Quantification of CO₂ masses trapped through free convection process in
isothermal brine saturated reservoir

Akand Islam^{*}, Alex Sun

Bureau of Economic Geology, University of Texas at Austin, Austin, TX 78758, USA

^{*}corresponding author: awislam@utexas.edu

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USA

Abstract. Dissolution trapping of supercritical CO₂ into formation brine has been investigated as a potential mechanism for reducing buoyancy force in carbon storage formations. This study attempts to quantify how much CO₂ can be stored through dissolution trapping assuming the free-phase CO₂ will be dissolved continuously on the top of perturbed brine phase. To our knowledge, most former investigations focused on physical explanations of density-driven free convection instability. Our aim is to compute the amount of CO₂ (by mass) captured by dissolution trapping until the model reservoir reaches steady state. The numerical experimentation is done using dimensionless mass and momentum conservation laws. The major problem parameter here is the Rayleigh number, for which we carry out an extensive survey to find out its low and high ends based on field and observed data from the literature and in-house database. Because density difference is the main driving force, we also investigate the effects of possible impurities retained in CO₂ stream and the resulting effect on density contrast. We study results of both homogeneous and heterogeneous permeable reservoirs. Also, different boundary conditions (Neumann, Dirichlet, and periodic) are compared to understand their effects. The simulations are run until almost complete saturation (~99%) is reached. For a test case ($T = 40$ °C, $P = 50$ bar) of geologic and thermophysical conditions, we have found that on average 0.33 – 15 g CO₂ will dissolve per year until a heterogeneous unit reservoir volume of 1 m³ reaches complete saturations. For the case of homogeneous reservoir this amount is 0.28 – 6 g.

Keywords: CO₂ geologic sequestration; free convection; convective instability; solutal Rayleigh number; impurities

^{*}corresponding author: awislam@utexas.edu

Notations

c	concentration [mol/m ³]
D	Diffusion coefficient [m ² /s]
g	gravitational acceleration [m/s ²]
i	component id
H	Reservoir thickness [m]
k	mean permeability [m ²]
L	Reservoir length [m], characteristic length
m	molality
n	number of nodes
N	number of components [-]
p	pressure [Pa]
λ	wave number [-]
Pe	Peclet number [-]
Ra	Rayleigh number, $Ra = \frac{\Delta\rho g k H}{\phi D \mu}$ [-]
$\bar{c}$	average deposition [%]
T	Temperature [°C]
t	time [s]
U	characteristic velocity [m/s]
u	velocity [m/s]
V	Dykstra-Parsons coefficient
X	mass fraction [kg solute/kg solution]
x	distance along x-axis
z	distance along z-axis

Greek letters

α	thermal diffusivity
β	coefficient of density increase by concentration [m^3/mol]
ϕ	porosity [-]
μ	viscosity [$\text{kgm}^{-1}\text{s}^{-1}$]
ρ	density [kg/m^3]
ψ	stream function [$\text{m}^3\text{m}^{-1}\text{s}^{-1}$]

Superscript

*	Dimensionless quantity
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Subscripts

0	initial value
dp	Dykstra-Parsons
i	node in x – direction
j	node in z – direction
r	reference value
x	x-coordinate
z	z-coordinate
∞	infinite dilution

1. Introduction. Carbon capture and storage is being studied as a viable technology to reduce global greenhouse gas emissions into atmosphere [1]. The overall energy demand has increased as population increases and standard of living progresses. The energy usages are directly related

to CO₂ emissions [2], which are now higher than 30 gigatons a year [3, 4]. To mitigate CO₂ emissions and thus enable global transitioning to a low-carbon energy infrastructure, industrial-scale sequestration of CO₂ into deep sedimentary basins ($\geq 800\text{m}$) has been identified as the most promising geoengineering control measure [5-12]. Among all sedimentary basins, the most abundant potential storage sites are brine saturated deep saline aquifers. Other possible sites include depleted oil and gas reservoirs, coal beds, and seabed sediments [13]. Estimates of the subterranean storage capacity of CO₂ range from 1700 to 14000 gigatons or higher [3, 9, 13]. This wide range reflects incomplete knowledge on properties that constitute a suitable storage site, as well as the lack of conclusive geological data of potential sites.

