Linking H_2 concentration and redox state (fH_2 and fO_2) in H_2O-H_2-NaCl fluids

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Hydrothermal processes such as serpentinization involve complex H_2-rich fluid mixtures whose thermodynamic properties are poorly known. One parameter that needs to be better constrained is the molal activity coefficient of H_2 dissolved in the fluid ($\gamma H_2(aq)$). Knowledge of this parameter is critical to link the H_2 molality ($mH_2(aq)$, commonly measured in hydrothermal fluids) and the H_2 fugacity ($fH_2(v)$, one redox parameter) by using:

$$fH_2(v) = \gamma H_2(aq).\ mH_2(aq).K$$

where K denotes the equilibrium constant for the reaction:

$$H_2(aq) = H_2(v) \qquad (Rn1)$$

It is commonly assumed that $\gamma H_2(aq) \equiv \gamma CO_2(aq)$ at the same T. However the few determinations of $\gamma H_2(aq)$ suggest a complex dependence with P, T and NaCl concentration in the fluid [1] and so systematic data are needed.

In this study we have developed an experimental setup allowing $\gamma H_2(aq)$ to be determined in H_2O-H_2-NaCl fluids at T = 250 to 450°C and P = 20 to 50MPa. The experiments are performed in large-volume Dickson-Seyfried bombs (1) fitted with an $Au_{20}Pd_{80}$ (wt.%) H_2 permeable membrane for *in situ* fH_2 monitoring and (2) allowing fluid sampling and $mH_2(aq)$ determination by gas chromatography in the fluid aliquot. Knowing the equilibrium constant of the reaction ($Rn1$) from SUPCRT92 [2] allows $\gamma H_2(aq)$ to be determined.

Results were first obtained for the H_2O-H_2 system at P = 50MPa. Determined $\gamma H_2(aq)$ are very close to 1 (1.11±0.08 at 250°C and 0.99±0.08 at 300°C), in agreement with the value expected for such fluids ($\gamma H_2(aq)$ = 1), and confirming the validity of our experimental setup. Experiments with NaCl-bearing fluids are now being conducted at T = 250 to 450°C and P = 20 to 50MPa. Results will be compared with the available data and applications for the calculation of fH_2 and fO_2 in hydrothermal fluids illustrated.

[1] Ding, K., and Seyfried, Jr., W.E. (1990). *EOS, Transactions American Geophysical Union*, 71:1680.

[2] Johnson, J.W., Oelkers, E.H., and Helgeson, H.C. (1992). *Computers & Geosciences*, 18(7): 899-947.