Abstract
In this paper we follow a similar procedure as proposed by Koval (1963) to analytically model the performance of gravitationally-unstable flow in porous media. The Koval model is analogous to the Buckley-Leverett method and multiplies the heterogeneity index of the system as an input \( (H\text{-factor}) \) with the fluid-flow (here gravity) induced instability factor, \( E \) to obtain the Koval factor \( K_G = HE \). This paper only considers the gravity induced instability factor \( E \) \((H=1)\). The Koval factor is implemented in a modified fractional flow function that includes a dilution effect when the CO\(_2\) moves away from the interface to describe countercurrent gravity flow. The pseudo two-phase flow problem provides the average concentration of CO\(_2\) in the brine as a function of distance.

The \( K_G \)-factor can be used in commercial simulators to account for the density-driven natural convection, which cannot be currently captured because the grid cells are typically orders of magnitude larger than the wavelength of the initial fingers. Such natural convection effects occur in storage of greenhouse gases in aquifers and EOR processes using carbon dioxide or other solvents.

A comparison of the analytical model with the horizontally-averaged concentrations obtained from 2-D numerical simulations provides a correlation for calculation of the \( K_G \)-factor for different Rayleigh numbers. The model shows a rarefaction followed by shock-like behavior because the CO\(_2\) concentration decreases away from the gaseous CO\(_2\)-liquid interface. The agreement between the analytical model and full numerical simulation is practically acceptable. We leave the introduction of the heterogeneity factor for future work.

Introduction
When a denser fluid is placed on top of a lighter one in the gravity field, it can lead to Rayleigh-Taylor instabilities (Taylor, 1950). This phenomenon is of importance for many fields of science and engineering (e.g. see Sharp, 1984); however, we confine our interest to CO\(_2\)-brine and CO\(_2\)-oil systems, relevant for CO\(_2\) sequestration and enhanced oil recovery processes. The instabilities initiated by local density increase of brine (or oil), caused by dissolution of CO\(_2\), increases the mass-transfer rate of CO\(_2\) in brine (or oil) (Ennis-King and Paterson, 2000; Farajzadeh et al, 2007, 2009; Yang and Gu, 2006; Pau et al., 2010; Hassanzadeh et al., 2007; Neufeld et al., 2010; Riaz et al., 2006; Riaz and Tchelepi, 2006). The increase of the mass-transfer rate is equivalent to the dissolution of a larger amount of CO\(_2\) in a shorter period of time and faster propagation of CO\(_2\) in porous media (aquifers and hydrocarbon reservoirs). The large volume of dissolved CO\(_2\) remains permanently in the liquid (at least as long as the pressure remains unchanged) and poses no threat of leakage, which is favorable for geological storage of CO\(_2\).

Moreover, one of the challenges in the application of chemically-enhanced oil recovery methods for the naturally fractured reservoirs using solvents is the slow mass transfer between the solvent in the fracture and the crude oil in the matrix. By injecting a solvent that is miscible with oil and increases the density of oil (for instance CO\(_2\)), natural convection phenomenon could enhance the transfer rates, increase the mixing between the solvent and the oil, and accelerate the oil production. Naturally, the time required for the initiation of the convection and in case of injection of costly solvents the recycling of the solvent play a crucial role in the (economic) success of the proposed method of oil production.
The efficiency of mixing in density-driven natural convection is governed by the Rayleigh number, which includes the reservoir permeability and the density difference. Stability analysis of the saturated porous layers under density-driven natural convection effects indicates that the time required for the initiation of natural convection is proportional to $-\frac{1}{Ra^2}$ and the critical wavelength is proportional to $-\frac{1}{Ra^2}$ (Meulenbroek et al., 2011; Riaz et al., 2006). The critical wavelength, $\lambda_c$, in an indication of the grid size required to capture the initiation of the initial fingers. Let us define Rayleigh number by:

$$Ra = \frac{k\Delta\rho gh}{\varphi \mu \phi}.$$  (1)

