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Solute activity coefficients in high-temperature hydrothermal fluids revisited

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Accurate knowledge of solute thermodynamic properties is crucial for geochemical modelling of hydrothermal ore formation. However, two major problems basically prohibit geochemical modelling for important types of hydrothermal ore deposits such as porphyry-Cu, or the hotter parts of others such as VMS: (1) for the relevant temperature-pressure conditions, the HKF model for standard properties is outside its assumed range of validity and, (2) how to calculate reliability activities of dissolved species (e.g., ore metal complexes) has remained elusive. Regarding (2), the few existing relevant experimental determinations reach only to 350 °C and indicate very small bulk activity coefficients (i.e., for the complete solute irrespective of speciation) in the order of 10^{-2} at molal concentrations. It is plausible to expect that they may be even smaller at "supercritical" temperatures and moderate pressures given the low dielectric constant of water. Obviously, the common practices of either ignoring activity coefficients and/or relying on untested predictive values could lead to severe errors in modelling as was already pointed out decades ago [1,2]. I will review some recent experimental studies in our lab, showing that even for supposedly simple minor components such as KCl in NaCl-dominated solutions, current predictive models seem to be deficient, at least for the activity coefficient ratios. This applies for water densities of 0.5 g cm^{-3} and below but may be more generally the case. I will speculate about the possible origin of these problems, highlighting previous findings from the physical chemistry literature that have received little attention in the geochemical community [3].

[1] Brimhall G.H., Crerar D.A., 1987. Ore fluids: Magmatic to Supergene. Rev. Miner. Geoch.17, 235-321.

[2] Anderson, G.M., 2005. Thermodynamics of natural systems. Cambridge University Press.

[3] Levelt Sengers, J.M.H. et al., 1992. Fluid Phase Equilibria 81, 85-107.