From the context of viability and public safety it is essential that the long term (tens of thousands of years) underground storage of sequestered CO₂ is assured. In the US, the primary goals of its carbon storage program are to (a) develop technologies to help the industry to predict CO₂ storage capacity in geologic formations with a ± 30 percent margin of error and (b) ensure that at least 99 percent of the injected CO₂ is stored permanently [14]. Sequestration may occur through a combination of different physical and chemical trapping mechanisms [15]. Because the injected CO₂ is buoyant, the presence of an impermeable caprock, typically a shale or clay layer, below which the CO₂ can pool and spread under gravity, is critical for any storage site. Although in principle hydrodynamic trapping provides the dominant trapping mechanism for sequestering CO₂, unintended migration through fractures, faults, and abandoned wells can occur [16-20]. There are other processes which take place over different timescales. They are: residual trapping through which small pockets of CO₂ are immobilized in the brine by capillary forces; mineral trapping in which chemical reactions with host rocks lead to the precipitation of CO₂; and dissolution trapping by which CO₂ dissolves into the host brine [3, 21-23]. Our primary motivation of this research is quantification of the maximum possible amount of trapped CO₂ through dissolution trapping via convective mixing.

This study is addressing the post injection period. Generally injection occurs at high pressures to ensure that CO₂ is in the so-called supercritical state, where it occupies a considerably smaller

volume than it would otherwise do as gaseous phase at lower pressures. Given that the pores of the injection formation are already brine saturated, after injection has ceased dense brine with CO₂ will overlay lighter brine without CO₂, causing an unstable setting and potentially leading to free convective mixing would result in more secure storage. Geothermal gradients affect the convection process insignificantly even after long periods [24, 25]. On the other hand, geochemical field observation in natural CO₂ reservoirs suggests that convective dissolution plays a non-neglecting role for the transport of CO₂ [26]. In term of natural convection, geochemical interactions can enhance dissolution by 5% in the first 500-year period and the effects become more significant in time [22]. The batch reaction effects in CO₂ storage are discussed extensively in the literature [27-30]. Here the geothermal and geochemical effects are not considered. Convective mixing could be a major contributor to the dissolution rate because it continuously provides new fresh brine to regions with high CO₂ concentration, thereby allowing more CO₂ to dissolve [31, 32].

For the post-injection period, it is important to understand the role of convective mixing in CO₂ sequestration and the time scales involved, which have direct implications on the safety and effectiveness of geologic carbon storage. The long term behavior of the flow cannot be predicted from the initial concentration profiles, especially for large Ra (≥ 100) numbers [24, 33, 34]. Because of unstable flow, the dissolution rate fluctuates about a constant mean value. Pruess and Zhang [35] reported that the dissolution rate during active convection is approximately 1.5 to 2 times larger than the dissolution rate at the onset of convection. They concluded that the dissolution is enhanced by the convective process significantly, provided that the dissolution rate is a function of $t^{-1/2}$ in the absence of convection. Laboratory results from Kneafsey and Pruess [36], Faisal et al. [37], and Yang and Gu [38] are consistent with simulation results both qualitatively and quantitatively. The convective mixing is studied from various contexts, such as linear stability analysis [25, 34, 39-44], anisotropic permeability effect [32, 43, 45], hydrodynamic dispersion [46], geothermal effect [22, 24, 25, 44], and geochemical interactions [22, 47]. Field scale simulations are computationally expensive. Hassanzadeh et al. [48] presented simple scaling relationships that characterize the long term behavior of the mixing process for better understanding the final disposition of CO₂ in saline aquifers. For

computational study, the authors also reported the appropriate grid block size needed for numerically error free modeling. Pau et al. [33] performed high resolution numerical simulations and concluded that the CO₂ mass flux due to density driven convection is ~25% higher when the simulations are conducted for a 3D geometry than that for a 2D domain. Chen et al. [49] also investigated 3D simulations. In most cases the aforementioned publications have discussed the physics of plume advancements during the injection time (typically 30-40 years) and/or early periods of post injection periods, ranging from a couple of hundreds to thousands of years. Here we give particular emphasis on longer periods of simulations so that the reservoir model investigated reaches about 99% saturation. Because convective mixing is unstable, complete settlement is not possible in theory.