For the typical values of $k = 1$ Darcy, $\Delta\rho = 10$ kg/m$^3$, $g = 10$ m/s$^2$, $H = 50$ m, $\mu_w = 1$ cP, $D = 2\times10^{-9}$ m$^2$/s, and $\varphi = 0.2$ we obtain: $Ra = 1.25\times10^5$, which using the analysis of Meulenbroek et al. (2011) provides: $\lambda_c = 110H / Ra \sim 0.009H = 50$ cm. This implies that accurate estimation of the amount of dissolved CO$_2$ in brine under these conditions requires grid sizes much smaller than 50 cm. For highly permeable and heterogeneous porous media the required grid size may be too small to resolve even with massively parallel architectures (Lu and Lichtner, 2007; Pau et al., 2010). This necessitates the development of simpler models that could approximately quantify the amount of dissolved CO$_2$ after injection period taking into account the instabilities.

In miscible displacement the viscosity difference between the solvent and the oil leads to development of fingers that adversely affects the oil recovery. Because of the fingering behavior the evolution of concentration of the solvent cannot be predicted by fractional-flow-based miscible displacement models, originally described by Peaceman and Rachford (1962). Koval (1963) developed a simple model to account for the instabilities (fingering behavior) observed in the miscible displacements. In his model the fractional-flow function in the Buckley-Leverett equation is replaced by:

$$f_{koral}(c) = \frac{1}{1 + \frac{1 - c}{c} \frac{1}{K_G}}.$$  (2)

where the Koval factor $K_G = H_k E$. Here $E$ is the “effective” viscosity ratio and is defined as the ratio between the oil viscosity and the viscosity of mixture of oil and solvent in which the volume fraction of the solvent is $c_e$. It turns out from experimental results that $c_e = 0.22$. $E$ is chosen such that the results of the model fit the experimental data. For heterogeneous reservoirs $E$ is multiplied by the Koval heterogeneity index, $H_k$, which is related to the Dykстра-Parson’s coefficient by

$$\log H_k = \frac{V_{DP}}{(1 - V_{DP})^{0.2}}.$$  (3)

The degree of the heterogeneity of the permeability field determines the character of the density-driven natural convection flow in porous media. Similar to the instabilities induced by the viscosity difference between the fluids (Waggoner et al; 1991), instabilities induced by a density difference can lead to fingering, channeling, and dispersive regimes depending on the degree of the permeability variance (Dykstra-Parson’s coefficient) and the correlation length of the porous medium (Farajzadeh et al, 2011; Ranganathan et al; 2011). The dispersive regime (characteristic of flow in media with a high degree of heterogeneity) can be analytically modeled by choosing an effective dispersion coefficient in a diffusion-based model. In the channeling regime, which occurs for medium degree of heterogeneity, there is no correlation with the measures of heterogeneity and the transfer rate of CO$_2$ in water and therefore the method proposed by Koval cannot be applied without modification. Here, we confine our interest to find an analytical model for low degree of heterogeneity, which leads to fingering regime. It could be validated that for a low degree of heterogeneity the effect of heterogeneity on the transfer rate is relatively small.

The objective of this paper is to develop an analytical model that predicts the performance of gravitationally unstable flow in porous media. Our special focus will be on the inclusion of the effect of fingering on the transfer rate of CO$_2$ in brine. We use an analogous procedure as proposed by Koval (1963). The proposed model is similar to the Buckley-Leverett method for gravity dominated flow. The flow function uses a “gravity fingering index” as an input ($K_G$-factor). The solution provides the average concentration of CO$_2$ in the brine as a function of distance and eventually the total mass of dissolved CO$_2$. The structure of the paper is as follows. First we describe the physical model and provide the ensuing equations. Next we use method of matched asymptotic expansions to obtain an approximate analytical solution for most of the described equations. Afterwards, we introduce empirical parameters into the model to take into account the fingering behavior and compare the results of the proposed model to the numerical simulations. Finally we draw the main conclusions of this study.
2. Physical Model

Figure 1 schematically shows the purpose of the model, i.e., to capture the averaged behavior of the process in the direction of the flow. If there is no instability, there will be a short transition zone (here the ensuing error function is represented as a shock) between the CO$_2$ containing brine and the initial brine that contains no CO$_2$. This occurs when the flow regime is diffusive (e.g. at initial stages of the process) or dispersive (for highly heterogeneous media). This behavior can be accurately modeled through diffusion-based models, albeit with an effective diffusion coefficient. When instabilities occur, the concentration front moves faster and there is a gradual change from high (horizontally averaged) concentration of CO$_2$ at the top to the initial concentration.

