

Solubility and solution mechanism of H₂O in alkali silicate melts at high pressure

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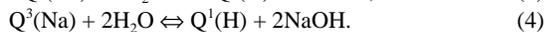
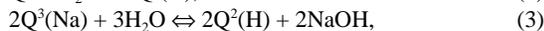
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The solubility behavior of H₂O in melts in the system Na₂O-SiO₂-H₂O were determined by Raman and NMR methods in the 0.8-2 GPa and 1000^o-1300^oC pressure and temperature range. The NBO/Si-range of the melts (0-1) was chosen to cover that of most natural magmatic liquids.

The H₂O solubility (X_w) ranges between 18 and 45 mol% (O=1) with (∂X_w/∂P)_T~14-18 mol% H₂O/GPa. The (∂X_w/∂P)_T is negatively correlated with NBO/Si (=Na/Si) of the melt. The (∂X_w/∂T)_P is in the -0.03 - +0.05 mol% H₂O/^oC range, and is negatively correlated with NBO/Si. Melts with NBO/Si similar to basaltic liquids show (∂X_w/∂T)_P<0, whereas more polymerized melts exhibit (∂X_w/∂T)_P>0. The [∂X_w/∂(NBO/Si)]_{P,T} is in the -3 - -8 mol% H₂O/(NBO/Si) range. Complete miscibility between hydrous melt and aqueous fluid occurs in the 0.8-2 GPa pressure range for melts with NBO/Si≤0.5 at T>1100^oC. Miscibility occurs at lower pressure the more polymerized the melt. From the solubility data, the partial molar volume of H₂O in the melts, V_w^m, ranges between 10 and 13 cm³/mol with (∂V_w^m/∂T)_P ranging between 0 and -0.01/^oC.

Results from ¹H-²⁹Si CP MAS and ¹H NMR and Raman spectroscopy of quenched melts (glass) with ~20 mol% H₂O indicate the formation of Si-OH and Na-OH groups in addition to molecular H₂O. In quenched SiO₂-H₂O melts, protons are associated with Q³-species (ie, 1H⁺/Si). In less polymerized melts (Na/Si = 0.25-1), protons are associated with both Q³ and Q² structural units (1H⁺/Si and 2H⁺/Si). In the most depolymerized melts (Na/Si>0.5), protons were also associated with Q¹ structural units (3H⁺/Si). Evidence for Na-OH groups was observed in all Na-bearing melts.

Relations between abundance of Qⁿ-pecies (n=0, 1, 2, and 3) and H₂O content are consistent with formation of OH-groups via H₂O-solution mechanisms such as (in addition to molecular H₂O):



Reactions (1-3) dominate in felsic liquids (NBO/Si<0.5), whereas (3-4) is the predominant solution mechanism for less polymerized melts (NBO/Si>0.5). These solution mechanisms help explain the melt composition dependent H₂O solubility and partial molar volume of H₂O in these alkali silicate melts.