For free convection problems, Ra governs the overall calculations [13, 50-53]. Different from most previous studies that assume arbitrarily high value of Ra , we use realistic Ra values based on field data reported in the literature. Besides the reservoir data (permeability, porosity, etc.), thermophysical conditions (i.e., density change in brine due to CO₂ dissolution, diffusion constant, and viscosity) also affect Ra . In addition, it is unlikely that the injected CO₂ stream is completely pure. The flue-gas streams from power generation and other industrial and chemical processes often have impurities such as, N₂, Ar, O₂, CO, and, potentially, H₂, SO_x, NO_x, and other acid species [54]. Density changes, due to the non-condensable impurities which cannot be liquefied at ambient temperature can affect the buoyancy [55]. This is surveyed in order to determine the overall effect on convective mixing. To understand the effect of CO₂ free convection in porous media, three types of boundary conditions, Neumann [22, 34, 45, 49, 56], Dirichlet [50], and periodic [33, 35] have been applied. We present detailed comparisons of these conditions in terms of trapping. The mass of CO₂ trapped is quantified for both low and high ends of Ra 's for the cases of homogeneous and heterogeneous reservoirs.

2. The model reservoir and governing equations. The model reservoir for the mechanistic study is a 2D domain filled with brine saturated porous medium with height H , and length L as illustrated in **Figure 1**. Initially, there is no dissolved CO₂ in the system. We assume the

interface at the top is relatively sharp and is not deflected by the density fingering in the underlying brine. This condition is motivated by the continuous phase equilibrium establishment [32]. For purely horizontal flux at the top (infinite resistance for the brine to flow upward beyond the interface) we obtain $u_z(x, z=0, t) = 0$. Capillary transition between gas and brine phases is ignored. Hence this is reduced to a single phase problem. The Boussinesq approximation and Darcy flow model are applied. The velocity-induced dispersion effect is ignored. Because this is a free convection problem Ra is low enough to expect only the laminar flow. Isothermal condition is kept valid throughout the entire process. Therefore energy balance is not taken into consideration. Density contrast due to concentration difference is the only driving force to impose free convection. The governing equations are as follows:

(a) Continuity equation,

$$\phi \frac{\partial \rho}{\partial t} + \frac{\partial (\rho u_x)}{\partial x} + \frac{\partial (\rho u_z)}{\partial z} = 0. \quad (0)$$

(b) Darcy's law,

$$u_x = -\frac{k_x}{\mu} \frac{\partial p}{\partial x}, \quad (0)$$

$$u_z = -\frac{k_z}{\mu} \left(\frac{\partial p}{\partial z} - \rho g \right). \quad (0)$$

(c) Mass transport equation,

$$\phi \frac{\partial c}{\partial t} + u_x \frac{\partial c}{\partial x} + u_z \frac{\partial c}{\partial z} = \phi D \left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial z^2} \right). \quad (0)$$

In order to account for the density change due to dissolved CO_2 , a simplified equation-of-state is used

$$\rho = \rho_0 [1 + \beta (c - c_0)], \quad (5)$$

where $\beta = \frac{1}{\rho_0} \left[\frac{\partial \rho}{\partial c} \right]_T$.

After differentiation we obtain

$$\frac{\partial \rho}{\partial x} = \rho_0 \beta \frac{\partial c}{\partial x}. \quad (6)$$

To eliminate the pressure term Eqs. 2 and 3 are cross differentiated. We get

$$\frac{1}{k_z} \frac{\partial u_z}{\partial x} - \frac{1}{k_x} \frac{\partial u_x}{\partial z} = \frac{g \rho_0 \beta}{\mu} \frac{\partial c}{\partial x} \quad (7)$$

2.1 Dimensionless form of the equations:

The dimensionless variables are defined by,

$$\begin{aligned} x^* &= \frac{x}{H}, \quad z^* = \frac{z}{H}, \quad u_x^* = \frac{H u_x}{\phi D}, \quad u_z^* = \frac{H u_z}{\phi D}, \quad k_x^* = \frac{k_x}{k}, \quad k_z^* = \frac{k_z}{k}, \quad \psi^* = \frac{\psi}{\phi D}, \\ t^* &= \frac{Dt}{H^2}, \quad c^* = \frac{c - c_r}{c_0 - c_r}, \quad u_x^* = -\frac{\partial \psi^*}{\partial z^*}, \quad u_z^* = \frac{\partial \psi^*}{\partial x^*}, \quad Ra = \frac{\Delta \rho g k H}{\phi D \mu}, \quad \text{and } \Delta \rho = \rho_0 \beta (c - c_0). \end{aligned} \quad (8)$$

Upon following the Boussinesq approximation the final forms of dimensionless equations are reduced to (* is dropped for clarity)

$$k_z \frac{\partial^2 \psi}{\partial x^2} + k_x \frac{\partial^2 \psi}{\partial z^2} = Ra \frac{\partial c}{\partial x}, \quad (9)$$

$$\frac{\partial c}{\partial t} + u_x \frac{\partial c}{\partial x} + u_z \frac{\partial c}{\partial z} = \frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial z^2}. \quad (10)$$

The coupled Eqs. 9 and 10 are solved to compute ψ and c .

