Numerical experiments of density driven CO₂ saturated brine migration in heterogeneous two-dimensional geologic fabric materials

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Abstract. CO₂ geological sequestration has been recognized as one of the potential solutions for reducing anthropogenic atmospheric emissions. To better estimate the storage efficiency (capacity) of subsurface geologic formations and to understand the plume migration mechanism through porous geologic formations, this study performs numerical simulations of density driven CO₂-saturated brine flow in highly resolved heterogeneous 2D fabrics. While large-scale convective currents driven by gravity have been cited as important for dissolution of CO₂ into brine, the impact of small-scale heterogeneity on the time scale of dissolution and final dissolved concentration is not well understood. Two aspects of small-scale geologic heterogeneity are examined here: 1) the correlation length of the sedimentary structures, or depositional fabric, of the material; and 2) grain size distribution (range of porosity and permeability). Twenty seven clastic facies with three different fabrics were evaluated. These materials have variable horizontal correlation lengths (1:1 to 50:1) and range from extremely well sorted upper-coarse sand to very poorly sorted upper-coarse silt. We consider density contrast between CO₂-saturated brine and initial brine as the main driving force of plume migration due to natural convection. Migration behavior from both vertical and horizontal line sources are explored. The analysis is done using conservation of mass and momentum, governed by Darcy's law. Results show that effective convective migration is possible only in a limited range of facies at the scales investigated (100's sq. cm.). Only the extremely well and well sorted coarse sands have mean permeability and porosity that exceed the critical Rayleigh number ($\approx 4\pi^2$). Results indicate that convective and dissolution processes are influenced primarily by the range in grain size (permeability), while the correlation length of the sedimentary fabric has minor effect.

Keywords: CO₂ sequestration; natural convection; darcy flow; solubility trapping

Notations.

c	concentration [mol/m ³]
D	Diffusion coefficient [m ² /s]
g	gravitational acceleration [m/s ²]

H	Reservoir thickness [m]
k	mean permeability [m ²]
L	Reservoir length [m], characteristic length
n	number of nodes
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 concentration [%]
T	temperature [°C]
t	time [s]
u	velocity [m/s]
x	distance along x-axis
z	distance along z-axis

Greek letters

β	coefficient of density increase by concentration [m ³ /mol]
ϕ	porosity [-]
μ	viscosity [kgm ⁻¹ s ⁻¹]
ρ	density [kg/m ³]
ψ	stream function [m ³ m ⁻¹ s ⁻¹]

Superscript

*	Dimensionless quantity

Subscripts

i	node in x – direction
j	node in z – direction
x	x-coordinate
z	z-coordinate

Acronyms

EWUCSa	Extreme Well Upper Coarse Sand
EWUFSa	Extreme Well Upper Fine Sand
EWUSi	Extreme Well Upper Silt
WSUCSa	Well-sorted Upper Coarse Sand
WSUFSa	Well-sorted Upper Fine Sand
WSUSi	Well-sorted Upper Silt
PSUCSa	Poorly-sorted Upper Coarse Sand
PSUFSa	Poorly-sorted Upper Fine Sand
PSUSi	Poorly-sorted Upper Silt

