trachyte from the basal unit of the Breccia Museo deposit from the Phlegrean Field (Italy) (Ricci, 2000; Freda et al., 2003; Di Matteo et al., 2004).

Dry glasses were synthesized by fusion of oxides and carbonates at 1873 K for 2 h in open platinum crucibles at ambient pressure in a conventional chamber furnace (Ohlhorst et al., 2001). To improve homogeneity, the quenched glass was crushed and melted again for another 2 h at the same temperature. The glass was finally rapidly quenched onto a brass plate.

The composition of glasses, displayed in Table 1, was analyzed by electron microprobe, Cameca SX100. The measurements were realized using 5 nA beam current, 15 kV accelerating voltage, a defocused beam with 20 μm in diameter and 8 s counting time with the exception of Na and K for which 2 s counting time was used to avoid alkali

loss in particular for the wet samples. Composition of the phonolite is close to the natural analog while the synthesized trachyte has higher iron content compared to the natural trachyte. The ratio of non-bridging oxygen over tetrahedral cations (NBO/T) is close to zero for both anhydrous melts. In the calculation of NBO/T the Fe³⁺/Fe_{total} ratios determined via colorimetry for air-melted glasses was used and Fe³⁺ was assumed to be a network former and Fe²⁺ a network modifier. The atomic ratio of Na/K is 1.32 for the phonolite and 1.09 for the trachyte.

For preparation of hydrous samples, the obtained dry glasses were crushed and sieved to obtain two fine powder fractions with grain size of 200–500 μm and less than 200 μm. The two fractions were mixed in weight ratio of 1:1 to minimize the pore volume. The obtained mixture was loaded in Au₈₀Pd₂₀ capsules (30 mm length, 4 mm diameter, 0.2 mm wall thickness) and doped with doubly distilled water in several steps to improve the homogenization. The heating was carried out in an Internally Heated Pressure Vessel (IHPV) pressurized with Ar at 1473 K and 0.4 GPa for approximately 20 h. Oxygen fugacity is not controlled but is typically above NNO (Schuessler et al., 2008). Zhang and Ni (2010) showed that at 1373–1673 K molecular hydrogen diffusion does not significantly increase total hydrous component diffusivity as long as oxygen fugacity is higher than NNO-1. The samples were quenched isobarically, following a ramp (50 K/min to 673 K, 10 K/min from 673 to 573 K, 5 K/min to room temperature) defined to reduce the stress in the glass. The weight of each capsule was measured before and after annealing to test for possible leakage. Sample assemblages were always H₂O-undersaturated at experimental conditions so that water was completely dissolved in the melt and bubble-free glass pieces with water content ranging from 0.5 to 5.5 wt.% were obtained. Quench crystals were present in some of the synthesized glasses, but this is not a problem for the diffusion experiments since such crystals rapidly dissolve during heating to the target temperature. For the IR calibration a set of smaller samples (ca. 100 mg) was synthesized in an IHPV equipped with a rapid quench device (Berndt et al., 2002). Fast cooling prevented formation of quench crystals in this case.

2.2. Experimental techniques

2.2.1. Diffusion couple experiments in IHPVs

Diffusion couples consisted of two glass cylinders (3 mm length, 2.8 mm diameter) drilled out from the prepared dry and hydrous glasses and polished on one of the base planes. The two cylinders, having different water contents, were placed together with the polished surface in direct contact and loaded in Pt capsules. Each sample assemblage was prepared in a way to minimize air and free space in the capsule.

Water diffusion experiments in an IHPV at Leibniz University of Hannover were performed at 2 or 3 kbar pressure and temperatures from 1373 to 1677 K with run durations between 108 and 1188 s. For most of the IHPV experiments a new rapid heating–rapid quench device (RHQ) was used, as described below. For comparison, one experiment was run with a standard sample holder (NQ) heating the sample at a rate of 30 K/min and initial cooling at a rate of 100 K/min.

Rapid quench devices for IHPVs were developed and described in detail by Roux and Lefèvre (1992), Holloway et al. (1992) and Berndt et al. (2002). They were designed to prevent melts from crystallization during quench or any other quench effect. The new RHQ device used in this study (Fig. 1) was aimed to minimize the durations of both heating and cooling in the experiments. Thus, it is particularly useful for studying short term processes such as diffusion of fast species or viscous flow in low-viscosity melts. The sample is fixed with a ceramic glue (Cerambond®) on a ceramic rod that has an iron wedge in the basal part. The ceramic rod is free to move in a ceramic tube between a storage position in the cold part of the oven and the

Table 1
Composition of phonolitic and trachytic melts measured by electron microprobe.

Sample	Phonolite		Trachyte	
	wt.%	(n)	wt.%	(n)
SiO ₂	58.89	(42)	60.53	(39)
TiO ₂	0.76	(2)	0.48	(4)
Al ₂ O ₃	19.87	(19)	17.83	(24)
FeO ^a	3.61	(35)	7.14	(43)
CaO	3.90	(15)	1.72	(14)
MgO	0.69	(4)	0.21	(4)
Na ₂ O	5.96	(44)	5.22	(24)
K ₂ O	6.87	(25)	7.28	(19)
Total	100.55		100.45	
Na/K	1.32		1.09	
NBO/T	0.06		0.01	

Notes. Microprobe analyses are based on 30 measurements on three fragments of each glass. One standard deviation is given in parentheses.

Na/K = atomic ratio of alkali elements.

NBO/T = (Na⁺ + K⁺ + 2 Ca²⁺ + 2 Mg²⁺ + 2 Fe²⁺ - Al³⁺ - Fe³⁺) / (Si⁴⁺ + Ti⁴⁺ + Al³⁺ + Fe³⁺).

^a All iron is given as FeO.

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