Novel Geochemical Calculation for Underground Geological Storage of CO₂: From Multiphase Geochemical Equilibrium to Compositional Reservoir Simulation

Fernando de A. Medeiros, Duncan Paterson, Erling H. Stenby, Wei Yan

*Center for Energy and Resources Engineering, Department of Chemistry, Technical University of Denmark, 2800 Kongens Lyngby, Denmark

Abstract

Underground geological storage (UGS) of CO₂ is one of the most promising technologies to realize large reduction of CO₂ emission in the near future. It entails the injection of CO₂-rich streams mostly in deep saline aquifers and depleted petroleum reservoirs. It is by nature a multiphase flow process involving multiple physics. To monitor or to assess the feasibility of such a process, it is crucial to have simulation tools that can accurately describe the involved physics. One distinctive feature in CO₂ storage simulation is multiphase equilibrium (phase equilibrium between all the fluid and solid phases) and geochemical reactions (speciation and mineral dissolution/precipitation), which underlies CO₂ partitioning, CO₂ transfer with fluids, and the ultimate fate of injected CO₂. Simulation tools for CO₂ storage analysis must rely on robust and fast chemical and phase equilibrium (CPE) calculation involving electrolytes, minerals, and fluid phases. Nevertheless, the highly nonlinear nature of the CPE problem has been a standing challenge to the robustness and efficiency of any CO₂ storage simulator. We have recently developed a novel formulation for multiphase reaction equilibrium and the relevant algorithms that can be used for geochemical calculation in CO₂ storage simulation. The formulation and the relevant algorithms, developed based on the RAND method, are a non-stoichiometric Gibbs energy minimization approach, in contrast to the methods using the law of mass action behind the well-known geochemical simulators PHREEQC or TOUGHREACT. Our RAND-based algorithms handle chemical and phase equilibrium simultaneously, solve a smaller set of equations, provide second-order convergence, and ensure convergence through Gibbs energy minimization. We illustrate here the application of the novel geochemical calculation method using three different examples of importance to CO₂ storage in saline aquifers. The examples require tools with varying complexity from standalone equilibrium calculation to three-dimensional compositional simulation, all with the RAND method at their cores:

- Fixed CO₂-fugacity calculation: The calculation is useful to understand the path of the geochemical reactions in saline aquifers, to screen mineral formations for their CO₂ storage potential, and to identify mineral phases that might occur during CO₂ injection. It is essentially an equilibrium calculation at fixed chemical potentials. We show how to apply RAND to an open system by extending Michelsen’s Q-function approach. The resulting algorithm is a minimization algorithm and more robust than the root-finding procedure employed in many geochemical simulators.

- 1D salt precipitation analysis: The RAND-based module was incorporated into a 1D multiphase compositional flow simulator that solves the mass and volume conservations implicitly. The resulting tool was used to analyze salt precipitation during CO₂ injection in a 1D aquifer. The large amount of CO₂ injected causes water vaporization and mineral precipitation. The precipitated minerals depend on the brine composition and the interplay between fluid flow and the reaction equilibrium. The tool is used to show how the mineral precipitation sequence varies with the injection condition, and how the precipitation amount and mineral composition vary spatially and evolve with time.

- 3D compositional simulation: A fully implicit 3D compositional simulator involving complete phase and geochemical equilibrium has been developed based on our geochemical module. The simulator calculates phase equilibrium and geochemical reaction equilibrium simultaneously. We provide a detailed procedure on how to couple the geochemical module in the solution of the governing equations using a fully implicit scheme. Results for CO₂ injection in a 3D saline aquifer are provided. The current simulation only includes two fluid phases (CO₂ and brine). Since there is no restriction in our RAND approach on the number of phases handled simultaneously, the extension to cases involving other fluid phases, like an oil phase in a depleted reservoir, should be readily achieved in the future.
This work presents a novel geochemical calculation method that can handle complex multiphase equilibrium and geochemical reaction equilibrium simultaneously. It provides a promising robust and efficient geochemical module for general-purpose CO₂ storage simulations in both aquifers and depleted petroleum reservoirs.

*Keywords*: Geochemical modeling; Multiphase reaction equilibrium; CO₂ storage; Salt precipitation; Simulation