2.2 Initial and boundary conditions:

The initial conditions are

$$\psi = 0, \quad c = 0. \quad (11)$$

The Dirichlet boundary conditions are

$$\psi(x, z)=0, c(x=0, z; x=1, z; x, z=1)=0. \quad (12)$$

The Neumann boundary conditions are presented by

$$\psi(x, z)=0, \frac{\partial c}{\partial x}(x=0, z; x=1, z)=0, \frac{\partial c}{\partial z}(x, z=1)=0 \quad (13)$$

The periodic boundary conditions read

$$\psi(x, z)=0, \text{ continuous } c \text{ at lateral boundaries, } \frac{\partial c}{\partial z}(x, z=1)=0. \quad (14)$$

Fig 1. Model domain for the mechanistic study.

3. Discussions on Ra . From the classical definition, we know Ra is ratio of the driving strength of buoyancy to the dissipative effects of diffusion and viscosity. Reservoir formation parameters (ϕ, k, H) and thermophysical parameters ($\Delta\rho, D, \mu$) of associated fluid determine the value of Ra , which means Ra may vary by orders of magnitudes depending on reservoir characteristics

and fluid's physical behavior. However for this particular problem $\Delta\rho$, D , and μ are subjected to change with temperatures and pressures and these can be varied by couple of factors up to five, if we assume typical temperature and pressure of geologic sequestration up to 105 °C and ≤ 600 bar, respectively [57]. To measure these parameters it is assumed that CO₂ and brine phase are in thermodynamic equilibrium. $\Delta\rho$ can be at most ~ 20 kg/m³ compared to pure water density. Among common gases, only CO₂, SO₂, Ar, Kr increase the density of the dissolved phase. After applying simple additive mixing rule of molar volumes [32] and considering the impurities in CO₂ streams (Ar, CO, CO₂, N₂, NO, O₂, and SO₂) listed in Refs. [54, 58-61] we find that $\Delta\rho$ is within the maximum limit (~ 16 kg/m³). Table 1 shows mass fractions of possible impurities dissolved in brine. The mixing rule followed is shown below

$$\bar{\rho} = \sum_{i \neq \text{CO}_2}^{N-1} \rho_i^{\infty} X_i + \rho_{\text{CO}_2} \left(1 - \sum_{i \neq \text{CO}_2}^{N-1} X_i \right) \quad (15)$$

Table 1. Infinite dilution densities and mass fractions of possible dissolved impurities.

Impurity (<i>i</i>)	ρ_i^{∞}	X_i
Ar	1225	2.52e-5
CO	751	1.77e-5
N ₂	785	1.18e-5
NO	<1340	5.6e-7
O ₂	964	2.29e-5
SO ₂	1664	2.46e-2
H ₂ S	979	1.85e-3
CH ₄	465	2.55e-5

The D can differ by one order-of-magnitude ($8.91 \times 10^{-10} - 8.2 \times 10^{-9}$ m²/s) based on temperatures and pressures [62]. Recently, Islam and Carlson [63] investigated μ calculations for geologic sequestration and found the range of value as 0.3-1.3 mPa.s. The most significant effect on these parameters is that of temperature, while the effects of concentration, pressure, and salinity are minor. Typically, reservoir porosity ϕ and average k vary between 1-40%, and 1-4000 mD, respectively. H ranges from a few to a couple of hundred meters [24]. These parameter values give rise to Ra in the range $10 < Ra < 10^5$. However the upper bound of the Ra range is extremely

unlikely. To our knowledge, from the real field data the highest Ra for geologic sequestration is reported to be $\sim 80,000$. In general $Ra \leq 1750$ and $Ra > 1750$ are considered moderate and high Rayleigh number regimes, respectively [13]. Ra 's compiled from the peer-reviewed literature are presented in Table 2 [25, 32, 33, 39, 47-49, 56, 64-67]. The reservoirs with ID 36-50 in this table are collected from Texas Atlas, a database source by Texas Bureau of Economic Geology [68]. The database has a collection of 3200 fields. Because we are interested in the range of Ra , especially the high ends, only the data of very low and high permeable reservoirs are shown after sorting.

