

Correlation between sulphur diffusion and sulphur speciation in silicate melts

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Degassing of silicate melts is an important process in nature (e.g., volcanic eruptions) as well as in industrial glass production (e.g., fining of glass melts). One of the most abundant volatile components in volcanic gases is sulphur (mostly as SO₂ and H₂S) which has a large climatic impact due to the formation of aerosols in the stratosphere. Diffusion of sulphur is a rate-controlling step during melt degassing. Sulphur can be present in the melt in different oxidation states (from 2- to 6+) and the diffusion of sulphur is expected to be strongly dependent on sulphur speciation. The main objective of our research project is to investigate the coupling between sulphur diffusion and speciation in simple silicate melts. Sodium trisilicate (NS3) and soda-lime-silica (NCS) glass melts were doped with 1000 to 2000 ppm S by weight using mixtures of glass powder and Na₂SO₄, Na₂SO₃, Na₂S, FeS₂ or native sulphur powder. These mixtures were either sealed in Au capsules and processed in an internally heated argon pressure vessel (IHPV) at 1000°C, 100 MPa, or melted at one atmosphere in Pt or corundum crucibles. Diffusion experiments were carried out using the diffusion couple technique with one half of the couple being a sulphur-doped glass cylinder. Diffusion experiments were processed at different temperatures and 100 MPa total argon pressure in an IHPV.

Sulphur speciation in the diffusion samples was analysed at ANKA (Forschungszentrum Karlsruhe) using XANES (X-ray Absorption Near Edge Structure) spectroscopy, indicating that sulphate is the dominating sulphur species at oxidising conditions in the IHPV (pH₂ ~ 0.2 bar) [1]. Diffusion profiles were measured using an electron microprobe. Our results indicate that sulphur diffusion is closely related to the melt viscosity in NS3 melts at 1000°C (diffusion coefficient of $\log D = -13.37 \pm 0.12$, D in m²/s). With increasing temperature sulphur diffusion increases to $\log D = -11.74 \pm 0.10$ at 1250°C. First results on NCS melts show that the sulphur diffusion is about one order of magnitude slower than the diffusion of network formers calculated by the Eyring equation, i.e. we found $\log D = -13.81 \pm 0.10$ at 1100°C and $\log D = -13.22 \pm 0.14$ at 1200°C). Experiments at higher pressures (0.75 to 2.0 GPa) in a PCA at 1100°C showed no measurable pressure dependence of sulphur diffusion. Further experiments of our work will focus on the effect of *f*O₂ and melt water content on sulphur diffusion and other simple silicate systems.

References

[1] L. Backnäs, J. Stelling, H. Behrens, J. Göttlicher, S. Mangold, O. Verheijen, R. G. C. Beerkens & J. Deubener (2008): Journal of the American Ceramic Society 91, 721-727.

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