Effects of geochemical reaction on double diffusive natural convection of CO_2 in brine saturated geothermal reservoir

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ABSTRACT

In order for mitigation of greenhouse gas emissions, as we know, the storage of CO_2 in deep geological formations is considered as a promising option. Besides convective mixing an understanding of geochemical reaction that affects the long term storage process in deep saline aquifers is of great importance. In this study we numerically investigate double diffusive natural convection of CO_2 in brine saturated geothermal reservoir which is anisotropic in permeability variations, impervious from the sides, and is open to CO_2 at the top. Our primary objective is to understand effects of geochemical reactions of different rates and orders on density driven natural convection of CO_2 due to concentration and geothermal gradients. We present propagation of CO_2 plumes over long period of time by analyzing different combination of problem parameters: Damkohler number ($0.01 \leqslant Da \leqslant 10^5$) with variation of 1st, 2nd, and 3rd order reactions; solutal Rayleigh number ($500 \leqslant Ra_s \leqslant 2000$); the buoyancy ratio ($2 \leqslant N \leqslant 100$); Dykstra–Parsons coefficient ($0.35 \leqslant V_{dp} \leqslant 0.85$); and fixed Lewis and Prandtl numbers. Reaction order is set from relevant stoichiometric ratio. In each case results are quantified in term of percentage of CO_2 deposition. It is found that mineral interactions make traceable difference of depositions as reaction rate increases, especially when $Da > 10^3$. In one hand, compared to very minimum geochemical interaction ($Da \sim 0.01$) the strong reaction effects ($Da \sim 10^4$) can make difference of more than 5% in the period of 500 years of trapping. On the other hand, at a fixed equivalent Da reaction order also makes substantial distinction as deposition time passes. Heterogeneity plays a vital role. Geothermal gradient has very negligible effect, however only after very long time (>500 years).

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1. Introduction

Deep saline aquifers have the highest storage potential to bridge the gap between climate change mitigation and CO_2 emissions [30,15,1,18,22,2,20]. Physical, residual, solubility, and mineralogic trapping are the proposed mechanisms by which CO_2 is hold in the saline aquifers and does not escape to the surface [16,29,2,24]. Among these mechanisms, mineralogic trapping is the only technique that sequesters the CO_2 while others are referred as storage mechanisms [10]. Hence, modeling of geochemical processes is a necessary predictive tool for long-term CO_2 disposal in deep aquifers. When CO_2 is injected into a saline aquifer, it can react with other chemical species in aqueous phase as well as constituents on the solid phase. The geochemical changes caused by CO_2 injection into aquifers include acidification and carbonation of the native brine and potential mineral dissolution and precipitation reactions driven by the aqueous composition changes [25]:

$$CO_2(g) = CO_2(aq)$$

$$H_2O + CO_2(aq) = H_2CO_3$$

$$H_2CO_3 = H^+ + HCO_3^-$$

$$CaCO_3(S) + H^+ = Ca^{+2} + HCO_3^-$$
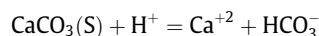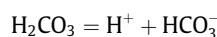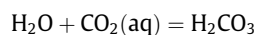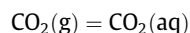
High HCO_3^- concentration in the aqueous phase is a driving force for CO_2 to be dissolved more in the aqueous phase [21]. Such reactions can change the hydrological and chemical properties of subsurface systems, alter flow pathways, and eventually affect contaminant transport and behavior significantly [33]. Mineral dissolution and precipitation reactions are important for evaluating the potential CO_2 storage capacity and can also influence the performance of the injection well. Many researchers investigated the geochemistry in the CO_2 sequestration, mostly related to kinetics problem [8]. Noh et al. [25] proposed a mathematical formulation by which geochemistry is integrated with the multi-phase flow. Local equilibrium was assumed in the formulation. The theory led to graphical

