DENSITY DRIVEN (INCLUDING GEOTHERMAL EFFECT) NATURAL CONVECTION OF CARBON DIOXIDE IN BRINE SATURATED POROUS MEDIA IN THE CONTEXT OF GEOLOGICAL SEQUESTRATION

*Akand W. ISLAM, **Muhammad A. R. SHARIF, *Eric S. CARLSON

*Department of Chemical & Biological Engineering
**Department of Aerospace Engineering & Mechanics
The University of Alabama
Tuscaloosa, AL 35487, USA
e-mail: awislam@crimson.ua.edu

ABSTRACT
Double diffusive natural convection of carbon dioxide in two-dimensional cavities filled with brine saturated porous media is numerically investigated in this study. Vertical gradients of carbon dioxide concentration and temperature are imposed across the height of the cavity. The objective is to understand the dissolution of carbon dioxide through natural convection process over long period of time after sequestration into subsurface porous media aquifer. The problem parameters are the solutal Rayleigh number \( 100 \leq Ra_s \leq 10000 \), the buoyancy ratio \( 2 \leq N \leq 100 \), the thermal Rayleigh number \( 2 \leq Ra_T \leq 100 \), the cavity aspect ratio \( 0.5 \leq A \leq 2 \), and a fixed Lewis number \( Le = 301 \). It is found that the \( \text{CO}_2 \) plumes move faster when \( Ra_s \) is increased, however slow down with decreasing \( N \). For every simulation run, the average \( \text{CO}_2 \) dissolution \( \bar{S} = \sum_{i,j}^{n} \sum_{i,j}^{n} C_{i,j} \) in the reservoir is computed. At early stage \( (\leq 10 \text{ years}) \) of the convection process, the \( \text{CO}_2 \) dissolution are same for all cases studied. After 500 years the dissolution is found to be around 0.63 for \( N = 100 \), and around 0.47 for \( N = 2 \), respectively. After 2000 years the dissolution rate is extremely slow. When the reservoir aspect ratio \( (A) \) is changed, the dissolution rate changes slightly. The rate is slightly higher in laterally wide reservoir, which makes it better candidate than the deeper aquifer from the context of \( \text{CO}_2 \) sequestration.

INTRODUCTION
In order to control carbon emissions, the use of technologies to capture and store \( \text{CO}_2 \) has rapidly emerged as an important physically and economically viable method these days. Various \( \text{CO}_2 \) storage methods in geological formations, such as depleted oil and gas fields, un-mineable coal seams, saline-filled basalt formations, etc., have been suggested. This process promises to reduce the cost of achieving considerable reductions in \( \text{CO}_2 \) emissions over the next few decades (Hassanzadeh et al., 2005a). Geological storage in underground saline formation involves injecting supercritical \( \text{CO}_2 \) at high pressure into a saline aquifer capped by a rock formation. Following injection, \( \text{CO}_2 \) is accumulated between the cap and aquifer surface. Eventually, the \( \text{CO}_2 \) is trapped by two different mechanisms, namely; capillary trapping and solubility trapping. In capillary trapping, part of the \( \text{CO}_2 \) rises through porous rock formation above due to buoyancy and capillary forces and gets trapped into the rock pores. On the other hand, at the interface between the \( \text{CO}_2 \)-rich
phase and brine (Farajzadeh et al., 2009), dissolution of CO$_2$ into brine starts by molecular diffusion increasing the brine density (Islam and Carlson, 2012) (by about 1%) on the aquifer top surface which then sinks down into the brine by natural convection due to solute gradient. This phenomenon is termed as solubility trapping. Another destabilizing agent is the naturally occurring geothermal temperature gradient (typically ~3 °C/100 meter depth), which induces some sort of upward natural convection of the brine. The interaction of these two opposing processes, termed as double diffusive transport, determines the resultant rate of dissolution/deposition of the CO$_2$ in the brine. The geothermal gradient is partially compensated by the geo-pressure gradient (Lindeberg and Wesselberg, 1997) (normally ~10 bar/100 meter depth). The convective mixing enhances dissolution of CO$_2$ through continuously removing CO$_2$-rich brine from the top layer and bringing under-saturated brine into contact with the downward advancing CO$_2$ plume. For the design, operation, and maintenance of such a geologic CO$_2$ storage facility, it is very important to quantify the rate of dissolution and understand the transport mechanism. The time scale of the solubility trapping is very large ranging hundreds to thousands of years during which the high pressure free phase CO$_2$, accumulated between the aquifer free surface and the top rock formation may leak out through rock fractures (Hassanzadeh et al., 2005b). The chance of leaking reduces when a significant amount of CO$_2$ dissolves into the brine.