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1. Introduction. Injection of CO₂ into geological formations is widely regarded as a promising strategy for reducing global atmospheric CO₂ emissions (Bachu et al., 1994; Lackner, 2003; Orr Jr., 2004; Schrag, 2007). To evaluate injection scenarios and estimate CO₂ storage capacity, an accurate understanding of subsurface CO₂ migration is important (MacMinn et al., 2010). One of the primary mechanisms for stable long-term geological storage of CO₂ is the dissolution of injected CO₂ within ambient brine. The layer of dense, saturated brine forms by the processes of diffusion, dispersion and mechanical mixing during injection and, once it has reached sufficient thickness, becomes rapidly unstable to convective overturning (Ennis-King and Paterson, 2005; Islam et al., 2013; Riaz et al., 2006). Convection is expected in most sequestration sites, and controls the dissolution rate and therefore reduces the long term risk of leakage (Islam and Sun, 2015; Neufeld et al., 2010). Convective dissolution occurs in most natural CO₂ reservoirs (Sathaye et al., 2014) and is anticipated for engineered CO₂ storage. When CO₂ dissolves in the brine, the brine density increases by 2 – 3 % (Garcia, 2001; Islam and Carlson, 2012; Teng et al., 1997) and thereby, the risk of upward leakage of CO₂ is reduced upon dissolving. This results in advancing the saturated front with random fingers of dense brine extending downward away from the overlying CO₂ plume. The downward fingers are replaced by the upward migration of under-saturated brine. Through this progression the dissolution rate is greatly enhanced compared to purely diffusive transport of the dissolved CO₂. Even with the help of convective mixing, it may take thousands of years before the plume is completely trapped; therefore, it is important to have accurate estimates of the plume migration dynamics and convection behavior for long periods in the formation after injection stops (Elenius et al.,

2015; Islam and Sun, 2015). Convection enhances the rate of transport over diffusion alone and thereby increases the efficiency of the solubility trapping. In some cases natural formations may appear to be unsuitable for storage because of their low vertical permeability. However many formations have considerable permeability anisotropy where the horizontal permeability may be several times greater than the vertical permeability (Cheng et al., 2012). If the horizontal permeability is sufficiently high, they can be potentially attractive as the storage sites as long as the vertical permeability is not too low. Here we investigate advancement of saturation front through natural convection in different geologic fabrics whose formations are different based on horizontal to vertical correlation lengths and grain size distributions. Based on injection sites the saturation fronts may advance from both side boundary and top of the aquifer. We consider both scenarios in our study. Our main objective is to find out which facies show higher convection rate and therefore are suitable candidates for CO₂ sequestration.

Mathematical models and numerical simulators are essential tools for understanding our conceptualization and numerical implementation of CO₂ convection in geologic formations. The long time scale of the storage process in realistic geologic formations renders experimental investigations impractical. Although some laboratory studies (Ennis-King and Paterson, 2005; Kneafsey and Pruess, 2010; Moghaddam et al., 2012; Neufeld et al., 2011; Watson et al., 2012) have been performed to evaluate density driven natural convection processes, they cannot replicate specific cases, e.g., heterogeneous permeability distributions, the reservoir size, etc. Hence the viable alternative is numerical experiment which is the motivation behind this work.

2. The geologic fabric materials and the governing equations. The 2D geostatistical models used in the paper are built using a stochastic simulation technique that utilizes Fast Fourier Transforms (FFT) to generate heterogeneous property fields that we henceforth call “fabrics”. This program known as FFTSIM (Jennings Jr and Ward, 2000), generates log-normally distributed geological property fields with the desired mean, variance and correlation lengths. While the correlation lengths of the generated fabrics are controlled by user input, the grain sizes of the fabrics are populated based on a variety of rock/facies types with defined median grain size and sorting (grain size histogram), characteristically described by geologists (Beard and Weyl, 1973; Folk and Ward, 1957). The nine facies used in the models are displayed in **Fig. 1**. The rows have fixed sorting (standard deviation) while the columns have fixed median grain size values. Moving down the rows, sorting becomes worse, indicating wider grain size distributions and greater heterogeneity.

Three horizontal to vertical correlation lengths are generated stochastically (50:1, 10:1 and 1:1) and properties (i.e. porosity, permeability) are assigned based on the nine different facies, resulting in 27 models representing different facies (grain size distributions) and fabrics (horizontal correlation lengths). As the horizontal correlation length increases the level of anisotropy in the vertical direction increases.

