

## H<sub>2</sub>O diffusion in silicate glasses and melts

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Diffusion of H<sub>2</sub>O in silicate melts and glasses plays a critical role in bubble growth that powers explosive volcanic eruptions and in stability of glass. Although extensive work has been carried out, there are still some major gaps in our understanding of the diffusion process. This work is aimed at understanding how the diffusivity of H<sub>2</sub>O depends on the anhydrous composition, the pressure, and the dissolved total H<sub>2</sub>O concentration (H<sub>2</sub>O<sub>t</sub>).

Samples used in this study include KS and Rhy (natural obsidian glass from Mono Craters, California, USA; initial H<sub>2</sub>O<sub>t</sub> 0.8% by weight for KS and 1.9% for Rhy), NSL (natural peralkaline rhyolitic glass from New Zealand, with 2.0% H<sub>2</sub>O<sub>t</sub>), Ab (synthetic albite glass with 1.8% initial H<sub>2</sub>O<sub>t</sub>), AOQ (synthetic haplogranitic glass with nominal composition of Qz28Ab38Or34 containing either 1.3% H<sub>2</sub>O<sub>t</sub> or 2.2%), T3 (synthetic glass with nominal composition of Qz50An50 containing 2.0% H<sub>2</sub>O<sub>t</sub>). Table 1 lists the composition of the samples measured by electron microprobe.

Dehydration experiments were carried out at 584–605°C and 0.5 to 5 kbar Ar pressure in a rapid-quench cold-seal furnace. After heating of 16 to 73 hours, each quenched experimental wafer is sectioned to obtain a slice from which the H<sub>2</sub>O diffusion profile is measured by infrared spectroscopy. The Bruker IFS88 IR spectrometer with microscope (A-590) setup at Hannover is used with a slit width of 5 to 25 µm. The H<sub>2</sub>O<sub>t</sub> concentration is obtained by summing up the OH and molecular H<sub>2</sub>O

(H<sub>2</sub>O<sub>m</sub>) species concentrations measured at 4520 cm<sup>-1</sup> and 5230 cm<sup>-1</sup> combination bands.

All H<sub>2</sub>O<sub>t</sub> concentration profiles are well fit by assuming that H<sub>2</sub>O<sub>t</sub> diffusivity ( $D_{H_2O_t}$ ) is proportional to H<sub>2</sub>O<sub>t</sub> (Fig. 1). They can also be fit well by assuming that molecular H<sub>2</sub>O is the diffusing species and  $D_{H_2O_m}$  is independent of H<sub>2</sub>O<sub>t</sub>. These results are consistent with both Zhang *et al.* (1991) and Nowak and Behrens (1997) since H<sub>2</sub>O<sub>t</sub> is relatively low. The effect of different fitting procedures to the resulting  $D_{H_2O_t}$  is examined and found to be within 20% relative.

Experimental data show that  $D_{H_2O_t}$  decreases with increasing pressure. For AOQ composition at 600 °C and 1% H<sub>2</sub>O<sub>t</sub>,  $D_{H_2O_t}$  decreases from 0.25 µm<sup>2</sup>/s at 0.5 kbar, to 0.133 µm<sup>2</sup>/s at 2 kbar, and to 0.080 µm<sup>2</sup>/s at 5 kbar. The activation volume for H<sub>2</sub>O diffusion inferred from the data is 17 ± 8 (2σ error) cm<sup>3</sup>/mol, somewhat greater than but still in agreement within experimental error with that inferred from high temperature diffusivity data for AOQ (5 to 11 cm<sup>3</sup>/mol, Nowak and Behrens, 1997).