Comprehensive investigations have been carried out on double-diffusive convection where heat and solute are the diffusive components (Oldenburg and Pruess, 1998; Stern, 1975). An extensive work has been done in the context of porous media, especially related to the environmental problems and the transport of contaminants (Cooper et al., 1997; Green, 1984; Mojtabi et al., 2005; Poulikakos, 1986). Studies investigating stability analysis for the onset time for convection, the preferred wavelength for the growth of convective fingers, growth rates, and solutal and thermal effects have also been published (Bhaduria, 2006; Javaheri et al., 2010; Poulikakos, 1986; Sodha and Kumar, 1985). The extensive time scale of the storage process operation renders the experimental investigation of the process impractical. The viable alternative is to study the double diffusive process numerically, which is the motivation behind this work. The use of modeling and simulation to make predictions on the timescale is obviously impossible to validate, since even in a field operation one cannot history match more than a small amount of the relevant time period (Ennis-King and Paterson, 2003).

In this paper the double-diffusive convection of CO$_2$ in brine under vertical thermal and solutal gradients is numerically studied considering reservoir conditions suitable to geologic sequestration. Realistic simulation inputs, obtained through extensive literature review, are provided for the computations. Results are presented graphically in terms of the propagation of the CO$_2$ front through the aquifer with time for various cavity configurations ($0.5 \leq A \leq 2$) and for a range of the solutal Rayleigh number ($100 \leq Ra_s \leq 10000$) and thermal Rayleigh number ($2 \leq Ra_T \leq 100$). The effects of variation of these geometric and hydrodynamic parameters on the CO$_2$ front propagation and dissolution are analyzed and evaluated.

**DESCRIPTION OF THE PROBLEM AND GOVERNING EQUATIONS**

The geometry under consideration is a two-dimensional rectangular cavity reservoir, sketched in **Figure 1**, filled with porous medium saturated with brine (H$_2$O+NaCl), with a height $H$ and length $L$. The permeability of the porous medium is $\varphi$. Initially the fluid is at rest and there is no
CO₂ dissolved. Boundary conditions are no fluid flow across all boundaries and no solute fluxes across lateral and bottom boundaries at all time. Also there is no heat flux across the lateral boundaries while the top and bottom boundaries are maintained at isothermal cold and hot temperatures, respectively. We assume that CO₂-liquid interface is relatively sharp and fixed at the top boundary; meaning pressure change due to dissolution is negligible. This approximation is reasonable below a depth of 1000 m which is the usual case of geological sequestration (Ennis-King and Paterson, 2005). The brine is initially quiescent and the medium is homogeneous in terms of porosity and permeability. The presence of a capillary transition zone between the gas and the brine phase is disregarded. Therefore only the liquid phase is modeled and the presence of the gas phase at the top is represented by a boundary condition. The Boussinesq approximation and Darcy flow model are assumed valid. Moreover, we assume that velocity-based dispersion and capillary effects are negligible and that geochemical reactions are not occurred. We only expect a laminar flow regime since Rayleigh number is low. Boundary conditions for the temperature are opposite to concentration, because the geo-temperature at the bottom of the reservoir is higher than that at the top, the actual temperature difference varies from place to place. The density gradient owing to concentration and temperature variations are the source of natural convection here. For such a system, the governing equations of flow and concentration field are:

(a) Continuity equation
\[
\frac{\partial \rho}{\partial t} + \frac{\partial (\rho u_x)}{\partial x} + \frac{\partial (\rho u_z)}{\partial z} = 0
\]  
(1)

(b) Darcy’s law
\[
u_x = -\frac{k}{\mu} \frac{\partial p}{\partial x},
\]
(2)
\[
u_z = -\frac{k}{\mu} \left( \frac{\partial p}{\partial z} - \rho g \right).
\]
(3)

(c) Concentration transport equation
\[
\frac{\partial \rho c}{\partial t} + u_x \frac{\partial \rho c}{\partial x} + u_z \frac{\partial \rho c}{\partial z} = \varphi D \left( \frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial z^2} \right)
\]
(4)