The porosities of the facies used in the model are those experimentally determined by Beard and Weyl (Beard and Weyl, 1973) for wet packed sand with similar grain size properties. With the porosity, and the mean and standard deviation of the facies available, the corresponding permeability field of the model is generated using the well-established Berg's (Nelson, 1994) porosity-permeability correlation:

$$k = 5.1 \times 10^{-6} \times \phi^{5.1} \exp^{-1.385 p} D^2 \quad (1)$$

where k is the permeability in Darcy, ϕ is the percent porosity, D is the median diameter of the grains in millimeters and p is the sorting parameter in phi scale (phi is minus of log to the base 2 of the size in mm).

The model reservoir of geologic sandstone in this study is a 2D domain saturated with brine. The reservoir height and length are H and L , respectively. As mentioned we have investigated how dense CO_2 saturated interface spreads from lateral and vertical line sources, separately. The cases and corresponding boundary conditions are shown in **Fig. 2**. It is noteworthy that the interface is relatively sharp and is not deflected by the density fingering in the underlying brine. The condition is maintained by the continuous phase equilibrium establishment. The pressure change due to dissolution is negligible. The brine is initially quiescent. The presence of a capillary transition zone between the gas and the brine phase is disregarded. Hence only the liquid phase is modeled and the presence of the gas phase is represented by a boundary condition for the liquid phase. The Boussinesq approximation and Darcy flow model are assumed valid. The velocity-induced dispersion effect is ignored. Because this is a natural convection problem Ra is low enough to expect only the laminar flow. Isothermal condition is kept valid in the model aquifer. Therefore energy balance is not taken into consideration. Density contrast due to concentration difference is the only driving force to impose natural convection. The governing equations are as follows:

(a) Continuity equation,

$$\phi \frac{\partial \rho}{\partial t} + \frac{\partial(\rho u_x)}{\partial x} + \frac{\partial(\rho u_z)}{\partial z} = 0. \quad (2)$$

(b) Darcy's law,

$$u_x = -\frac{k_x}{\mu} \frac{\partial p}{\partial x}, \quad (3)$$

$$u_z = -\frac{k_z}{\mu} \left(\frac{\partial p}{\partial z} - \rho g \right). \quad (4)$$

(c) Mass transport equation,

$$\phi \frac{\partial c}{\partial t} + u_x \frac{\partial c}{\partial x} + u_z \frac{\partial c}{\partial z} = \phi D \left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial z^2} \right). \quad (5)$$

A simplified equation-of-state below is used to account the density change due to dissolved CO₂

$$\rho = \rho_0 [1 + \beta(c - c_0)], \quad (6)$$

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

After differentiation we obtain

$$\frac{\partial \rho}{\partial x} = \rho_0 \beta \frac{\partial c}{\partial x}. \quad (7)$$

Eqs. (3) and (4) are cross differentiated for eliminating the pressure term. We get

$$\frac{1}{k_z} \frac{\partial u_z}{\partial x} - \frac{1}{k_x} \frac{\partial u_x}{\partial z} = \frac{g \rho_0 \beta}{\mu} \frac{\partial c}{\partial x}. \quad (8)$$

2.1 Dimensionless form of the equations:

The dimensionless variables are defined by,

$$\begin{aligned}
x^* &= \frac{x}{H}, \quad z^* = \frac{z}{H}, \quad u_x^* = \frac{Hu_x}{\phi D}, \quad u_z^* = \frac{Hu_z}{\phi D}, \quad k_x^* = \frac{k_x}{k}, \quad k_z^* = \frac{k_z}{k}, \quad \psi^* = \frac{\psi}{\phi D}, \\
t^* &= \frac{Dt}{H^2}, \quad c^* = \frac{c - c_r}{c_0 - c_r}, \quad u_x^* = -\frac{\partial \psi^*}{\partial z^*}, \quad u_z^* = \frac{\partial \psi^*}{\partial x^*}, \quad Ra = \frac{\Delta \rho g k H}{\phi D \mu}, \quad \text{and } \Delta \rho = \rho_0 \beta (c - c_0).
\end{aligned} \tag{9}$$

As we have applied Boussinesq approximation the final forms of dimensionless equations thus are appeared as (after dropping *)

$$k_z \frac{\partial^2 \psi}{\partial x^2} + k_x \frac{\partial^2 \psi}{\partial z^2} = Ra \frac{\partial c}{\partial x}, \tag{10}$$

$$\frac{\partial c}{\partial t} + u_x \frac{\partial c}{\partial x} + u_z \frac{\partial c}{\partial z} = \frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial z^2}. \tag{11}$$

The coupled Eqs. 10 and 11 have been solved to obtain ψ and c .