The dependence of  $D_{H_2O_t}$  on anhydrous composition is examined at 5 kbar Ar pressure and 600°C.  $D_{H_2O_t}$  at 1 wt.% H<sub>2</sub>O<sub>t</sub> is 0.152, 0.080, 0.070, and 0.053 µm<sup>2</sup>/s for NSL, AOQ, Ab and KS/Rhy samples. For the T3 sample, the diffusion profile is too short to be measured, implying a maximum  $D_{H_2O_t}$  at 1% H<sub>2</sub>O<sub>t</sub> of ~0.002 µm<sup>2</sup>/s, much less than that in other glasses. For comparison,  $D_{H_2O_t}$  in SiO<sub>2</sub> glass (Moulson and Roberts, 1961) at 600°C and 1 bar is 0.044 µm<sup>2</sup>/s by a

TABLE 1. Anhydrous oxide concentrations (wt.%) of samples used

Sample	SiO <sub>2</sub>	TiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	FeO	MnO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	Sum
KS, Rhy	76.59	0.08	12.67	1.00	—	0.03	0.52	3.98	4.88	
NSL	75.45	0.19	10.05	4.29	0.12	0.00	0.17	5.27	4.56	100.10
Albite	69.19	0.00	18.83	0.00	0.00	0.00	0.00	11.96	0.02	100.00 <sup>†</sup>
AOQ	76.14	0.00	13.53	0.00	0.00	0.00	0.00	4.65	5.68	100.00 <sup>†</sup>
T3	71.64	0.00	18.48	0.00	0.00	0.00	10.13	0.00	0.00	100.00 <sup>†</sup>

<sup>†</sup> Normalized to 100%.

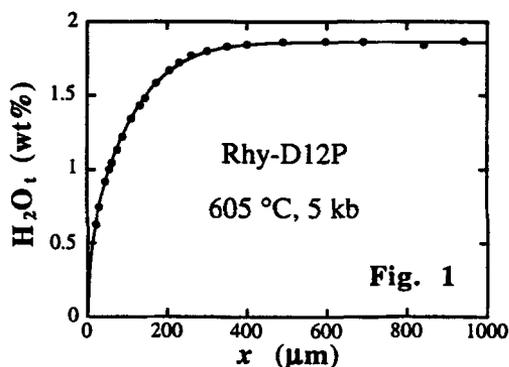


FIG. 1.

gross extrapolation to 1%  $H_2O_t$ , much greater than that in T3 glass even with the pressure effect being considered. Further,  $D_{H_2O_t}$  in basaltic melts is significantly greater (Zhang and Stolper, 1991). We therefore conclude that (i)  $D_{H_2O_t}$  values in fully polymerized and Ca-poor glasses and melts (AOQ, Ab and KS/Rhy) are not strongly dependent on the anhydrous composition, in agreement with Behrens and Nowak (1997); (ii)  $D_{H_2O_t}$  increases with extra alkalis unassociated with Al (NSL) or with a decrease in degree of polymerization (basalt); (iii) assuming  $H_2O_m$  is the diffusing species, the greater  $D_{H_2O_t}$  value in NSL is partially attributable to a greater  $H_2O_m$  concentration at a given  $H_2O_t$  compared with other samples, but also owing to a greater  $D_{H_2O_t}$  and (iv) high Ca concentration in polymerized  $SiO_2$ -rich melt reduces  $D_{H_2O_t}$  (T3). The reduction of  $D_{H_2O_t}$  by Ca may be due to complexing reactions between OH and Ca.

Both Ar solubility and diffusivity are obtained as a side product from this study since Ar is used as the pressure medium. The solubility of Ar in the glasses at 600°C and 5 kb is 0.52 wt.% in AOQ, 0.49% in KS/Rhy, 0.37% in NSL and 0.25% in Ab. The differences in the solubilities are small and consistent with other studies (e.g. White *et al.*, 1989). Examination of Ar diffusivity as a function of  $H_2O_t$  can reveal how the diffusivity of a molecular species depends on  $H_2O_t$ . This knowledge may be instructive to the understanding of  $H_2O_m$  diffusion. However, because Ar diffusion profile is relatively short (20 to 80  $\mu m$ ), loss of several  $\mu m$  glass from the edge during preparation and polishing of the sample has a significant effect on the measured profile. Hence, the effect of  $H_2O_t$  on Ar diffusion is not conclusive yet.

Work is in progress to investigate the effect of  $H_2O_t$  on  $D_{H_2O_t}$  at higher  $H_2O_t$  content, and to study diffusion in dacitic and andesitic glasses and melts.

## References

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