(d) Energy equation:
\[
\rho_0 C_p \left( \frac{\partial T}{\partial t} + u_x \frac{\partial T}{\partial x} + u_z \frac{\partial T}{\partial z} \right) = \kappa \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial z^2} \right)
\]
(5)

For small density variations due to temperature and concentration changes at constant pressure, the brine density is a linear function of temperature and concentration of solute as given by
\[
\rho = \rho_0 \left[ 1 + \beta_c (c - c_0) + \beta_T (T - T_0) \right]
\]
(6)
where
\[
\beta_c = \frac{1}{\rho_0} \left[ \frac{\partial \rho}{\partial c} \right] \quad \text{and} \quad \beta_T = \frac{1}{\rho_0} \left[ \frac{\partial \rho}{\partial T} \right]
\]
(7)

and we obtain
\[
\frac{\partial \rho}{\partial x} = \rho_0 \left[ \beta_c \frac{\partial c}{\partial x} + \beta_T \frac{\partial T}{\partial x} \right]
\]
(8)

After eliminating the pressure by cross-differentiation of equations (2) and (3) we get
\[
\frac{\partial u_x}{\partial x} - \frac{\partial u_z}{\partial z} = kg \rho_0 \left[ \beta_c \frac{\partial c}{\partial x} + \beta_T \frac{\partial T}{\partial x} \right]
\]
(9)

The equations to be solved are Eqs. (1), (4), (5), and (9) to obtain \(u_x, u_z, c\) and \(T\).

**Dimensionless form of the equations**

Considering the cavity height, \( H \), as the characteristic length and \( \varphi D / H \) as the characteristic velocity following dimensionless variables are defined,
After applying the Boussinesq approximation the final forms of dimensionless equations are

\[
\frac{\partial^2 \psi^*}{\partial x^*} + \frac{\partial^2 \psi^*}{\partial z^*} = Ra_t \left( \frac{\partial c^*}{\partial x^*} - \frac{1}{N} \frac{\partial T^*}{\partial x^*} \right)
\]

(10)

\[
\frac{\partial c^*}{\partial t^*} + u^*_x \frac{\partial c^*}{\partial x^*} + u^*_z \frac{\partial c^*}{\partial z^*} = \frac{\partial^2 c^*}{\partial x^*^2} + \frac{\partial^2 c^*}{\partial z^*^2}.
\]

(11)

\[
\frac{\partial T^*}{\partial t^*} + \rho u^*_x \frac{\partial T^*}{\partial x^*} + \rho u^*_z \frac{\partial T^*}{\partial z^*} = \frac{1}{Pr} \left( \frac{\partial^2 T^*}{\partial x^*^2} + \frac{\partial^2 T^*}{\partial z^*^2} \right)
\]

(12)

where \( N = -\frac{\beta c \Delta c}{\beta c \Delta T} \) is the buoyancy ratio and \( Pr = \frac{\rho c_p D}{\kappa} \).

**Boundary and initial conditions**

The initial condition is,

\[ c^* = 0, T^* = 0 \]  \( \text{at } t^* = 0 \) \( \psi^* = 0 \).  \( \text{at } t^* = 0, c^* = 0, T^* = 0 \).  \( \text{at } t^* = 0 \)  \( \psi^* = 0 \).

(13)

The boundary conditions are shown in Figure 1.

**Figure 1**: Schematic diagram of our hypothetical reservoir model

---

**NUMERICAL METHOD**

Numerical simulation of density-driven transport problem is very sensitive to discretization errors. The following criteria must be fulfilled for the stability of the transport equations given by

\[
\frac{u^* \Delta x^*}{\varphi \Delta x^*} \leq \frac{P_E}{2}
\]

(14)

where \( P_E = \frac{u^* \Delta x^*}{D \varphi} \). The simulation was performed with \( 81 \times 81 \) uniform rectangular grid cells for the cavity with aspect ratio of unity, and time step \( (\Delta t^*) \) was taken as \( 10^{-6} \) (equivalent to \(~29\) days based on \( H = 100 \) m) to meet above conditions. A sequential solution procedure was used to solve the elliptic Poisson equation (10) and the concentration and temperature transport equations (11) and (12). Equation (10) is solved by Point Gauss-Sidel iterative method with convergence criteria of

\[
\sum \left( \psi_{i+1}^* - \psi_i^* \right) < 10^{-6}
\]

where the superscript \( i \) denotes iteration step number.