Table 2. Compilations of Ra 's from field and hypothetical published data.

Reservoir ID	H	D	$\Delta\rho$	ϕ	$\mu \times 10^{-3}$	$k \times 9.87 \times 10^{-16}$	Ra
1	15	4.1e-9	4.65	0.06	0.60	30	142
2	10	4.0e-9	3.56	0.18	0.66	186	139
3	18	3.9e-9	1.58	0.05	0.78	40	74
4	8	3.7e-9	4.30	0.10	0.65	100	141
5	10	5.1e-9	1.51	0.07	0.64	16	11
6	13	5.5e-9	3.22	0.12	0.50	30	38
7	4	3.2e-9	4.41	0.13	0.74	6	3
8	81	3.7e-9	4.28	0.04	0.67	9	318
9	29	4.2e-9	3.42	0.20	0.63	6	11
10	9	4.5e-9	8.90	0.12	0.46	9	29
11	60	7.6e-9	4.09	0.09	0.36	137	1359
12	10	7.4e-9	3.40	0.06	0.39	75	146
13	10	5.0e-9	6.10	0.08	0.48	115	364
14	10	5.5e-9	6.15	0.12	0.44	9	19
15	10	2.7e-9	0	0.06	1.32	14	0
16	40	2.6e-9	6.03	0.22	0.82	67	351
17	4	3.7e-9	5.33	0.10	0.60	346	329
18	24	4.5e-9	3.37	0.11	0.61	10	27
19	13	4.3e-9	2.82	0.12	0.65	13	14
20	13	4.3e-9	4.78	0.12	0.57	32	67
21	40	5.0e-9	1.85	0.05	0.63	27	127
22	5	4.6e-9	4.53	0.06	0.55	109	162
23	26	5.9e-9	3.46	0.12	0.48	1	3
24	10	5.5e-9	2.94	0.10	0.52	130	133
25	60	4.29e-9	12	0.09	0.50	47	429
26	13	3.53e-9	12	0.09	0.50	6	14
27	0.125	2.0e-9	20	0.10	0.50	800	196
28	0.04	2.0e-9	20	0.10	0.50	1000	392

29	6	2.0e-9	20	0.10	0.50	80	943
30	8	2.0e-9	10.45	0.15	0.59	500	2317
31	20	2.15e-9	11.40	0.20	0.80	500	3251
32	160	2.2e-9	14.42	0.30	0.84	2000	81651
33	160	3.4e-9	14.42	0.30	0.58	1000	38209
34	160	4.9e-9	14.42	0.30	0.43	500	17653
35	160	6.3e-9	14.42	0.30	0.34	100	3476
36	19	2.0e-9	10.00	0.25	0.80	4000	19030
37	33	4.0e-9	10.4	0.24	0.80	3394	15122
38	6	2.0e-9	11	0.30	0.90	2500	3023
39	10	2.5e-9	10.4	0.24	0.89	2460	4700
40	8	2.5e-9	8.5	0.34	0.89	2400	2091
41	50	4.0e-9	15.9	0.31	0.49	2350	3016
42	7	2.0e-9	5.9	0.33	0.40	2290	3514
43	15	2.0e-9	14.90	0.26	0.40	2000	24139
44	5	3.0e-9	4.80	0.22	0.30	1758	2090
45	40	2.1e-9	12.0	0.39	0.18	0.1	3
46	16	2.0e-9	10.0	0.10	0.40	1	19
47	18	6.0e-9	16.0	0.11	0.80	2	10
48	12	5.0e-9	15.0	0.08	0.60	5	36
49	111	5.0e-9	14.0	0.08	0.60	10	680
50	20	2.0e-9	10.0	0.09	0.40	20	545

1-24: Alberta basins (Canada), 25: Nisku aquifer (Canada), 26: Glauconitic Ss (Canada), 27-33: Hypothetical, 32-35: North Sea (Norway), 36: Sulphur Bluff, 37: Hawkins, 38-Withers North, 39: Pewitt Ranch, 40: Durke, 41: Webster, 42: Tomoconnor, 43: Talco, 44: Tomoconnor, 45: Salt Flat 46: Sand Hill, 47: Union, 48: Shafter Lake, 49: Wasson 72, 50: Nolley. The maximum and minimum Ra 's obtained are colored red.