2.1. Formulation

We consider a one-dimensional porous medium of length $H$ that is initially saturated with water. The vertical coordinate, $z$, is taken positive in the downward direction. The constant porosity of the porous medium is $\phi$ and its permeability is $k$. Initially there is no CO$_2$ dissolved in water ($c_i = 0$). We assume a no flow boundary at the bottom of the porous medium. CO$_2$ is continuously supplied from the top, i.e., the CO$_2$ concentration at the top is kept constant and therefore the water at the interface is fully saturated with CO$_2$. The water and water saturated with CO$_2$ (referred to as “mixture” and represented by $m$) are considered as two separate phases; (1) pure water with density $\rho_w$ and viscosity $\mu_w$ and (2) a mixture phase with density $\rho_m$ and viscosity $\mu_m$. We disregard the presence of a capillary transition zone between water and mixture phases and assume that the relative permeability of the mixture is proportional to its saturation denoted by $S_m$. In the same way is the relative permeability of the water phase proportional to its saturation $S_w = 1 - S_m$.

2.2. Governing Equations

The motion of fluids in a porous medium can be described by Darcy’s law. The Darcy equation for the mixture can be written as

$$u_m = -\lambda_m \left( \frac{\partial p_m}{\partial z} - \rho_m g \right).$$

The Darcy equation for water reads

$$u_w = -\lambda_w \left( \frac{\partial p_w}{\partial z} - \rho_w g \right).$$

where $u$ is the Darcy velocity, $p_\alpha$, ($\alpha = m, w$) is the pressure of phase $\alpha$, $\rho_\alpha$ is the density of phase $\alpha$, and $g$ is the acceleration due to gravity, and $\lambda_\alpha = kk_\alpha / \mu_\alpha$ ($\alpha = m, w$) is the mobility, which is the ratio between the phase permeability $kk_\alpha$ and the viscosity $\mu_\alpha$. Subscripts $m$ and $w$ denote the CO$_2$+water mixture and the initial water, respectively. Both viscosity and density are assumed to be functions of the CO$_2$ concentration.

The saturated density difference between an aqueous solution of CO$_2$ and pure water is given by $c_p P_f$, where the value of the $c_p = 0.261 \text{ kg/m}^3/\text{bar}$ for pure water (see Gmelin p 72). $c_p$ will be less for formation brines, because the solubility of CO$_2$ in water decreases with increasing salinity. Therefore, the density of the mixture can be assumed to increase according to the following relationship:
\[ \Delta \rho = \rho_m - \rho_w = c_{\rho} P_{s} S_m^n, \]  

where \( S_m^n \) represents a dilution effect as the gravity fingers move away from the interface.

The saturation exponent \( n \) can have values between 0 and 1. We assume ideal mixing and use Boussinesq approximation, i.e., we only consider the density variations in Eqs. (4) and (5). With this assumption and because there is no source or sink in our model, the volume conservation is equivalent to mass conservation and hence

\[ u_m + u_w = 0, \]  

which implies a countercurrent flow. We can derive an expression for the Darcy velocity of the mixture, i.e.,

\[ u_m + u_w = 0 = u_m - \frac{\lambda_w}{\lambda_m} \left( \frac{\partial p}{\partial z} - \rho_m g \right) - \lambda_w (\rho_m - \rho_w) g. \]  

Hence it follows that

\[ u_m = \frac{\lambda_w \lambda_m}{\lambda_w + \lambda_m} (\rho_m - \rho_w) g. \]  

The conservation law for the mixture including diffusion reads:

\[ \varphi \frac{\partial S_m}{\partial t} + \frac{\partial u_m}{\partial z} = \varphi D \frac{\partial^2 S_m}{\partial z^2}. \]  