Equations (11) and (12) are solved using the Alternating Direction Implicit (ADI) method where the convection terms are discretized using the upwind differencing and the diffusion terms are discretized using the central differencing. The developed code prediction for natural convection in a cavity was checked against the literature benchmark solutions (Hassanzadeh et al., 2005a; Simpson and Clement, 2003) and the results are in satisfactory agreement with the published simulations independent of the number of grid cells. It is required that the interface concentration be perturbed in order to observe the downward propagation of the finger like concentration fronts. Therefore a sinusoidal perturbation is used on the top interface in the form

\[
c^* \left( x^*, z^* = 0, t^* = 0 \right) = 1 + 0.01 \sin(2\pi x^*)
\]

(15)
where the wave number \((n)\) of 24 is used. Basically the long term behavior is not dependent on the initial perturbation (Farajzadeh et al. 2007). Moreover, 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). Pore-level instabilities (Yortsos et al., 1997) is ignored here, however.

The important assumptions in this study are the homogeneity and isotropy of the porous medium. The effect of velocity-induced dispersion is ignored and the flow field as single-phase is considered. Additionally other mechanisms like, precipitation and geochemical reactions are not accounted for simplicity.

**RESULTS AND DISCUSSIONS**

The double diffusion is imposed here through the buoyancy ratio \((N)\) which was varied between 2 and 100. This represents vast differences of geothermal gradients. The geothermal gradient is not same everywhere; for instance, very high gradient can be observed as 20 °C/100 m at the Mid-Atlantic Rift, whereas in Iceland there is almost no gradient (~0 °C/100 m) (Lerner and Lerner, 2003). The direction of the flow due to thermal buoyancy forces is anticlockwise because temperature at the bottom of the reservoir is always higher than at the top. On the other hand, the flow owing to concentration gradient is clockwise, opposing the thermal flow. The solutal Rayleigh number, \(Ra_s\), is varied from 100 – 10,000. The porosity \((\varphi)\) of a typical reservoir (Oldenburg and Pruess, 1998), of 0.30, and recently reported (Newell et al., 2011) diffusion coefficient \((D)\) of brine filled porous media, of \(4 \times 10^{-9}\) m²/s, respectively, are taken for calculations. In order to calculate the Prandtl number, \(Pr\), in the energy equation, the required inputs are collected from Sharqawy et al (Sharqawy et al., 2010). \(Pr\) is calculated to be 0.0062. To understand the natural convection of different reservoir shapes, the aspect ratio \((A)\) is varied 0.5, 1, and 2 representing deep, square, and shallow reservoir configurations, respectively. The thermal diffusivity \((\alpha)\) is considered constant assigning the value \(3.7 \times 10^{-7}\) m²/s (Javaheri et al., 2010). Hence, the Lewis number, \(Le\), is fixed at 310. To discuss the results in terms of a combination of the solutal Rayleigh number, \(Ra_s\), and the buoyancy ratio, \(N\), the equivalent Rayleigh number, \(Ra_e\), expressed as

\[
Ra_e = Ra_s \left(1 + \frac{1}{N}\right)
\]

is introduced.

**Effects of \(Ra_s\) and \(N\)**

First, various scenarios of varying \(Ra_e\) from low (100) to relatively high (10,000) and \(N\) from 2 to 100 for the case of \(A = 1\) (square shape) are discussed. Figure 2 shows the concentration distribution for \(Ra_s = 100\) and \(N = 100\) where only the top side of the vertical domain is zoomed out. The corresponding \(Ra_e\) for this case is 99. By definition [Equation (16)] \(Ra_e\) does not change significantly unless \(N\) is low and close to 1. Equation (16) also implies that when the buoyancy ratio is higher, the thermal effect on CO₂ dissolution is relatively minor. When \(Ra_e\) is low (such as 99), even though the induced perturbations at the top interface initiate very tiny convection cells, they cannot survive as the time marches. For low \(Ra_e\) the dissolution is completely diffusion dominated and therefore propagation of CO₂ concentration front into brine is extremely slow. This is unfavorable because in such case CO₂ will have to be trapped over the aquifer for a very long time (thousands of years) and may caveat leakage through any fracture in the top rock formations. Average dissolution (or
concentration) of CO$_2$ in the model reservoir is defined as

$$
\bar{S} = \frac{\sum_i \sum_j c_{i,j}}{n_i \times n_j}
$$

(18)

