

The effect of water on fluorine and chlorine solubility and solution mechanisms in aluminosilicate melts at high pressure and high temperature

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The influence of water on F and Cl solubility and structural complexing in silicate melts is important to understand the difference in fractionation of F and Cl during hydrous mantle melting. This is illustrated in a recent study of Mt Shasta melt inclusions (Le Voyer et al. 2010) shows two distinct Cl/F ratios. These two ratios can be modeled by the melting of a peridotite that had been metasomatized by low or high amount of aqueous fluid, using F and Cl partition coefficients between a peridotite and hydrous basaltic melts (Dalou et al, in review).

Here, we address, therefore, the effect of water on the solution behavior of F and Cl by combining solubility measurements of F and Cl, and Raman data of F or Cl-bearing peralkaline aluminosilicate glasses (quenched melt). Six compositions along the join $\text{Na}_2\text{Si}_3\text{O}_7\text{-Na}_2(\text{NaAl})_3\text{O}_7$ were used, keeping a constant NBO/T and varying Al/(Al+Si). Starting materials were doped with 5 wt% Cl in the form of PdCl_2 or 10 wt% F in the form of AgF_2 , which release gaseous F_2 or Cl_2 , respectively, during an experiment. Starting material was loaded together with 2 to 10 wt% of water, in order to maintain water undersaturation content (Mysen and Cody, 2004). Samples were synthesized at 1400 °C and 1.5 GPa.

The F solubility increases from 1.4 to 5.2 wt% (4.5 to 7.8 mol%) with increasing water from 2 to 10 wt% , in the Al-bearing melt system (5 mol% Al_2O_3).

The Cl solubility decreases from 3.4 to 2.1 wt% (5.7 to 3.4 mol%)with water increasing from 2 to 10 wt% , in the Al-free melt system. Increasing Al content (5 to 10 mol%) reduces the effect of water on Cl solubility.

In F-bearing glasses (quenched melt), a peak around 870-910 cm^{-1} in their Raman spectra is assigned to Si-F bonding. This peak is absent in spectra of anhydrous F-bearing glasses, but integrated area, and, therefore, concentration of Si-F bonds, increases with increasing water. In anhydrous Al-free glasses, solution of F is in the form of NaF-type complexes, whereas in hydrous silicate glasses water may exist as NaOH-bearing and SiF-bearing complexes. When Al is added to the hydrous system, AlF-bearing complexes appear, in addition to the SiF-bearing complexes. This suggests, therefore, that the OH-complexing governs F solubility mechanism in silicate melts. In Cl-bearing glasses on the other hand, there is no indication in the Raman spectra of changes of Cl speciation as water is dissolved in aluminosilicate melt.

Here we show that F and Cl solution behaviors depend strongly on the amount of water dissolved in the melt, which has profound effect in melt structure. These data are consistent with changes of F and Cl partition coefficients with water during hydrous melting and explain why Cl/F ratios in arc magmas can be produced by variable amounts of water involved in magma genesis.