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Volumetric Properties and Liquid-Vapor Phase Relations of CO$_{2}$-H$_{2}$O Fluids in Upper Crustal Contact-Metamorphic Aureoules

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Recent experimental measurements reveal the densities (ρ), excess molar volumes ($V^{\rm ex}$), relative buoyancies (RB), and liquid-vapor phase relations of CO$_{2}$-H$_{2}$O fluids at 300-700 °C, 5-200 MPa (RB = $\rho_{\rm H_2O}/\rho$, where $\rho_{\rm H_2O}$ is the density of pure water, and ρ is the density of the CO$_{2}$-H$_{2}$O mixture). At temperatures $>$ 500 °C, excess molar volumes for CO$_{2}$-H$_{2}$O fluids are relatively small (peak values ranging between approximately 1 and 20 cm$^{3}\cdot$mol^{-1}) and nearly symmetric about the midpoint composition (0.4 $\leq$ $X_{\rm CO_2}$ $\leq$ 0.6 at $V^{\rm ex}$(max)); consequently, volumetric properties are predicted with good accuracy by a largely empirical, modified Redlich-Kwong thermodynamic model (Kerrick and Jacobs,

1981). However, at {\it T} $\leq$ $500\deg$C, excess molar volumes (i) increase sharply with decreasing temperature and pressure (down to {\it P}$_{\rm sat}$, the saturation vapor pressure of pure water) and (ii) become strongly asymmetric toward H$_{2}$O; therefore, a more theoretically robust equation of state (e.g., the modified Lee-Kesler formulation developed by Duan et al. 1992a,b) is required to accurately represent volumetric properties and liquid-vapor phase relations. Reliable thermodynamic modeling is especially challenging for 300-$400\deg$C, {\it P} $\leq$ 50 MPa, as peak values for {\it V}$^{\rm ex}$ (at a given temperature) range from approximately 50 to 270 cm^{3}$\cdot$mol^{-1} for fluids that contain $<$ 30 mole % CO$_{2}$. Due to this extreme thermodynamic nonideality, concentrations of dissolved CO$_{2}$ as low as 3-5 mole percent will dramatically decrease fluid density. In addition, if the CO$_{2}$ content of the fluid is sufficiently high (between approximately 1 and 25 mole % CO$_{2}$, depending on the particular {\it P}-{\it T} conditions) at {\it T} $<$ $374\deg$C and {\it P}$_{\rm sat}$ $<$ {\it P} $<$ {\it P}$_{\rm c}$ ({\it P}$_{\rm c}$ = the critical pressure in the CO$_{2}$-H$_{2}$O system at 265 $<$ {\it T} $<$ $374\deg$C), a comparatively CO$_{2}$-rich vapor phase---with a density far below that of pure H$_{2}$O at the same {\it P} and {\it T}---will form. These results indicate that small amounts of CO$_{2}$ can profoundly affect the rates and patterns of hydrothermal fluid flow in shallow contact-metamorphic aureoules, and other upper crustal, magmatic-hydrothermal systems.$ $

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