After about 4 years the average dissolution is found to be 0.016 while after 100 years that is 0.046. After 500 years the average dissolution of CO$_2$ in the aquifer will be only 0.094. During early periods, CO$_2$ dissolves in the brine slowly and as time passes the diffusion dominated dissolution rate is slightly enhanced.

Figure 2: Concentration profiles for $Ra_s = 100, N = 100$, at (a) $t = 4$, (b) $t = 100$, and (c) $t = 500$ years.

When $N$ is decreased to 50, the $Ra_e$ becomes 98 and correspondingly produces almost the same concentration distribution for $Ra_e = 99$ as shown in Figure 2. When the thermal buoyancy effect is increased by substantially decreasing $N (=2)$, the $Ra_e$ decreases to 50. At this low equivalent Rayleigh number, CO$_2$ dissolution rate is extremely slow (not shown).

The CO$_2$ propagation over the time is shown in Figure 3 when $Ra_s$ is increased to 1,000 with $N = 100$ producing $Ra_e$ of 990. In this case we see that, even up to 100 years the concentration front propagation is primarily diffusive and therefore the average dissolution reaches to only 0.05. However, after long time, the convective mixing becomes traceable. Figure 3c shows CO$_2$ concentration fingers after 500 years and some of them reach around half of height of the reservoir producing average dissolution of 0.15. Since initial perturbations of the wavelength of 24 were introduced, convection cells start to evolve in 23 segments and they merge together as the fluid convection becomes stronger. Nevertheless, some fingers grow faster than the others and the finger wavelength increases due to random but stronger
nonlinear interactions. It is also observed that the CO$_2$ finger front movement is relatively faster close to the sidewalls than the central region. Decreasing the value of $N$ to 50 exhibits marginal difference ($Ra_e = 980$), and is not shown separately. Further decrease of $N$ to 2 decreases the equivalent Rayleigh number to 500 which basically shows only diffusion even after 500 years (not shown here). Although the interface is perturbed, the imposed initial disturbances are damped and the CO$_2$ front advance as diffusion like manner.

Figure 3: Concentration profiles for $Ra_s = 1000$, $N = 100$, at (a) $t = 20$, (b) $t = 100$, and (c) $t = 500$ years.

For the case of $Ra_s = 10,000$ and $N = 100$ ($Ra_e = 9,900$), Figure 4 shows the CO$_2$ concentration distribution at several time periods. Here the $Ra_e$ is high enough to instigate natural convection even at early periods. Small convection cells are clearly seen in Figure 4a after the simulation is run up to 4 years. At this time, the average concentration is 0.02. The CO$_2$ concentration is distributed more evenly as time elapses. The 23 fingers generated initially reduce to 8 after 10 years with a corresponding dissolution of 0.03. The fingers keep moving downward dissolving more CO$_2$ in brine ($\bar{S} = 0.07$ after 20 years). Many of the fingers are observed to undergo strong interactions while others are in the process of being faded. When the CO$_2$ front first reaches to the bottom of the reservoir (after 68 years) only one isolated big central plume survives, while two other fingers on both sides of the central plume, originating from single feeding site on top, are still in competition. Dynamics of the fluid movements becomes irregular. As the time passes new feeding sites appear and the older ones are banished. In other regions, multiple fingers can attach to a single feeding site, a process which is governed by diffusion. The non-linear dynamics then
selects one over the others as the favorable flow path (Riaz et al., 2006). This is noteworthy that the merging process starts at the stem sides rather than at the tip of the fingers and from there they spread to the rest of the system.