Replacing Eq. (9) in Eq. (10) leads to

\[ \varphi \frac{\partial S_m}{\partial t} + \frac{\partial}{\partial z} \left( \frac{\lambda_w \lambda_m}{\lambda_w + \lambda_m} (\rho_m - \rho_w) g \right) = \varphi D \frac{\partial^2 S_m}{\partial z^2}. \]  

2.3. Dimensionless form of the equations

We scale the velocity with a characteristic velocity \( u_c = k c P_{s} g / \mu_w \), length with \( H \), and \( t \) with \( \varphi H / u_c \). Therefore, Eq. (11) leads to:

\[ \frac{\partial S_m}{\partial \tau} + \frac{\partial}{\partial \xi} \left( \frac{S_m^n k_{rw}}{\Lambda^{-1} + 1} \right) = \varepsilon \frac{\partial^2 S_m}{\partial \xi^2}. \]  

where \( \tau = (u_c / \varphi H) t \), \( \xi = z / H \), \( \Lambda = \lambda_m / \lambda_w \), \( \varepsilon = 1 / Pe \), and \( Pe = u_c H / D = Ra \). We define the “fractional flow function” as

\[ f(S_m) = \frac{S_m^n k_{rw}}{\Lambda^{-1} + 1}. \]  

Therefore, Eq. (12) can be written as

\[ \frac{\partial S_m}{\partial \tau} + \frac{\partial f(S_m)}{\partial S_m} \frac{\partial S_m}{\partial \xi} = \varepsilon \frac{\partial^2 S_m}{\partial \xi^2}. \]

The Peclet number, \( Pe \), is defined as the ratio between the convective and diffusive fluxes. For typical values mentioned earlier we will obtain \( u_c = 2 \times 10^{-7} \) m/s. Using \( D = 2 \times 10^{-9} \) m²/s we obtain: \( Pe = 5000 \). Zimmerman and Homsy (1992) noted that for \( Pe >> 1 \)
it is reasonable to model diffusion as constant and isotropic because at large Peclet numbers all averaged quantities are independent of any anisotropy (see also Booth, 2008). Hence taking $D$ as constant is justified. We notice that for very large Peclet numbers ($Pe \to \infty$ or $\varepsilon \to 0$) Eq. (14) converts to the classical Buckley-Leverett equation, albeit with a different fractional-flow function.

At the displacement problems the initiation of the flow is forced by injection. In our case, the flow rate cannot be forced. Therefore a crucial parameter of the model is to define the top boundary condition. If we assume that the saturation of the mixture is one, it is not immediately clear how unstable convective motion would initiate. Here we will investigate the approximate analytical solution of Eq. (14) with the method of matched asymptotic expansions and compare it with the numerical solution. The method is briefly explained in Appendix A.

2.4. Approximate solutions

The approximate solution consists of an inner solution in the domain $R_1 = [0, \varepsilon)$, where diffusion dominates and an outer solution in $R_2 = (\varepsilon, 1]$ that is convection dominated. It turns out that the outer solution consists of a rarefaction solution combined with a shock and that the inner solution concerns the stationary diffusion equation.

2.4.1. Rarefaction (or outer) and shock solutions

In the absence of diffusion the solution consists of rarefactions, constant states and shocks. Firstly, we derive the rarefaction solution and the constant state. We can use a coordinate transformation $\eta = \xi / \tau$ and obtain from Eq. (14)

$$-\eta + \frac{df(S_m)}{dS_m} \frac{\partial S_m}{\partial \eta} = 0 \ .$$

The solution of Eq. (15) is either a constant state $dS_w / d\eta = 0$ or

$$\eta = \frac{df(S_m)}{dS_m} \ .$$

This equation can be solved to obtain $S_m = S_m(\eta)$ on condition that the second derivative of the fractional-flow function does not change the sign (Landau and Lifshitz, 1959; Silin et al., 2009), i.e., in the absence of shocks. Replacing the parameters in Eq. (13) we obtain

$$\eta = \frac{d}{dS_m} \left\{ \frac{(1 - S_m) S_m^u}{(1 - S_m) - K_G S_m} \right\} \ ,$$