2.2 Initial and boundary conditions:

The initial conditions ($t = 0$) are

$$\psi = 0, \quad c = 0. \tag{12}$$

The boundary conditions are presented in **Fig. 2**.

3. Numerical methods. As we know numerical simulation of density-driven transport problem is very sensitive to discretization errors. The following CFL criteria, hence are fulfilled for the stability of the transport equations given by

$$\frac{u \Delta t}{\phi \Delta x} \leq \frac{P_E}{2}, \tag{13}$$

where $P_E = \frac{u \Delta x}{D \phi}$. The simulation was performed with 128×128 uniform rectangular grid cells with

aspect ratio of unity, and time step (Δt) was taken as 10^{-6} (equivalent to ~ 111 seconds based on $H = 1$ m) to meet above conditions. We have applied a sequential solution procedure to solve the elliptic Poisson equation (10) and the solute transport equation (11). Eq. (10) has been solved by Point Gauss-Sidel iterative method with convergence criteria of $\sum_{s=1}^{\infty} \left(\frac{\psi^{s+1} - \psi^s}{\psi^s} \right) < 10^{-6}$ where the superscript s denotes iteration step number. The Alternating Direction Implicit (ADI) method is used to solve Eq. (11) where

the convection terms are discretized using the upwind differencing and the diffusion terms are discretized using the central differencing. The developed Matlab R2013b (www.mathworks.com) code was validated against the benchmark solutions (Hassanzadeh et al., 2005; Islam and Sun, 2015; Simpson and Clement, 2003) and the results were in satisfactory agreement independent of mesh sizes. The interface boundary concentration is sinusoidally perturbed shown in Eq. (14) in order to observe the non-linear behavior, i.e., the fingering behavior.

$$c = 1 + 0.01 \sin(2\pi nx). \quad (14)$$

The wave number (n) of 24 is used. It should be noted that the long term behavior is not dependent on the initial perturbation. More so, the growth of the perturbations is a weak function of the wavelength. Pore-level perturbations and thermo-mechanical fluctuations cause the perturbation to start the finger like plumes in reality (Gunn and Krantz, 1980; Landau and Lifshitz, 1969). However, pore-level instabilities (Yortsos, 1997) is ignored here for simplicity. The effect of velocity-induced dispersion is not considered. Additionally other mechanisms like, precipitation and geochemical reactions are not accounted.

4. Results and Discussions. The resultant permeability distributions of facies derived from Eq. (1) are shown in **Fig. 3**. Colorbars are in absolute scale, mD. The level of heterogeneity varies as low as by 2 times in EWUSCSa to maximum 7 orders of magnitude in PSUCSa. Because of extreme variations the permeability contrasts are not visible in absolute scales for the case of poorly sorted facies. Hence to clarify the distributions in logarithmic scales are exhibited for Fabrics 1 and 3 in **Fig. 4**. The least permeable formation is EWUSi of which mean permeability 2.5 mD. As a whole, in the case of well and poorly sorted facies the heterogeneity ranges are very wide. However, interestingly, the high permeable regions are quite few, discrete. The permeability variations are far away than normal distributions. For instance, in PSUCSa of Fabric 2 mean permeability is 4400 mD where the median is only 86 mD. In PSUSi mean and median permeability are 340 and 0.67 mD, respectively.

