

Testing the validity of density models for H₂O-NaCl-CO₂ fluids under hydrothermal conditions: new in situ experimental density data and phase relations

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Hydrothermal fluids of the H₂O-CO₂-NaCl system are important players in transcrustal magma systems. Whether derived directly from magma outgassing or of meteorite origin, these fluids are subject to significant variations in pressure (P) and temperature (T), which modify their volumetric properties and thus affect their circulation in magmas and host rocks, and their ability to transport other elements, including metals, between the different reservoirs (magma, host rock, atmosphere, hydrosphere).

To date, the experimental data available to model the evolution of the density of CO₂- and NaCl-bearing fluids as a function of pressure and temperature are limited to surface conditions and virtually non-existent at temperatures above 500 K [1,2]. As a result, the thermodynamic models available for the ternary H₂O-CO₂-NaCl system are only applicable to CO₂ storage conditions in “cold” saline aquifers [3] or rely on extensive extrapolations, either empirical or supported by molecular-dynamic simulations [4-6]. This limitation (or lack of validation) of thermodynamic models at T > 500 K hinders the development of models describing chemical exchanges associated with magmatic outgassing or high-temperature fluid circulation, for example linked to the formation of Cu-Au-Mo porphyry deposits in volcanic arcs.

Here we present new density data on the binary H₂O-NaCl and H₂O-CO₂ systems up to 900 K and 1.5 kbar to verify the validity of available density models. The measurements were conducted by the X-ray absorption method in a hydrothermal autoclave, which allows direct monitoring of liquid-vapor separation. In situ measured densities and fluid separation show a strong deviation from the most commonly employed equations of state [6,7], underlining the need to develop models supported by well-supplied experimental databases.

[1] Schmidt et al., 1995. *Geochim. Cosmochim. Acta* 59, 3953-3959. [2] Song et al. 2013. *J. Chem. Engin. Data* 58, 3342-3350. [3] Li et al., 2011. *Energia Procedia* 4, 3817-3824. [4] Duan, Z.H., and Zhang, Z.G. 2006. *Geochem. Cosmochim. Acta* 70, 2311-2324. [5] Mao, S. et al., 2015. *Appl. Geochem.* 54, 56-64. [6] Driesner, 2007. *Geochim. Cosmochim. Acta* 71, 4902-