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The Fidelity of Olivine-Hosted Melt Inclusions as Recorders of Pre-Eruptive Water Content and Oxygen Fugacity

Glenn Gaetani, Julie O'Leary, and Nobumichi Shimizu

Department of Geology and Geophysics, Woods Hole Oceanographic Institution, Woods Hole MA, USA

Olivine-hosted melt inclusions represent an important source of information on both the pre-eruptive H_2O contents and oxygen fugacities of basaltic magmas [1]. The principal uncertainty involved with deriving pre-eruptive H_2O concentrations from melt inclusions is the potential for diffusive loss or gain of H^+ (protons) through the host olivine. Further, it has been proposed that the proton flux associated with H_2O loss/gain affects the oxidation state of the inclusion [2,3]. Results from hydration and dehydration experiments carried out on natural inclusion-bearing olivines analyzed by SIMS and XANES confirm that H_2O re-equilibratrion occurs rapidly via proton diffusion through the host olivine, and demonstrate that re-equilibration of oxygen fugacity within the inclusion occurs on comparable timescales via diffusion of point defects. Therefore, an olivine-hosted melt inclusion provides a reliable record of both the H_2O content and oxygen fugacity of the external melt with which it most recently equilibrated. However, efficient re-equilibration of both H_2O and oxygen fugacity limits the utility of olivine-hosted melt inclusions as indicators of mantle processes.

Hydration experiments were performed on olivines from Puu Wahi, a scoria cone on the NE rift zone of Mauna Loa volcano. Melt inclusions initially containing 0.36±0.05 wt% H_2O were held at 1 GPa and 1250°C in water enriched in ^{18}O ($^{18}O/\Sigma O = 0.977$) and D ($^{2}H/\Sigma H = 0.998$) to map the transport of protons and oxygen during equilibration of melt inclusions with an external fluid. Dehydration experiments were carried out for 1 to 18 hrs at 1 bar and 1250 °C on inclusion-bearing olivines in scoria erupted from Cerro Negro volcano, Nicaragua, in 1999. The initial concentration of H_2O in these melt inclusions was uniformly high (3.6±0.6 wt%). All run products were analyzed for major elements by electron microprobe and for H_2O by SIMS on the Cameca 1280 ion microprobe at WHOI. The oxidation state of Fe was determined by XANES at beamline 13-IDC of the Advanced Photon Source at Argonne National Laboratory.

The D/H ratios of the melt inclusions from our hydration experiments range from 18.4-25.6, as compared to $\sim 1.448 \times 10^{-4}$ for mantle-derived basalt, indicating significant addition of deuterium. The $^{18}O/^{17}O$ ratios of melt inclusions are within uncertainty of natural ratios for mantle-derived materials. The H_2O content of individual melt inclusions increased by as much as 3.9 wt %, while the oxidation state of Fe in the hydrated melt inclusions is not significantly different from the starting materials.

The concentration of H_2O in melt inclusions from the dehydration experiments ranges from 2.8 to 0.05 wt%, and dehydration is nearly complete after 18 hours. The δD value of the melt inclusions increases significantly as dehydration progresses and is consistent with a calculated diffusive fractionation of hydrogen isotopes. Neither diffusive fractionation of H_2O nor equilibrium fractionation via vapor loss is consistent with the isotopic enrichment observed in dehydrated melt inclusions. The oxidation state of Fe ranges from $Fe^{3+}/\Sigma Fe = 0.58 \pm 0.04$ (NiNiO+4) for the starting materials to $Fe^{3+}/\Sigma Fe = 0.21 \pm 0.03$ (NiNiO+0.6) for melt inclusions that were heated for 18 hours, and re-equilibrates on the same timescale as H_2O.

References: [1] K.A. Kelley, E. Cottrell, Science 325, 605 (2009); [2] A. V. Sobolev, L. V. Danyushevsky, *J Petrol* **35**, 1183 (1994); [3] L. V. Danyushevsky, A. W. McNeill, A. V. Sobolev, *Chem Geol* **183**, 5 (2002).