Figure 4: Concentration profiles for $Ra_s = 10000$, $N = 100$, at (a) $t = 4$, (b) $t = 10$, (c) $t = 20$, (d) $t = 68$, (e) $t = 100$, and (f) $t = 500$ years.
The larger fingers travel with less interactions with the neighboring fingers. Moreover, large fingers are connected to the thin diffusive shear layer at discrete locations serving as the feeding sites of high-density fluid for the fingers. The animation of the transient advancement of the plumes at $Ra_e = 9,900$ may be watched at the website (http://www.youtube.com/watch?v=oyUNj7HEaro). Though CO$_2$ reaches at the bottom relatively early, the saturation is below 0.20. Figure 4f shows simulation results for 500 years. The later stage behavior of the convection process cannot be ascertained from the results of early stage and the prediction of the rate of dissolution is very complex. Porosity, permeability, density difference, temperature difference, etc. are the factors which all are retained in the Rayleigh number and therefore the complexity in the flow behavior is strongly dependent on this Rayleigh number (both solutal and thermal). It is already apprehended that the concentration front spreads faster for larger equivalent Rayleigh number. Average dissolution of CO$_2$ in the reservoir after 100 and 500 years is 0.21 and 0.62, respectively for $N = 100$. When $N$ is reduced to 2, these corresponding dissipations are 0.16 and 0.47, respectively. The concentration contours of this case ($N = 2$) are shown in Figure 5. At early stages CO$_2$ dissolutions are same (~0.02) for both $N = 100$ and 2. This is because at the early stages the transport process is mostly diffusion dominated. As mentioned earlier, the CO$_2$ front reaches the bottom of the aquifer after 68 years when $N = 100$ while the front advances little more than half of the reservoir depth for $N = 2$ even after 100 years.
Effects of $A$

In order to understand the effects of reservoir aspect ratio on the transport process additional computations are performed for $A = 0.5$ (laterally wide) and $A = 2$ (deep well). At early stages ($< 20$ years), the diffusion dominated average dissolution of CO$_2$ are almost same ($\bar{S} \sim 0.02$ after 4 years and 0.07 after 20 years) for any aspect ratio while at later stages ($> 100$ years) the average dissolution varies slightly with aspect ratio. Figure 6 shows the evolution of the CO$_2$ concentration front with time for $Ra_s = 10,000$, $N = 100$, and $A = 0.5$. The CO$_2$ distribution after 100 years is very uneven.

Interestingly, the mixing is very slow near the side walls. It is also noted that the reservoir aspect ratio has little effect on the CO$_2$ dissolution. When the thermal effect is made significant by decreasing $N$ to 2, the
concentration distribution obtained at 100 and 500 years are shown in Figure 7. Unlike for \( N = 100 \), the mixing seems even and almost symmetrical about the vertical midline at 100 years. In this case, the increased thermal effect counteracts with the solutal buoyancy effects making the dissolution process slower. Therefore the average dissolution is low (0.17). Over time the front propagation accelerates little more resulting in average dissolution 0.48 after 500 years.

For a deeper reservoir \((A = 2)\), Figure 8 shows the results for the case \( Ra_s = 10,000 \) and \( N = 100 \). In this case also, the early stage behavior is same as that for \( A = 0.5 \) or 1. The 23 cells merge to form only 4 plumes after 20 years where concentrations at left walls are invariant and one front advances along the edge of the right wall. The middle three \( CO_2 \) fronts merge to only a long one as time marches. However, the plume at the right edge becomes bigger pushing others to left. Thus the dissolution accelerates, reaching 0.60 after 500 years. When the thermal effect is increased \((N = 2)\) the long term dissolution process changes. After 100 years of simulation, plumes of various random sizes and shapes are traced. During the 500 year period only one big \( CO_2 \) front is survived, which is yet to reach the bottom. Average concentration of the aquifer at this time is 0.46. These are shown in Figure 9. Actually internal interactions among the fingers decrease, and the time required for \( CO_2 \) front to touch the bottom of the reservoir increases with increasing aspect ratio. These are consistent with the findings by Farajzadeh et al (Farajzadeh et al., 2007).
Figure 8: Concentration profiles for $Ra_s = 10000$, $N = 100$, $A = 2.0$, at (a) $t = 4$, (b) $t = 20$, (c) $t = 100$, (d) $t = 500$ years.

Figure 9: Concentration profiles for $Ra_s = 10000$, $N = 2$, $A = 2.0$, at (a) $t = 100$, (b) $t = 500$ years.

