



Theoretical study of the influence of chemical reactions and physical parameters on the convective dissolution of CO₂ in aqueous solutions

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Subsurface carbon sequestration has emerged as a promising solution to the problem of increasing atmospheric carbon dioxide (CO₂) levels. How does the efficiency of such a sequestration process depend on the physical and chemical characteristics of the storage site? This question is emblematic of the need to better understand the dynamics of CO₂ in subsurface formations, and in particular, the properties of the convective dissolution of CO₂ in the salt water of aquifers. This dissolution is known to improve the safety of the sequestration by reducing the risks of leaks of CO₂ to the atmosphere. Buoyancy-driven convection makes this dissolution faster by transporting dissolved CO₂ further away from the interface. Indeed, upon injection, the less dense CO₂ phase rises above the aqueous layer where it starts to dissolve. The dissolved CO₂ increases the density of the aqueous solution, thereby creating a layer of denser CO₂-rich solution above less dense solution. This unstable density gradient in the gravity field is at the origin of convection. In this framework, we theoretically investigate the effect of CO₂ pressure, salt concentration, temperature, and chemical reactions on the dissolution-driven convection of CO₂ in aqueous solutions. On the basis of a linear stability analysis, we assess the stability of the time-dependent density profiles developing when CO₂ dissolves in an aqueous layer below it. We predict that increasing CO₂ pressure destabilizes the system with regard to buoyancy-driven convection, because it increases the density gradient at the origin of the instability. By contrast, increasing salt concentration or temperature stabilizes the system via effects on CO₂ solubility, solutal expansion coefficient, diffusion coefficient and on the viscosity and density of the solution. We also show that a reaction of CO₂ with chemical species dissolved in the aqueous solution can either enhance or decrease the amplitude of the convective dissolution compared to the non reactive one. On the basis of a reaction-diffusion-convection model, we classify the various possible cases and show that the difference between the solutal expansion coefficients of the reactant and of the product governs the type of density profile building up in the aqueous solution and thus the stability of the system. By contrast to non reactive density profiles, reactive density profiles can feature a minimum that induces a delay of the buoyancy-driven convection. This work identifies the parameters that could influence the dissolution-driven convection in the aquifers, and thus impact the safety of the sequestration. In other words, this theoretical study shows that it is crucial to analyse the composition and reactivity of potential storage sites to choose those that will be most efficient for long-term CO₂ sequestration.