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Directionality of Thermal Diffusive Transport in Subsurface Carbon Dioxide and Hydrogen Storage

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Underground storage of hydrogen (H_2) fuel and carbon dioxide (CO_2) are technologies pursued to mitigate climate change and advance the energy transition. Once injected underground, CO_2 and H_2 will exist as partially-soluble multiphase fluid-water-rock systems: the injected fluids can flow through the porous rock in a connected plume, become disconnected and trapped as ganglia surrounded by groundwater within the storage rock pore space, and also dissolve and migrate through the aqueous phase. Recent analyses —many pioneered by the Benson research team [1,2,3] —have highlighted that the concentration gradients induced by differing capillary pressure between fluid ganglia can drive diffusive transport ("Ostwald ripening"). We build on these previous studies by incorporating the nonideality of gases under geologic conditions, with specific focus on the entropic and thermodiffusive effects resulting from geothermal gradients. Our analysis, based on thermodynamic first principles, provides numerical estimates for CO_2 and H_2 at conditions relevant to underground storage reservoirs. We show that the direction of diffusive transport in isothermal systems is upwards (as found by prior work) for both gases. However, the entropic contributions to the chemical potential of the dissolved species are so significant that we predict a reversal in the direction of diffusive transport —downwards —in deeper storage reservoirs and those with larger geothermal gradients. For CO_2, relatively low geothermal gradients of 10 C/km are sufficient to induce downwards diffusion at depths relevant to storage. Diffusive transport of H_2 is less affected but still reverses direction under typical gradients, e.g., 30 C/km, at a depth of 1000 m. Contrary to previous studies, we find that diffusion and density-driven convection will likely work in concert to drive aqueous phase transport of these fluids: both mechanisms work to drive CO_2 downwards, and H_2 upwards, for conditions representative of their respective engineered storage reservoirs.[4]

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References

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