

## Modeling of dehydrogenation of olivine during magma ascent

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Despite the recognised crucial role of H in affecting so many processes and rates of the deep Earth, the actual amount of H contained in Nominally Anhydrous Minerals (NAMs) in the mantle remains unclear. The H content in olivine from mantle xenoliths or megacrysts transported by magmas in different tectonic settings is highly variable from about <1 ppm to ca. 400 ppm H<sub>2</sub>O (e.g., Beran and Libowitzky, 2006). This could reflect variable local water contents in the mantle, different transport histories of the olivines and host magma, or a combination of the two. The possibility of H loss from the olivines during magma transport is supported by the experimentally determined H diffusion rates in olivine (e.g., Mackwell and Kohlstedt, 1990) and by the H gradients in natural crystals (e.g., Demouchy *et al.*, 2006). However, much uncertainty remains as to the significance of both, the maximum water contents, and the H profiles, since the effect of the ascent paths and conditions have not been considered, e.g. water saturated vs. undersaturated or multi-stage histories.

Here we present results of a dehydrogenation diffusion model of olivine that clarifies under which conditions of transport times, crystal sizes, and magma decompression paths the H content measured from naturally occurring olivine may reflect the original water content of the upper mantle. We find that even for a grain size of 2 cm, magma ascent rates of > 10 m/s are required to preserve at least most (e.g., 90 %) of the initial water content of an olivine originated from the mantle. The simulations also show that (i) homogeneous H distribution within naturally occurring NAMs may indicate extensive diffusive loss of H rather than being representative of upper mantle H concentrations as is commonly assumed, (ii) a short stage of a few days in an intermediate magma reservoir prior to eruption completely re-equilibrates the water content with the new low pressure environment, even for crystals several cm in size. The fast rates of ascent required to preserve the highest water contents suggest that the H contents of natural crystals reflect the transport path of their magma rather than their mantle sources.

### References

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## Thermodynamic model for the simultaneous exsolution of two fluids from silicic magmas

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Chlorine is the predominant constituent in magmatic and hydrothermal fluids. From intrusive to shallow epithermal settings the salinity and composition of hydrothermal fluids changes dramatically and this is partly due to the subcritical phase separation in the H<sub>2</sub>O-NaCl system. It is still unclear (1) what is the salinity of exsolving magmatic fluids, (2) whether one or two fluid phases simultaneously separate from the magmas, and (3) how analytical data from melt inclusions can be linked to fluid chemistry or be used to monitor the devolatilization paths. We propose a new thermodynamic model for chlorine in silica-rich melts that is based on Darken's formalism for solute in a multicomponent solvent. Thermodynamic properties of NaCl are related to the molten-salt standard state *via* the increment,  $I_{\text{NaCl}}$  (kJ) = 11 + 0.03 T(K) - 1.33 p(kbar), which was calibrated by experimental data on the chlorine solubility in granitic melts. Interaction between NaCl and H<sub>2</sub>O in the melt structure is defined by the regular solution parameter,  $W_{\text{NaCl-H}_2\text{O}}$  (kJ) = -53 + 0.04 T(K) + 2.67 p(kbar). Properties of the H<sub>2</sub>O-NaCl-KCl fluids are described by the Kosinski-Anderko-Pitzer equation of state. Phase equilibrium computations in the granite-H<sub>2</sub>O-Cl<sub>2</sub>O<sub>1</sub> space reveal existence of a temperature minimum at 5.4 wt. % H<sub>2</sub>O and 0.13 wt. % Cl in the melt (at 200 MPa) where very small changes in the melt Cl/H<sub>2</sub>O ratio produce large variations in the fluid salinity. At subcritical conditions, the thermal minimum becomes an invariant point (Cl/H<sub>2</sub>O = 0.037 at 100 MPa) where all differentiation paths of chlorine-bearing granitic magmas terminate by simultaneous saturation with the vapor and brine. Here the Cl and H<sub>2</sub>O concentrations in the melt remain invariantly buffered despite the fact that the largest amount of fluids is produced at this stage. The thermodynamic model demonstrates that: (1) experimental data require only minor deviations from mixing of halide and aqueous components in the melt, i.e., the trends of chlorine and water solubilities are largely related to non-ideal activity-composition relationships in the aqueous fluid, (2) natural silicic magmas with Cl/H<sub>2</sub>O < 0.04 first saturate with a low-density aqueous vapor and subsequently exsolve concentrated brines at the solidus; decompression favors vapor saturation whereas crystallization promotes brine separation, and (3) realistic predictions of metal to hydrothermal fluids must consider both the melt-brine and melt-vapor partitioning. It remains to be understood whether the "vapor-then-brine" mechanism of fluid saturation in the shallow-level magma bodies promotes the vapor-mediated metal transport or it leads to metal enrichment in the melt up to high degrees of fractionation, followed by partitioning to late-stage brines. The new thermodynamic model offers the missing link between the melt and fluid chemistry in crustal magmatic reservoirs and it can be used in quantitative modeling and interpretation of devolatilization and ore-forming processes.