First we investigate propagation of CO₂ saturated front from lateral boundary as shown in Fig. 2a. The saturation (or, average concentration) spread around the fabric domain with respect to time is calculated as follows

$$\bar{c} = \frac{\int_0^x \int_0^z c(x, z, t) dx dz}{\int_0^1 \int_0^1 dx dz}. \quad (15)$$

Fig. 5 shows average concentrations of CO₂ in different facies of fabric 1. EWUCSa facies renders highest saturation among all other configurations where EWUFSa shows the minimum. The permeability of EWUSi, WSUSi, and all poorly sorted facies are so low that the resultant *Ra*'s are smaller than the critical value ($\approx 4\pi^2$). Hence for these formations we have not observed any free convection in the simulations, and we anticipate similar behavior in natural settings with similar facies. Meckel et al., (2015) have presented such specimens in their studies of CO₂ invasion-percolation. The convection is purely diffusion like and consequently is extremely slow. They have not been presented in comparisons. The results are consistent per permeability distributions of the facies. As mentioned, although in some very discrete points the permeability are high in WSUCSa and PSUCSa mostly they are packed with extremely low permeable formations. The slow diffusion of these cases can be enhanced through immobilization and forced convection. We have found following descending order after sorting the suitability of sequestration based on the amount of CO₂ can be trapped through dissolution and natural convection processes.

$$\text{EWUCSa} \rightarrow \text{WSUCSa} \rightarrow \text{PSUCSa} \rightarrow \text{WSUFSa} \rightarrow \text{EWUFSa} \quad (16)$$

The coarse formations are relatively suitable for sequestration than the fine formations, and obviously the silts, because of higher permeability which allow CO₂ fronts to find favorable paths for advancing. Especially in silt formations the plumes are arrested from the very beginning resulting in no free convection. To observe how CO₂ plumes advance laterally is shown in **Figs. 6 and 7**. The axes are in non-dimensional node numbers. As initial perturbations of the wavelength of 24 were induced convection cells evolve in 23 segments and they merge from tips in order to reach mechanical equilibrium locally. Convection becomes stronger with time. Some fingers grow faster than the others and the finger wavelength amplifies due to random but stronger nonlinear interactions. There is some tenacity of the initial behavior and the convection pattern experienced persists for few moments before the number of fingers starts to decrease exhibiting intrinsic properties (Farajzadeh et al., 2011; Islam et al., 2014). Many of the fingers seem to undergo strong interactions while others in the process of disappearing. The convection cells are surrounded by diffusive shear layers which ultimately act as the feeding sites. For increased heterogeneity the convection cells get more dynamicity and thus CO₂ dissolution in the facies becomes faster. The flow patterns are almost same in the fabrics of same formations (EWUCSa) because their permeability variations are almost same except the distributions vary by correlations lengths.

Though very minor, the effect of correlations lengths will be shown in later part of this discussion. **Figs. 8 and 9** show results of fabric 2 and 3, respectively, indicating similar observations experienced in fabric 1.

When the CO₂ saturation is prescribed to spread from the top boundary, the results presented in **Fig. 10** seem similar to previous cases of side boundary. However, because of gravity effect quantitatively saturations are higher by few times. The results are shown for EWUCSa and WSUCSa formations as they are better candidates. In reality, CO₂ plumes may flow from both side and top boundaries based on injection sites and how CO₂ is flowing after injection is ceased (MacMinn et al., 2010). Irrespective of boundary locations the advancement of plume is mainly controlled by the permeability provided other reservoir and thermo-physical properties of CO₂-brine remain same. The faster vertical motion can enhance the stable storage of dissolved CO₂ in deeper parts of the formation. This vertical spreading involves more contact area between dissolved CO₂ and the host rock.

Fig. 11a shows results of concentrations for different correlation lengths in EWUCSa. It is clearly revealed that for the same level of heterogeneity the correlation length's effect is not significant. The formation with higher correlation length can be little more saturated after long time. On the other hand the effect is quite obvious with different permeability variations at the same correlation length. These calculations are shown in **Fig. 11b**. Actually characterization of the saturated CO₂ transport due to natural convection is highly dependent on permeability variance with small influence of correlation length. It is important to note here that the flow regimes (fingering, channeling, dispersive) are largely reliant on both correlation length and heterogeneity simultaneously (Farajzadeh et al., 2011), however in our case we have presented results in term of mass transport where correlation lengths are found to be not influential. This is because; when plumes reach to any low permeable layers they spread laterally and advance downward as soon as get favorable high permable paths. On the other hand, in the case of lateral flow whenever the plumes are barred by horizontal layers immediately move through vertical favorable paths.

