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Control of carbon dioxide convective dissolution with chemical reactions in porous media: Enhanced dissolution flux

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Dissolution-driven convection in partially miscible systems has regained much interest in the context of CO_2 sequestration [1]. A buoyantly unstable density stratification can build upon dissolution of CO_2 into brine, thereby driving convection. Dissolution and convection are known to improve the safety of the sequestration process by reducing the risks of leaks of CO_2 to the atmosphere. The temporal evolution of dissolution-driven convective dynamics has been characterized in detail and it has been shown that the dissolution flux reaches a steady-state value before shutdown [2]. The question remains, however, as to how the efficiency of such process depends on the chemical properties of the storage site and how chemical reactions can affect the steady-state fluxes. Reactions can indeed accelerate or slow down the development of dissolution-driven convection in partially miscible stratifications when they impact the density profile in the host fluid phase. The different possible dynamics taking place during the convective dissolution of CO_2 into a host fluid containing a dissolved reactant B have been classified for a chemical reaction $CO_2 + B \rightarrow C$ (product), assuming that all species diffuse at the same rate [3,4]. We now consider the more general case where solutes can diffuse at different rates. Using nonlinear simulations, we show that depending on the type of density profile developing in the host fluid, one or two convection zones can be observed. Although changing the diffusivity ratios can have non-trivial effects, decreasing the diffusion coefficient of product C or increasing the diffusion coefficient of reactant B both accelerate the development of convection and amplifies the global reaction rate in the host solution. Both effects contribute to a larger dissolution flux and a faster storage of CO_2 into the host fluid. We compare our results with experimental results showing that reactions accelerate the development of buoyancy-driven fingering during the convective dissolution of CO_2 into aqueous reactive solutions of alkali hydroxides [5].

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Primary authors: RONGY, Laurence (Universite libre de Bruxelles (ULB)); LOODTS, Vanessa (Universite libre de Bruxelles (ULB)); THOMAS, Carelle (Universite libre de Bruxelles (ULB)); SAGHOU, Hind (Universite libre de Bruxelles (ULB)); KNAEPEN, Bernard (Universite libre de Bruxelles (ULB)); DE WIT, Anne (Universite libre de Bruxelles (ULB))

Presenter: RONGY, Laurence (Universite libre de Bruxelles (ULB))

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