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FLUIDS IN THE CRUST

In-situ measurements of fluorine and chlorine speciation and partitioning between melts and aqueous fluids in the Na_2O-Al_2O_3-SiO_2-H_2O system†

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Abstract

The effect of pressure and temperature on the structure of silicate melts coexisting with silica-saturated aqueous electrolyte fluids enriched in fluorine or chlorine in the Na_2O-Al_2O_3-SiO_2-H_2O system has been described. In situ measurements were conducted with the samples at desired temperatures and pressures in a hydrothermal diamond-anvil cell (HDAC) by using microRaman and FTIR spectroscopy techniques. The data were acquired at temperatures and pressures up to 800 °C and 1264 MPa, respectively.

In silicate melts, the intensity of the infrared bands assigned to the stretch vibration of OH-groups is smaller than those of coexisting molecular H_2O when F and Cl are present in the melt structure. This difference reflects the interaction of F or Cl with H_2O in the melts. With decreasing pressure and temperature (*P*-*T*) conditions, SiF complexes are favored in the melt over that in coexisting fluid, perhaps because of decreasing silicate concentration in fluids with decreasing temperature and pressure. In these melts, the solubility of Cl, likely in the form of NaCl, increases with decreasing *P*-*T* conditions, whereas the abundance of such complexes in coexisting fluids decreases in favor of HCl.

Our experimental data were employed to model the ascent of a fluid-saturated magma from the upper mantle to the shallow crust. This modeling offers insights into F and Cl partitioning between and the speciation of F and Cl in melts and magmatic fluids. We suggest that the formation of stable SiF and NaCl complexes and their increasing solubilities in silicate melts during magma ascent may explain the late volcanic degassing of F and Cl compared with the degassing behavior of other volatile species.

Keywords: Fluorine, chlorine, speciation, aqueous fluids, hydrous melts, structure, spectroscopy, hydrothermal diamond-anvil cell