where, we have replaced $\mu_w / \mu_m$ with $K_G$ similar to Koval (1963). The term in brackets is plotted in Figure 2. At downstream of the diffusive layer, the solution of Eq. (17) for different values of $K_G$ starts at the saturation corresponding to the highest phase velocity (maximum of the fraction-flow function) with a rarefaction solution. We refer to $K_G$ as the “gravity fingering index” and it can be used as fitting parameter to obtain agreement between the numerical and analytical results. The solution will include a shock for $n > 0$. The general rarefaction solution of Eq. (17) is
\[ \eta = \left\{ \begin{array}{l}
\frac{S_m^n}{(1 - S_m)} \frac{1}{K_G} + 1 + \frac{n(1 - S_m)}{S_m} \frac{1}{K_G} + 1 \left( \frac{1 - S_m}{S_m} \right) \frac{1 + \frac{1 - S_m}{S_m}}{K_G} + 1 \left( \frac{1 - S_m}{S_m} \right) \frac{1}{K_G} + 1 \right) \end{array} \right\}. \quad (18) \]

For \( K_G = 1 \), i.e., \( \mu_m = \mu_w \) and \( n = 0 \) the solution is simplified to:

\[ \eta = \left( 1 - 2S_m \right), \quad (19) \]

or

\[ S_m = \frac{1}{2} \left( 1 - \eta \right) = \frac{1}{2} \left( 1 - \frac{\xi}{\tau} \right) = \frac{1}{2} \left( 1 - \frac{\varphi \mu_w}{k \left( \rho_m - \rho_w \right) g \left( t \right)} \right). \quad (20) \]

The rarefaction solution will be followed by a shock if \( n \) has a non-zero value. The shock occurs when the following condition is satisfied:

\[ \frac{f(S_m, \text{shock})}{S_m, \text{shock}} = \frac{df(S_m)}{dS_m}, \quad (21) \]

where we have assumed that initially there is no CO\(_2\) dissolved in the brine. Using Eq. (13) the shock saturation equals:

\[ S_m, \text{shock} = \frac{1}{2} K_G (n - 1) - 2n + \frac{\sqrt{K_G^2(n - 1)^2 + 4K_Gn}}{n(K_G - 1)}. \quad (22) \]

Figure 2: Plot of dimensionless phase velocity, i.e., term in brackets in RHS of Eq. (17) versus saturation. We use \( n = 0.22 \). Just downstream of the diffusion layer all solutions start at the saturation corresponding to the highest phase velocity with a rarefaction possibly followed by a shock.
**2.4.2. Diffusion equation (or inner) solution**

In $R_1$ we rescale the $z$-coordinate: $X = \frac{\xi}{\varepsilon}$ in Eq. (12). This leads to

$$
\varepsilon \frac{\partial S_{m}^{in}}{\partial \tau} + \frac{\partial f\left( S_{m}^{in} \right)}{\partial X} = \frac{\partial^2 S_{m}^{in}}{\partial X^2} .
$$

(23)

The term $\varepsilon \partial S_{m}^{in} / \partial \tau$ is small with respect to the other terms and can be omitted. There Eq. (25) becomes

$$
\frac{\partial f\left( S_{m}^{in} \right)}{\partial X} = \frac{\partial^2 S_{m}^{in}}{\partial X^2} .
$$

(24)

Integration of Eq. (24) gives

$$
\frac{\partial S_{m}^{in}}{\partial X} = f\left( S_{m} \right) + \kappa ,
$$

(25)

where, $\kappa$ is a constant. The matching condition reads (Verhulst, 2000; van Dyke, 1975):

$$
\lim_{X \to \infty} S_{m}^{in} (X) = \lim_{\xi \to 0} S_{m}^{out} (\xi) = S_{m}^{0} .
$$

From Eq. (15) we have:

$$
\eta = 0 \rightarrow S_{m}^{0} = \frac{1}{2} \left( \frac{-2(n+1) + nK_{G} + \sqrt{4K_{G}(n+1) + n^2K_{G}^2}}{(n+1)(K_{G} - 1)} \right).
$$

(26)