For smaller Ra values, convective mixing does not have significant impact; for $Ra > 1000$, on the other hand, convective dissolution is expected to play significant role in long term stability of stored CO_2 . When Ra is less than the critical value ($4\pi^2$) no convection cells will be formed and the CO_2 fronts will advance only by molecular diffusion, resulting in extremely slow mixing process. Convection in a 2D Rayleigh-Darcy cell for low and moderate values of Ra (≤ 1300) has been the subject of many studies [13, 69]. For $382 \leq Ra \leq 1300$, convective instabilities in the boundary layers cause a series of bifurcations that perturb but do not completely break down the background cellular structure of the flow. The perturbations take the form of “dripping” plumes which grow from the boundary layers but are washed around with background circulation. The dynamics render significant hysteresis in the regime, and so the values of Ra at

which bifurcations occur are not apparent. When $1300 \leq Ra \leq 5000$, the quasi-steady background rolls are completely broken down by the growth of destabilizing fronts from the upper and lower boundaries [13, 70-73]. For $Ra > 5000$ mass flux from the upper boundary becomes independent of Ra [74] and vigorous mixing is observed in early periods.

In the process of free convection, regime changes in time rather than with Ra . Primarily the flow dynamics are localized to the source of solute at the upper boundary and the layer thickness does not play a role until the fronts reach the bottom boundary. Hence, most regimes are parameter independent and the Ra only controls how many of these regimes are encountered before the bottom influence the dynamics and the flow transitions to a final Ra dependent regime [75]. After hitting the bottom boundary the regimes merge and thus flow becomes Ra - and perturbation independent. Hewitt [13] recently performed an extensive analysis specially on high Ra concentrating for porous medium.

4. Results and Discussions. Numerical methods applied in our calculations are discussed in Refs. by Islam et al. [22, 24, 44]. The simulations are performed with uniform grid size of 81×81 , and a time step of 10^{-6} (equivalent to about one month based on 100 m thick 2D domain) to meet the CFL conditions. The developed code predictions were checked against literature benchmark solutions [22, 56, 76]. Reproducibility of dissolutions in homogeneous medium is shown in Table 3 for $Ra = 1000$. The results were in satisfactory agreement independent of mesh sizes. Likewise, in this study we have also applied a sinusoidal perturbation condition having wave number of 24 in order to observe the non-linear behavior, i.e., the fingering behavior. Because we are interested in end result of overall dissolution, any pore-level instability that may occur in reality is ignored.

Table 3. Comparison of computed dissolution (homogeneous medium, $Ra = 1000$)

Time (yr)	Dissolution $\bar{c}$ (Eq. 16)			
	Islam et al. [16]	Hassanzadeh et al.[56]	Simpson and Clement [76]	present study

10	0.02	0.019	0.021	0.02
100	0.04	0.039	0.041	0.04
500	0.11	0.10	0.13	0.12

In order to quantify the amount of CO₂ that can be dissolved through free convection, we have chosen a very typical reservoir conditions at $T = 40$ °C and $P = 50$ bar. The reservoir size is assumed to 10^4 m³ ($H = L = 100$ m with unit length width) and the pore volume 2000 m³ ($\phi = 0.2$). Other formation and thermophysical parameters are such that $Ra = 1000$. The salinity of brine aquifer is considered to be 1.0 molality ($m = 1$). The overall dissolution in a definite time period t is computed by

$$\bar{c} = \frac{\int_0^x \int_0^z c(x, z, t) dx dz}{\int_0^1 \int_0^1 dx dz}. \quad (16)$$

Peclet number ($Pe = \frac{LU}{D}$) is the ratio of advection to diffusion. This generally explains the local convective non-linear flow nature of the plumes in a particular grid point. However, here we have calculated this as an average value so that it represents plume advancements globally. After using the non-dimensional scaling relations Pe reads as

$$Pe = \phi \bar{u}_z \bar{z}. \quad (17)$$

where $\bar{u}_z = \frac{\int_0^x \int_0^z u_z(x, z, t) dx dz}{\int_0^x \int_0^z dx dz}$ and $\bar{z} = \frac{\int_0^z z_{\max} dz}{\int_0^z dz}$. Here $z_{\max}$ is the location of the most advanced plume in z direction.