Figure 10 shows the average dissolution of $\text{CO}_2$ over time up to 500 years in a square
reservoir ($A = 1$) for various equivalent Rayleigh number, $Ra_e$. In early years ($< 80$) the average dissolution rate is faster than that at later times. Also the dissolution rate at any specific time increases with increasing $Ra_e$. For lower values of $Ra_e$, the CO$_2$ dissolution rate is slower because convective flow is weak and diffusion tends to dominate. To understand the behavior of the dissolution rate over a very long period, simulations were run up to 6,000 years at a fixed $Ra_e$ of 9,900 for a square reservoir and the dissolution rate is presented in Figure 11. It is noted that up to about 450 years the slope is very steep. After that the rate becomes slower and after 2,000 years, the rate of dissolution becomes asymptotic to about 0.95. Primarily, every concentration finger is surrounded by thin shear layer, and as they mix together over the time the layer becomes thicker and after long time the propagation again becomes diffusion dominated. To reach complete equilibrium stage, when CO$_2$ concentration in the aquifer is uniform everywhere, may require thousands of years. Figure 12 shows the effects of aspect ratios on CO$_2$ dissolution rate. It is observed that CO$_2$ dissolution rate increases slightly as the reservoir becomes laterally wide with increasing aspect ratio because fluid gets more free space to move in laterally wide reservoir enhancing the convection. Therefore from practical point of view laterally wide reservoir is better candidate than the deep aquifer for sequestration of CO$_2$.
CONCLUDING REMARKS

Double diffusive natural convection in a two dimensional brine saturated porous media, subjected to vertical concentration and temperature gradients, is investigated numerically. The study is focused on the influence of the solutal and thermal buoyancy forces on the propagation of the concentration front and its resulting dissolution into the brine in the context of CO$_2$ sequestration in underground reservoirs. It is found that the thermal effect does not interfere the natural convection process significantly unless the buoyancy ratio, $N$, is low and close to one. For higher values of $N$ ($>50$), the equivalent Rayleigh number, $Ra_e$, does not change much and the double diffusion is basically like density-driven mass transfer due to concentration variation. The convection process enhances with increasing $Ra_e$, which depends on reservoir characteristics and physical properties of the brine, viz., porosity, permeability, diffusivity, concentration gradient, thermal gradient, etc.

As the time passes the number of CO$_2$ fronts decrease due to decreasing convection process. At very initial stage ($<10$ years) the average CO$_2$ dissolution is same for all cases studied because of diffusion domination propagation. After 500 years of simulation CO$_2$ dissolution is over 0.60 for $N = 100$, and over 0.40 for $N = 2$. After 2,000 years the dissolution process again becomes very slow. The reservoir may be completely CO$_2$ saturated after thousands of years. Changing reservoir aspect ratio does not affect the average dissolution rate much, however the laterally wide reservoir is favorable than the deeper one.

NOTATIONS

$A$ aspect ratio, $H/L$ [-]
$c$ concentration [mol/m$^3$]
$C_p$ heat capacity at constant pressure [Jkg$^{-1}$K$^{-1}$]
$D$ Diffusion coefficient [m$^2$/s]
$g$ acceleration due to gravity [m/s$^2$]
$H$ porous medium height [m]
$k$ permeability [m$^{-2}$]
$L$ porous medium length [m]
$n$ number of nodes
$p$ pressure [Pa]
$Le$ Lewis number [-]
$Pe$ Peclet number [-]
$Pr$ Prandtl number [-]
$Ra$ Rayleigh number [-]
$\bar{S}$ average dissolution
$t$ time [s]
$u$ velocity [m/s]
$x$ distance along x-axis
$z$ distance along z-axis

Greek letters
$\alpha$ thermal diffusivity
$\beta_c$ coefficient of density increase by concentration [m$^3$/mol]
$\beta_T$ coefficient of thermal expansion[K$^{-1}$]
$\varphi$ porosity [-]
$\mu$ viscosity [kgm$^{-1}$s$^{-1}$]
$\kappa$ thermal conductivity []
$\rho$ density [kg/m$^3$]
$\psi$ stream function [m$^3$/m$^{-1}$s$^{-1}$]

Superscript
* Dimensionless quantity

Subscripts
0 initial value
c concentration
$i$ node in x – direction
$j$ node in z – direction
$r$ reference value
$s$ solutal
$T$ Temperature
$x$ x-coordinate
$z$ z-coordinate
REFERENCES

correlations and data. Desalination and Water Treatment 16, 354-380.