5. Conclusions. Simulations of natural geologic fabric materials with known properties (grain size distribution) and correlation length (natural deposition) were performed to study density driven free convection of CO₂ saturated brine. We aimed to sort out the materials to find out the trapping capacity in term of mass deposition. The side and top saturated boundary, separately, were considered for predictions. Both cases are representative of how CO₂ may migrate in saline aquifers. From the results obtained the following conclusions can be drawn:

- (a) The coarse sand materials are better candidates for increased CO₂ dissolution while the silts are the least preferable because of their low permeability. The facies can be ranked with respect to dissolution as EWUCSa → WSUCSa → PSUCSa → WSUFSa → EWUFSa. The others even do not meet critical conditions to drive any convection. In these cases the plumes are arrested from very beginning.
- (b) The vertical line source cases result in higher saturation than the lateral flows because of gravity effect.
- (c) In the same fabric with specific correlation length the convection rates can vary significantly in different facies because of wide range in grain sizes. This reveals that the heterogeneity has very strong effect in CO₂ dissolution.
- (d) In different facies with the same heterogeneity results do not vary much. This is due to the weak effect of correlation lengths. Dissolution magnitudes and rates are loosely dependent on the correlation length of sedimentary fabrics.

Acknowledgement. The authors acknowledge Texas Advanced Computing Center to provide SUs (service units) required to run simulations.

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Figures

Fig. 1. The 9 selected facies (highlighted with red squares) from a library of 54 sandstone facies used in this work. These facies are characteristic of a wide range of depositional processes and environments. Sorting is given in terms of Trask coefficient S_o and on the phi (ϕ) scale (Folk and Ward, 1957). Median grain diameters at the bottom are in millimeters. Wet packed porosities for these materials range from 43.1% (upper coarse extremely well sorted sand; upper left) to 32.6% (lower very fine sand; lower right) (Beard and Weyl, 1973, Table 1). The last column (Upper Coarse Silt) is not a part of their table; we assume porosities to be in the similar range as that of lower fine sand.

Fig. 2. Model geologic fabric assuming saturated boundary at (a) left and (b) top. Corresponding boundary conditions are shown.

(a)

(b)

(c)

Fig. 3. Permeability distributions with a ratio of horizontal-to-vertical correlation length of (a) Fabric 1, 50:1; (b) Fabric 2, 10:1; and (c) Fabric 3, 1:1. Each fabric resemblances to highlighted red squares in Fig. 1. The three rows from left to right represent EWUCSa, EWUFSa, EWUSi; WSUCSa, WSUFSa, WSUSi; and PSUCSa, PSUFSa, and PSUSi.

Fig. 4. $\log_{10} k$ of poorly sorted facies of (a) Fabric 1 and (b) Fabric 3.

Fig. 5. CO_2 concentrations over time for Fabric 1.

Fig 6. Concentration profiles of EWUCSa of Fabric 1 at (a) 1 and (b) 12 months.

Fig 7. Concentration profiles of EWUCSa of Fabric 3 at (a) 1 and (b) 12 months.

Fig. 8. CO₂ concentrations over time for Fabric 2.

Fig. 9. CO₂ concentrations over time for Fabric 3.

Fig. 10. CO₂ concentrations over time for Fabric 1 (saturated boundary at the top).

Fig. 11. CO₂ concentrations over time (a) for EWUCSa in different correlation lengths (b) for two different facies at the same correlation length of 10:1. Note that the effect of facies (grain size distribution) has a much greater influence than horizontal correlation length of fabric (permeability).