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Enhanced Cap Rock Integrity and Self-Sealing of the Immiscible Plume through Mineral Trapping during Prograde and Retrograde CO_2 Sequestration in Saline Aquifers

Successful long-term isolation of anthropogenic CO_2 within confined saline aquifers hinges on long-term integrity of the cap rock, which prevents upward CO2 migration. Such integrity must span both the relatively brief prograde (active-injection) and indefinite retrograde (post-injection) phases of the sequestration process. Concomitant lateral containment of CO_2 migration within the aquifer itself represents another significant component of successful isolation. Reactive transport modeling of CO_2 sequestration in shale-capped saline aquifers has identified four distinct mineral-trapping mechanisms that continuously enhance cap-rock integrity and self-seal the intra-aquifer immiscible plume throughout the prograde and retrograde regimes. In each case, carbonate precipitation results from kinetically controlled mass transfer that involves ambient formation minerals, the aqueous wetting phase, and immiscible CO_2, whose post-injection residual saturation catalyzes retrograde mineral trapping. Specifically, siderite-magnesite precipitates within the clay-rich shale cap rock while dawsonite precipitates throughout the intra-aquifer plume, whose lateral and upper margins are delimited by simultaneous, genetically distinct precipitation of siderite-magnesite-calcite rind. Although local porosity and permeability are reduced—and therefore CO_2 migration is retarded—by each of these four mechanisms, such reduction is most extreme in the cap rock because of its relatively large Fe and Mg concentrations, which derive from a preponderance of clay minerals. During a 20-year (10-year prograde, 10-year retrograde) simulation, mineral trapping from these four processes sequesters less than one percent by mass of injected CO_2. However, this seemingly negligible contribution has enormous strategic significance: by continuously enhancing cap rock integrity, it ensures isolation of the voluminous immiscible plume and solubility-trapped CO_2.