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MODELLING CHEMICAL EQUILIBRIUM PARTITIONING WITH THE GEMS-PSI CODE

D. Kulik, U. Berner, E. Curti

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1   WHAT IS EQUILIBRIUM PARTITIONING?

The term Equilibrium Partitioning denotes the (equilibrium) distribution of a chemical element (M) between two phases of variable composition – usually a mineral solid solution and an aqueous solution. A measurable quantity called *partition coefficient* is defined as:

$$Kd = \frac{[\mathrm{M}]_S}{[\mathrm{M}]_{AQ}} \qquad (1)$$

Here, $[\mathrm{M}]_S$ is the concentration of M in the solid phase (S), and $[\mathrm{M}]_{AQ}$ is that in the aqueous solution (AQ). Except for very simple systems, Kd is usually a complex function of temperature T, solid/liquid ratio s/l, total element inventory M_{TOT}, aqueous composition (pH, Eh, ionic strength I, concentrations of complexing ligands $[L]$), and host-mineral composition. Measured Kd values may also be influenced by sorption or precipitation kinetics. Thus, Kd is a conditional constant, i.e. it applies only to specific experimental conditions, and cannot be generalised. All this makes the empirical Kd value not the perfect choice for long-term predictions of trace element distributions; hence, more fundamental theoretical approaches are necessary.

Calculations of aqueous speciation and saturation indices of pure solids, like MCO_3 and $MOOH$, can easily be performed using widespread computer models, such as PHREEQC [1], but they are not always helpful in understanding the relationships between Kd and system variables. Experimental Kd values for trace metals are often significantly different from theoretical partition coefficients obtained from solubility products of pure solids. This fact indicates that the aqueous concentration of metals is not always controlled by simple, pure solid equilibria, but rather by other retention mechanisms involving the host mineral phases — namely, sorption, re-crystallisation or co-precipitation. In all these cases, the concept of a fixed thermodynamic solubility fails because of *variable compositions of both* solid and aqueous

phases; dissolved $[\mathrm{M}]_{AQ}$ would no longer be the solubility in the classical sense of the word. Attempts to use variable solubility products depending on the composition of aqueous solution, or of a mixed solid, were disappointing, because the law of mass-action alone does not seem to be sufficient for solving the solid-solution aqueous-solution (SSAS) equilibria. Some supporting tools, like the Lippmann functions and diagrams [2,3], can help in binary systems, but not in higher-order systems [4], or if two or more solid solutions are involved.

Hence, for an adequate thermodynamic description of partitioning, it is necessary to go back to the more basic concept introduced by Gibbs, which states that the chemical potential of M is the same in all co-existing phases at equilibrium. The equilibrium state is determined by finding mole amounts of all chemical species in all phases such that the total Gibbs free energy of the system is minimal at the given state variables (temperature T, pressure P, bulk mole composition vector **b**). In this approach, a variable-composition phase is fully defined by stoichiometry and mole amounts of its end-members (components, species), which need not necessarily exist as pure substances. The stability of each end-member is given by its standard molar Gibbs free energy G^o of formation from chemical elements. A deviation from ideal mixing with other end-members (excess partial molar Gibbs energy, $\overline{G}_{real} - \overline{G}_{ideal} = \overline{G}^{Ex} = RT \ln \gamma$) is described by the activity coefficient γ, a function of the actual phase composition.

2   METHODS OF SPECIATION CALCULATIONS

Two numerical methods of chemical thermodynamic modelling can be applied to heterogeneous aquatic systems: (i) Law-of-Mass-Action — Reaction Stoichiometry (LMA), and (ii) direct Gibbs Energy Minimisation (GEM). The LMA approach is common, and available in many speciation codes, such as PHREEQC [1], MINEQL [5] or EQ3/6 [6], some equipped with data

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