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Nomenclature

c	concentration [mol/m^3]	β_T	coefficient of thermal expansion [K^{-1}]
C_p	heat capacity at constant pressure [J kg^{-1} K^{-1}]	ϕ	porosity [–]
D	Diffusion coefficient [m^2/s]	μ	viscosity [kg m^{-1} s^{-1}]
Da	Damkohler number	κ	thermal conductivity
g	gravitational acceleration [m/s^2]	ρ	density [kg/m^3]
h	enthalpy [J kg^{-1}]	ψ	stream function [m^3 m^{-1} s^{-1}]
H	porous medium height [m]		
k	permeability [m^{-2}]	*Superscript*	
L	porous medium length [m]	$*$	dimensionless quantity
n	number of nodes		
p	pressure [Pa]	*Subscripts*	
λ	wave number [–]	0	initial value
Le	Lewis number [–]	a	stoichiometric coefficient of CO$_2$ brine phase
Pe	Peclet number [–]	b	stoichiometric coefficient of minerals
Pr	Prandtl number [–]	dp	Dykstra–Parsons
Ra	Rayleigh number [–]	i	node in x – direction
$\overline{S}$	average deposition [%]	j	node in z – direction
T	Temperature [°C]	m	minerals
t	time [s]	s	solutal
u	velocity [m/s]	x	x-coordinate
V	Dykstra–Parsons coefficient	z	z-coordinate
x	distance along x-axis		
z	distance along z-axis		

Greek letters

α	thermal diffusivity
β_c	coefficient of density increase by concentration [m^3/mol]

solution from which it is easy to see when and under what conditions mineralization occurs during the injection. Okamoto et al. [28] used GEM-GHG simulator and performed a sensitivity study of CO$_2$ mineralization to investigate effective parameters on CO$_2$ behavior over a long time. They used a real reservoir model of Naga-oka pilot test site in Japan. Reactive surface area and component of minerals were reported as essential parameters. Fischer et al. [6] conducted an experiment to study the significance of the CO$_2$-brine-rock interactions during the geological storage of the CO$_2$. In this work, CO$_2$-treated samples were compared against untreated samples to investigate the CO$_2$ effects on the mineralogy of a sandstone rock. Their experimental results showed the stabilization or precipitation of albite and dissolution of calcium-rich plagioclase, K-feldspar and anhydrite. Wolf et al. [39] investigated the in situ reactions involved in CO$_2$ sequestration through a micro-reaction system. In their study synchrotron X-ray Diffraction and Raman Spectroscopy were used to characterize the carbonation reaction. O'Connor et al. [27] also conducted an experiment to study mineral sequestration of CO$_2$. Several suggestions were documented to accelerate the slow geological processes including carbon dioxide activity increase in solution or the reactive surface area and imperfections into the crystal lattice through high energy attrition grinding, activation of minerals through thermally, etc. Wellman et al. [38] investigated the CO$_2$-brine-rock interactions by some corefloods, and then used TRANS-TOUGH simulator (combination of TOUGH-EOSCO2 with the chemistry code TRANS) to model their experimental work and to perform some sensitivity analysis. Lithology type and the magnitude of fluid flushed through the media were documented to be the two major controlling factors in this process. Rosenbauer et al. [31] showed that more CO$_2$ might be sequestered in a deep saline aquifer not only through the mineral trapping but also by the formation of bicarbonate ion (HCO$_3^-$) in the aqueous phase. Druckenmiller et al. [3] reported the main

parameters in terms of the geochemistry for a CO$_2$ sequestration process as, pressure; temperature; pH; brine concentration; and rock composition. It has been also included that the temperature effect is more pronounced compared to the pressure effect. Xu et al. [41] developed a reactive geochemical transport model in TOUGHREACT for a sandstone-shale system under high CO$_2$ pressures. The model was then used to interpret the mass transfer of aqueous chemical components, the alteration pattern of minerals, sequestration of CO$_2$, and changes of petrophysical properties (e.g., porosity) in a Gulf Coast aquifer. Schumacher and Anne [34] used Geochemist's Workbench software (GWB) to analyze the CO$_2$–water–rock interactions occurring during a carbon sequestration pilot test into a Mississippian oil reservoir. Several core and cutting samples as well as electric logs were used to model the static and dynamic of the pilot during the CO$_2$ injection. Simulation results show net dissolution during the injection stages while net precipitation for post-CO$_2$ injection.

In recent studies [13,14] double diffusive natural convection of CO$_2$ in brine saturated homogeneous and heterogeneous permeable geothermal reservoir were investigated. Here we have coupled the geochemistry and convective mixing in anisotropic homogeneous medium and have presented CO$_2$ settlement results to understand the role of these processes on the long term fate of the sequestration.

2. The reservoir model and governing equations

The hypothetical geothermal reservoir is considered to be a two-dimensional rectangular cavity filled with brine saturated porous medium with height H, and length L as shown in Fig. 1 Permeability varies spatially, i.e., $k = k(x, z)$. There is no flow condition across all boundaries and no solute flux across lateral and bottom boundaries are assumed. There is no heat flux across the lateral