$S_{m}^{0}$ is the saturation that separates the diffusive and rarefaction regimes. Notice that this is the saturation where the fractional-flow function attains its maximum. The second boundary for Eq. (24) reads: $S_{m}^{0} (0) = 1$. Expanding Eq. (24) gives

$$
\frac{dS_{m}^{in}}{dX} = f\left( S_{m}^{in} \right) + \kappa = \frac{(S_{m}^{in})^{n+1}K_{G} - (S_{m}^{in})^{n+2}K_{G} + \kappa K_{G}S_{m}^{in} + \kappa - \kappa S_{m}^{in}}{K_{G}S_{m}^{in} + 1 - S_{m}^{in}}.
$$

(27)

We only get bounded solution when

$$
\lim_{X \to \infty} \frac{dS_{m}^{in}}{dX} = \lim_{X \to \infty} \left( f\left( S_{m}^{in} \right) + \kappa \right) = 0 .
$$

(28)

This means that

$$
\kappa = -f\left( S_{m}^{0} \right).
$$

(29)

Replacing $\kappa$ in Eq. (25) and using the definition of the fractional-flow function in Eq. (13) we obtain

$$
\int_{1}^{\infty} dX = \int_{1}^{S_{m}^{0}} \frac{dS_{m}^{in}}{f\left( S_{m}^{in} \right) - f\left( S_{m}^{0} \right)}.
$$

(30)

Explicit integration of Eq. (30) is not trivial and should be solved numerically for different values of $n$ and $K_{G}$ to obtain the saturation profile. Nevertheless, for $n = 0$ and $n = 1$ it is possible to find an analytical solution. For $n = 1$ and $K_{G} = 1$, Eq. (26) gives:

$S_{m}^{0} = 1 / 2$ and the inner solution is simplified to

$$
S_{m}^{in} (X) = \frac{X + 4}{2X + 4} .
$$

(31)
Figure 3 plots the inner solution (Eq. 31) and outer solution (Eq. 19) of this case and compares it to the numerical solution. For \( n=1 \) and \( K_G = 1 \), the asymptotic value of the saturation is \( 2/3 \).

\[ \text{Figure 3: Inner solution (Eq. 31) and outer solution (19) and numerical solution of Eq. (12) for } Pe = 5000. \]

**Results and Discussion**

In this section we present the results of the 1-D analytical model and compare them to the results of the numerical simulations, which were performed on a 2-D porous medium with an aspect ratio of 1. A no-flow boundary condition was used at the sides and at the bottom of the medium. The concentration of CO\(_2\) at the top of the medium was assumed to be 1. To observe the fingering behavior the interface was initially perturbed. A fully implicit finite volume approach (Guçeri and Farouk, 1985) was used to solve the equations. The details of the numerical simulations can be found in Farajzadeh et al. (2007, 2011).

To compare the results, the concentration values of the 2D numerical simulations were averaged in the horizontal direction. An example is shown in Figure 4. The simulation of diffusion part of the problem requires grid cells that are in the order of \( 1/Ra \) in the vertical direction. Our numerical simulation consists of 81x81 grid cells and therefore we are not able to accurately model the diffusive regime, which is dominant before onset of the natural convection. However, this does not affect the estimated transfer rate (Farajzadeh et al., 2009).

\[ \text{Figure 4: The concentration profile of CO}_2 \text{ for } Ra = 2000 \text{ at } \tau = 6. \text{ The right plot is obtained by averaging the concentration value of left plot in the horizontal direction.} \]
Figure 5 through Figure 8 demonstrate the concentration profiles of CO$_2$ at different times for different Rayleigh numbers. The solid and dashed lines are the results of the analytical model and the numerical simulations, respectively. To consider the effect of fingering the dimensionless time should be scaled with the time-scaling factor, $\alpha_\tau$, in Eq. (18). This assures that the concentration front moves faster when fingering occurs. The agreement between the two models is acceptable, especially for lower Rayleigh numbers. We note from Figure 3 that the asymptotic saturation value $S_m$ near $z=0$ of the model is between 0.5 (for $n = 0$) and 2/3 ($n = 1$). However, the horizontally-averaged concentrations obtained from 2-D numerical simulations converge at a smaller value than this saturation value. The saturation value decreases further with increasing Rayleigh number. For higher Rayleigh numbers the value decreases to even smaller values as time progresses. Therefore, to obtain a reasonable match between the analytical and numerical models, the $K_G$-factor was modified such that the asymptotic values of the saturation values $S_m$ near the interface of the two models matched. We ignored time dependency of this value, i.e., we kept $K_G$-factor independent of time. It is interesting to mention that the best fit was obtained for $n = 0.22$ (Eq. 6) for all simulations. The other fitting parameters, i.e., $K_G$ and $\alpha_\tau$, are listed in Table 1. The $K_G$-factor increases with increasing Rayleigh number. Moreover, with increasing Rayleigh number the effect of convective mixing (gravity induced instability) becomes more pronounced and therefore the time required for CO$_2$ front to reach the bottom of the medium decreases (Riaz et al., 2006; Farajzadeh et al., 2007). On the other hand the density of the mixture increases with increasing Rayleigh number (Eq. 1). This means that the time-scaling factor ($\alpha_\tau$) includes both of these effects.