Before quantifying the heterogeneity and boundary effects, the plumes' advancements are analyzed for a typical case, i.e., $Ra = 1000$. First, we present results of homogeneous reservoir ($k_x = k_z$). As mentioned, the simulations are run until the reservoir is almost CO₂ saturated in the case of three boundary conditions discussed. **Figure 2** shows variation of the $\bar{c}$ vs. t . The results clearly show that after the injection has ended the brine reservoir will be about 99% saturated by CO₂ in around 51000 years' period. Because the interface is perturbed, i.e., non-uniform dissolution, complete 100% saturation may not be reached theoretically. Interestingly quantitatively results are the same (differ by 0.15%) for the case of both lateral Neumann and periodic boundary conditions. Pruess and Zhang [35] have also reported that total dissolution rates are not sensitive to the boundary conditions. However, per their calculations results can be up to 15% difference. They have not clearly mentioned between which boundary conditions this variation can occur. On the other hand, Dirichlet boundary condition limits dissolution up to only 40%. Unlike other two cases the effect of lower boundary will never be felt and thus will never be reverse mixed by the fronts. Actually for solutal convection this is not a realistic condition and therefore to forecast the CO₂ trapping or to understand the physics of flows only the Neumann and periodic boundary conditions are valid. **Figure 3** illustrates time lapse of Pe number. This shows that, though ultimate dissolutions remain the same in the case of Neumann and periodic conditions, the flow mechanisms differ apparently. As expected as time marches after onset of convection the flow gets more acceleration until around 1000 years. At this time the rate of mixing reaches to its peak and then upon the reverse mixing of the plumes after hitting bottom of the aquifer the initial density difference decreases causing deceleration of flows. The dynamics of the system slows down. Denser CO₂ saturated fluids can pinch-off and become disconnected from the big-chunk plumes. Among different boundaries there can be considerable variability in the advancement of fingers. The rate of advancement of the fastest finger can be varied much. Not surprisingly, Pe becomes stagnant in the case of Dirichlet condition after 10000 years of mixing. Because the unstable flows are highly non-linear and because of non-uniform local lateral pressure gradients in the plumes, Pe changes during the slowing down process and cannot be expected to be completely monotonic. As expected, at later times ($t > 10000$ years) the convective mixing process becomes slower. In order to visualize different natures of CO₂ flows among the boundary conditions concentration profiles, examples are presented in **Figures 4 - 7** for times $t = 500, 1000, 10000$, and 50000 years, respectively. Axes

and color scales are the same in all isochores' maps. Near the boundaries, the flow is influenced by short wave-length boundary-layer instabilities, which give rise to the rapid growth of protoplumes. These plumes are driven laterally and entrained into the interior big plume flow. Except for the Dirichlet condition, mixing process after 10000 years of convection becomes indifferent.

Fig. 2. CO₂ saturations over time in three boundary conditions.

Fig. 3. Time (t) lapse of Pe number for three boundary conditions.

Fig. 4. Concentration profiles at $t = 500$ years of Dirichlet, Neumann, and periodic Boundary conditions, respectively.

Fig. 5. Concentration profiles at $t = 1,000$ years.

Fig. 6. Concentration profiles at $t = 10,000$ years.

Fig. 7. Concentration profiles at $t = 50,000$ years.

Figure 8 shows results of saturations in heterogeneous reservoir compared to homogeneous. In heterogeneous medium we can expect to trap CO_2 completely much faster (~ 35000 years). As discussed earlier by the authors in [44, 45] this is due to the fact that the convective mixing in heterogeneous media is different than in homogeneous media because the permeability contrasts results in time dependent velocity fluctuations that enhances flow dynamics. To generate the permeability fields we used in-house simulator MDM (Matrix Decomposition Method) [77]. The typical Dykstra-Parson coefficient, $V_{dp} = 0.55$, was used. Concentration profiles for heterogeneous reservoir are shown in **Figure 9**. Our present results of time span for complete saturation do not match with the results reported in Refs [22, 24, 44, 78] because their findings were based on extrapolation of early periods' results. Because the flow dynamics are highly non-linear simple extrapolation they applied misrepresented the results of complete saturations.

