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AMORPHOUS MATERIALS: PROPERTIES, STRUCTURE, AND DURABILITY

The effect of H_2O on F and Cl solubility and solution mechanisms of in aluminosilicate melts at high pressure and high temperature†

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ABSTRACT

Effects of H_2O on the solution behavior of fluorine and chlorine in peralkaline sodium aluminosilicate glasses quenched from melts at high temperature (1400 °C) and pressure (1.5 GPa) were studied by combining solubility measurements and Raman spectroscopy. With increasing H_2O content from 0 to ~10 wt%, the fluorine solubility increases from 3.3 to 4.4 mol% in Al-free glasses and from 6.3 to 9.3 mol% in Al-rich glasses (10 mol% Al_2O_3). In contrast, in the same H_2O concentration range the chlorine solubility decreases from 5.7 to 3.4 mol% in Al-free glasses and from 3.6 to 1.7 mol% in Al-rich glasses.

In Al-free glasses, interaction between H_2O and the silicate to depolymerize the network is $Q^4 + H_2O \leftrightarrow Q^2(H)$ and $Q^3 + H_2O \leftrightarrow Q^2(H)$. The effect of water on silicate melt structure is different in halogen-bearing melts because in hydrous melt systems both F and Cl can act to depolymerize the melt further. For fluorine, this is accomplished via formation of Si-F, Al-F, and Na-F bonding in addition to Si-OH, whereas in chlorine-bearing hydrous melts, there is no interaction between Si^{4+} and Cl^-.

The halogen solubility in the magmatic liquid influences mineral/melt partition coefficients of chlorine and fluorine and implies partition coefficients different from unity. Moreover because of the contrasting effects of H_2O on fluorine and chlorine solubility, the Cl/F ratio in magmas formed in water-rich environments such as subduction zones can be a sensitive indicator of H_2O content during arc magmas genesis. Transport properties of melts, such as diffusion and viscosity, also vary differently in halogen-bearing hydrous melts compared with halogen-free systems. Moreover, those effects on melt properties are the strongest in F-bearing systems.

Keywords: Water, fluorine, chlorine, solubility, solution mechanism, aluminosilicate melt