Note that our model is only valid until CO$_2$ reaches the bottom of the medium. After this time CO$_2$ saturated solution will start to fill up the layer from the bottom. This filling up has been treated in a pioneering paper of Siddiqui and Lake (1997) and later by Bedrikovetsky et al. (2001), who addressed an analogous problem concerning the secondary migration of oil. Oil was moving through countercurrent gravity drainage from the source rock to the future reservoir initially filled with water. The problem is almost the same as the problem considered here except that the saturation exponents of the relative permeabilities are not one for the true two-phase (oil and water) conditions and the trivial fact that the gravity is pointing in the opposite direction. Another main difference is that in the migration problem the upstream boundary condition (in the source rock) is that the oil saturation is a given low value and is not governed by a diffusion process like here. Downstream there is a seal, which may be partially leaking, which is one of the complicating factors in the paper by Siddiqui and Lake (1997). In our case we assumed a no-flow boundary at the bottom.

In analogy of the Siddiqui-Lake paper we expect that a reflected wave will occur once the CO$_2$ saturated solution hits the bottom of the reservoir. Indeed, the saturation of the carbon dioxide containing aqueous phase near the bottom will increase until it is in the right side of Figure 2. Here the wave velocity (see Eq. (17)) will be negative (reflected wave). Consequently the reservoir will stably fill up from the bottom with the saturated solution. This aspect will be left for future work.

![Figure 5: Comparison between the analytical (solid lines, $n=0.22$. $K_G=1.66, \alpha_\tau =0.8$) and numerical (dashed lines) models at different times for $Ra = 1000$.](image-url)
Figure 6: Comparison between the analytical (solid lines, \( n=0.22 \), \( K_G = 2.6, \alpha = 0.45 \)) and numerical (dashed lines) models at different times for \( Ra = 2000 \).

Figure 7: Comparison between the analytical (solid lines, \( n=0.22 \), \( K_G = 4.0, \alpha = 0.37 \)) and numerical (dashed lines) models at different times for \( Ra = 5000 \).

Figure 8: Concentration profiles obtained from the analytical (solid lines, \( n = 0.22 \), \( K_G = 5.6, \alpha = 0.25 \)) and numerical (dashed lines) models at different times for \( Ra = 10000 \).
Table 1: The values of $K_G$ and $\alpha_T$ used as fitting parameters to obtain the match between the analytical and numerical models.

<table>
<thead>
<tr>
<th>Rayleigh Number</th>
<th>$\alpha_T$</th>
<th>$K_G$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1000</td>
<td>0.80</td>
<td>1.66</td>
</tr>
<tr>
<td>2000</td>
<td>0.45</td>
<td>2.6</td>
</tr>
<tr>
<td>5000</td>
<td>0.37</td>
<td>4.0</td>
</tr>
<tr>
<td>10000</td>
<td>0.25</td>
<td>5.6</td>
</tr>
</tbody>
</table>

In Figure 9 we plot the parameters reported in Table 1 as a function of Rayleigh number. The fitting exercise provides the following relationships

\[
K_G = 0.042 Ra^{0.532}, \quad \alpha_T = 6.85 Ra^{-0.436}.
\]

These empirical relations can be used to “effectively” model the gravity-induced instabilities.