Fig. 8. CO_2 saturation in homogeneous and heterogeneous media (inset - logarithmic permeability distributions).

Fig. 9. Concentration profiles for heterogeneous reservoir ($t = 500, 1000, 10000$, and 35000 years, clockwise).

Intensity of flow dynamics is governed by Ra 's. From **Figure 10** it is clearly observed that fingering interactions and movements are much pronounced for higher Ra 's. This is due to the fact that high velocity interacting fingers do not allow any single finger to clearly breakaway ahead of the front. On the other hand, for the smaller Ra 's slow moving fingers are isolated owing to the absence of neighboring fingers. **Figure 11** shows results of CO_2 saturations. In homogeneous reservoirs based on different geologic and thermophysical conditions the complete saturations can be reached in $5000 - 1,100,000$ years. Besides, in heterogeneous reservoirs as we can expect fast trapping time, i.e., $2000 - 900000$ years.

Fig. 10. Changes of Pe for different Ra 's in (a) homogeneous and (b) heterogeneous reservoir.

(a)

(b)

Fig. 11. Saturations' times for different Ra 's in (a) homogeneous and (b) heterogeneous reservoir.

Now to measure how much CO₂ can be immobilized through free convection, we have applied thermodynamics phase equilibrium (mass fraction of CO₂ in brine) and density calculations by following the schemes proposed by Islam and Carlson [79]. Mass fraction of CO₂ in brine at $T = 40^{\circ}\text{C}$, $P = 50$ bar, and $m = 1$ molality is 0.0209 kg CO₂/kg brine, and brine density is 1030 kg/m³. If we assume CO₂ is dissolved continuously through free convection as discussed and there is enough CO₂ injected for continuous supply at the interface the total CO₂ can be trapped is ~60 metric tons (Mt) in 2000 m³ porous volume ($V = 100 \times 100 \times 1 \times 0.2$). Based on these in the case of homogeneous medium trapping rate is 0.28 – 6 g per year in 1 m³ volume. On the other hand, in heterogeneous reservoirs 0.33 – 15 g CO₂ will convect. Considering CO₂ solubility at expected temperature (<100 °C) and pressure (<600 bar) conditions for geologic sequestration these trapping amounts may increase by at most 5 times. If the reservoir shapes are changed quantitatively average dissolutions remain almost equal [24].

5. Conclusions. In this work, density driven free convection in a 2D brine saturated isothermal reservoir, subjected to vertical concentration gradient due to dissolution of CO₂, is numerically investigated for detailed quantifications of settled amounts over the entire period of mixing process until the reservoir becomes completely saturated. To understand how the mixing progresses, time lapse of Pe is discussed. We run simulations until an average CO₂ dissolution of 99%. Ra is the governing non-dimensional parameter to study the free convection and is especially useful when applied in the context of CO₂ geologic sequestration. Because our aim is to calculate the amount of CO₂ that can be trapped through dissolution, different combinations of boundary conditions, and cases of homogeneous and heterogeneous permeability distributions are investigated. Based on the typical reservoir formations and thermophysical conditions ($V = 100 \times 100 \times 1$ m³, $\phi = 0.2$, $V_{dp} = 0.55$, $Ra = 100 - 50,000$, $T = 40$ °C, $P = 50$ bar, and $m = 1$) our findings are summarized as follows:

a) Neumann and Periodic boundary conditions show almost indifferent results of dissolutions. Dirichlet condition cannot be used to obtain the ultimate saturation. It can show saturation only up to 40%. Hence for conclusive results either Neumann or Periodic condition is to be used.

- b) Advancement rate of CO₂ fronts in term of Pe after onset of convection increases as time advances and at around, for instance, 1000 years the rate is maximum when $Ra = 1000$. After that flows slow down almost monotonically as density contrast between brine and CO₂ saturated brine decreases.
- c) In the case of homogeneous permeability distribution the reservoir will be 99% saturated after 5000 – 110,000 years of mixing process. In some heterogeneous reservoirs the saturation can be reached in 2000 – 90,000 years. Total 60 Mt CO₂ will be trapped in the model aquifer.
- d) 0.33 – 15 g CO₂ can be dissolved per year in every cubic volume of heterogeneous reservoir, whereas, in the case of a homogeneous reservoir, 0.28 – 6 g CO₂ is expected to be dissolved.

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