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Diffusion of Ar and CO₂ in Na aluminosilicate melts

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Next to H_2O , carbon dioxide is the second most abundant volatile in magmatic systems and has a strong effect on degassing processes of magmas. Even small amounts of dissolved CO_2 shift the fluid saturation limit of hydrous magmas to higher pressures and thus greater depth. Therefore it is discussed to be the driving force of bubble formation and growth in ascending magmas. Carbon dioxide emissions from active volcanoes are also a major source of greenhouse gases in the atmosphere over geological time. In contrast to H_2O diffusion there is still little information about the mobility of CO_2 in natural melt compositions.

Our previous work on the diffusion of carbon dioxide in natural simplified silicate melts from rhyolite to hawaiite demonstrated that the diffusivity of bulk CO_2 is independent from the melt composition (Nowak et al. 2004). In contrast to this, diffusion experiments in albitic glasses, that were depolymerized by the addition of excess Na₂O (NaAlSi₃O₈ + n Na₂O; n = 0 - 6 wt%) have shown an exponential increase of bulk CO₂ mobility with increasing degree of depolymerisation (Sierralta 2002). To shed light on this contradiction, we performed one dimensional CO₂ and argon diffusion couple experiments in synthesized iron free Na aluminosilicate melts as used by Sierralta (2002).

For each composition Ar-bearing and Ar-free glass cylinders (\emptyset 3 mm) were synthezised in an internally heated argon pressure vessel (IHPV) and then cut into slabs 4 mm long. According to the diffusion couple technique (Nowak and Behrens 1997) Pt jacketed Ar-bearing and Ar-free glass slabs were brought into contact and welded shut in a Pt capsule. The runs were performed in an IHPV at 500 MPa, 1423 to 1773 K and run times ranging from 1800 to 7800 s, assuring semi-infinite medium configuration. Concentration-distance profiles were monitored using electron beam microprobe, and error functions were fitted to the symmetrical profiles to derive diffusion coefficients

of bulk CO_2 . The results were compared with the corresponding CO_2 diffusion experiments performed by Sierralta (2002).

The results of the CO₂ diffusion experiments show an exponential increase of bulk CO₂ mobility with increasing degree of depolymerisation for all investigated temperatures in the compositional range studied, e.g. from log $D = -11.6 \pm 0.1$ (D in m²/s) to -11.2 ± 0.1 at 1423 K. The Ar diffusion data also increases exponentially, but with a significantly steeper slope, e.g. from log $D = -11.6 \pm 0.1$ to -10.6 ± 0.1 at 1423 K. As shown by Nowak et al. (2004) the diffusion of the inert volatile component argon can be used as a proxy for the diffusion of molecular CO₂. This indicates that the mobility of molecular CO₂ is increasing faster with increasing degree of depolymerisation than the mobility of bulk CO₂.

The new data also show that molecular CO_2 is a stable species in more depolymerized melt compositions, although IR spectra of the glasses at ambient conditions show a shifting of the CO_2 speciation from molecular CO_2 to CO_3^{2-} with increasing degree of depolymerisation. In AB + n Na₂O (n ≥ 4 wt%) CO₂ is incorporated mainly as CO_3^{2-} . To gain further knowledge on CO_2 speciation and diffusion mechanisms in aluminosilicate melts, we will model the CO_2 diffusion and speciation using the viscosity model by Giordano & Dingwell (2003) according to Nowak et al. (2004).

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