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In-situ characterization of carbon-speciation in silicate-C-O-H fluid and melt with temperature, pressure, and redox con

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Speciation and partitioning of C-bearing volatiles species in and between silicate-saturated C-O-H fluids and (C-O-H)-saturated melts have been determined in-situ with the samples to pressures and temperatures of ~2GPa and 900°C, respectively. Structural characterization was conducted with vibrational spectroscopy of samples contained in externally-heated, hydrothermal diamond anvil cells. The redox conditions were controlled near that of the $Fe+H_2O=FeO+H_2$ (reducing, RED) and $Ni+H_2O=NiO+H_2$ (oxidizing, OX) equilibria, respectively. Melts are, therefore saturated in H_2O, H_2, and C-bearing species (redox dependent) and coexisting fluids saturated in silicate components. Solubility of volatile and silicate components depend on both temperature and pressure.

The melt/fluid partition coefficients of the C-bearing species vary with redox conditions and temperature with the $\triangle H_{RED}{}^{melt/fluid}$ = 44(7) kJ/mol and $\triangle H_{OX}{}^{melt/fluid}$ = -70(32) kJ/mol. Pressure is a dependent variable and increases with increasing temperature. It is assumed no pressure effect of the partition coefficients.

The solution equilibria under reducing and oxidizing conditions, respectively, were; (1) $2CH_3{}^-+H_2O+Q^{n+1}=2CH_4+Q^n$ and (2) $2CO_3{}^{2-}+H_2O+2Q^{n+1}=HCO_3{}^-+2Q^n$, where the superscript, n, in the Q-species denotes number of bridging oxygen in the silicate species (Q-species). In the absence of H_2O equilibrium (1) changes to $CH_3{}^-+Q^n=CH_4+Q^{n+1}$. For oxidized carbon, there is an analogous expression expressing equilibrium between molecular CO_2 and structurally bound $CO_3{}^{2-}$-groups. Under both oxidizing and reducing conditions, the abundance ratios, $CH_4/CH_3{}^-$ and $HCO_3{}^-/CO_3{}^{2-}$ increase with temperature. The enthalpy change associated with the species transformation does, however, differ for fluids and melts and also for oxidized and reduced carbon ($\triangle H_{(1)}{}^{fluid}$ = -16(5) kJ/mol, $\triangle H_{(1)}{}^{melt}$ = -49(5) kJ/mol, $\triangle H_{(2)}{}^{fluid}$=81(14) kJ/mol). For the exchange equilibrium of CH_4 and CH_3 species, the temperature-dependent equilibrium constant yields $\triangle H$=34(3) kJ/mol.

Reactions (1) and (2) involve changes in silicate polymerization where increasing abundance ratios, $CH_4/CH_3{}^-$ and $CO_3{}^{2-}/HCO_3{}^-$ lead to increased silicate melt polymerization. As a result of the relations between speciation of C-bearing species and melt and fluid structure, stable isotope (C and H) and element partition coefficients between melts and fluids, which depend on and silicate polymerization and silicate speciation, also vary with speciation of C-bearing species in silicate-C-O-H systems. Pressure, temperature, and redox control on the C-speciation also govern those (and other) properties.

キーワード: COH volatiles, fluid structure, melt structure, high pressure, high temperature, redox conditions
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