

Thermodynamic databases for CO₂-water-rock systems

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Prediction of the fate of CO₂ and other gases injected in geological formations critically relies on the thermodynamic properties of the relevant solid, gaseous, and aqueous compounds. Compilations of such data are available for various geochemical modelling software, but parameters vary considerably among them. This study assesses the agreement of the geochemical predictions obtained with various available databases for the PHREEQC software, such as *phreeqc.dat*, *lnl.dat*, and *core10.dat*, based on comparisons to laboratory solubility measurements. The results revealed various degrees of systematic deviations of the predictions from laboratory data, with strong deviations at high temperature for *phreeqc.dat* and additionally for specific minerals like kaolinite and brucite. Furthermore, a lack of solubility measurements for many sulfide and silicate minerals prohibits the assessment of their stability.

To address some of these issues, a new thermodynamic database called *carbfix.dat* was created for PHREEQC [1]. Improvements include the revision of several mineral solubility expressions to improve consistency with recent experimental measurements, and the inclusion of new aqueous complex stability measurements as well as other aqueous species and solids relevant to subsurface CO₂ storage. To assess the variability in predictions of the fate of injected gases, exemplary geochemical models were run with different databases quantifying the importance of thermodynamic data for such simulations.

In addition to the above mentioned thermodynamic data, an internally consistent expression for the dissolution rate of basaltic glass, considered amongst the most favorable rocks for CO₂ storage, was re-calibrated based on existing rate measurements as a function of solution composition and temperature. This improves the capabilities of time-resolved models, as basalt dissolution often represents the rate-determining step in the carbonation of basalt-hosted systems.

[1] Voigt et al. (2018), Evaluation and refinement of thermodynamic databases for mineral carbonation, Energy Procedia 146, 81-91.