Figure 9: The fitting parameters as a function of Rayleigh number.

Figure 10 plots the mass of the dissolved CO$_2$ (normalized to the maximum mass that can be dissolved) for different Rayleigh numbers and compares it to the numerical model. For illustration purposes, the time has been divided to the corresponding Rayleigh number of each case. The analytical model is less accurate for the higher Rayleigh numbers. The disagreement is more pronounced at the initial stages and the analytical model under-predicts the dissolved mass. One possible reason for this is that the analytical model only consists of a convection part and there is no diffusion in the model. As mentioned earlier the transfer at the initial stages of the process is governed by diffusion. The numerical model, however, might over-predict the dissolved mass due to numerical dispersion.

Figure 10: The mass of CO$_2$ dissolved in water obtained from the analytical (dashed lines) and numerical (solid lines) models at different times for different Rayleigh numbers.
Conclusions

- Following a similar procedure as proposed by Koval (1963) we developed an analytical model to predict the performance of gravitationally unstable flow in porous media,

- The method of matched asymptotic expansions was used to obtain an approximate analytical solution for the relevant equations describing the diffusive and convective regimes. The analytical solution is in good agreement with the numerical results.

- The developed model takes the “gravity fingering index” (or \(K_G\)-factor), a time-scaling factor \(\alpha_\tau\), and a saturation exponent, \(n\), as input parameters and provides the average concentration of CO\(_2\) in the brine as a function of distance at different times.

- A comparison between the analytical model and the horizontally-averaged concentrations obtained from 2-D numerical simulations provides a correlation for calculation of the \(K_G\)-factor and the time-scaling factor \(\alpha_\tau\) for different Rayleigh numbers. The saturation exponent is \(n = 0.22\) for all cases.

- The model shows a rarefaction followed by shock-like behavior because the concentration in the fingers decreases \((n=0.22)\) away from the gaseous CO\(_2\)-liquid interface. The agreement between the Koval procedure and full numerical simulation is practically acceptable.

- The empirical relations between the \(K_G\)-factor and the Rayleigh number, and the time-scaling factor \(\alpha_\tau\) and the Rayleigh number can be used in commercial simulators to account for the density-driven natural convection, which cannot be currently captured because the grid cells are typically orders of magnitude larger than the wavelength of the initial fingers.

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Nomenclature

c Dimensionless concentration [-]

\(D\) Diffusion coefficient [m\(^2\)/s]

\(E\) Effective viscosity ratio

\(f\) Fractional-flow function

\(g\) acceleration due to gravity [m/s\(^2\)]

\(H\) Height of the porous medium [m]

\(H_k\) Koval heterogeneity factor [-]

\(k\) Permeability of the porous medium [m\(^2\)]

\(k_r\) Relative permeability [-]

\(K\) Koval factor

\(K_G\) Gravity fingering index

\(p\) Pressure [Pa]

\(Pe\) Peclet number [-]

\(Ra\) Rayleigh number [-]

\(S\) Phase saturation [-]

\(t\) Time [sec]

\(u_c\) Dimensionless velocity [-]

\(u\) Velocity [m/s]

\(V_{DP}\) Dykstra-Parsons coefficient

\(X\) Re-scaled z coordinate \((\xi/\varepsilon)\)

\(z\) Distance in z coordinate

Greek symbols

\(\alpha_\tau\) Time-scaling factor

\(\varepsilon\) \(1/Pe\)

\(\varphi\) Porosity of the porous medium [-]
\(\eta\) Transformation coordinate
\(\lambda_c\) Wavelength [m]
\(\lambda\) Mobility \((k_r/\mu)\)
\(\Lambda\) Mobility ratio of phase [-]
\(\mu\) Viscosity of the fluid [kg/m-sec]
\(\xi\) Dimensionless distance \((z/H)\)
\(\rho\) Density of the fluid [kg/m³]
\(\tau\) Dimensionless time [-]

Subscripts
\(i\) Initial value of the quantity
\(m\) Mixture phase
\(s\) Pure solvent
\(w\) Water phase
\(z\) Quantity in \(z\)-direction

References
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Appendix A: Matched Asymptotic Expansions

We consider the equation

\[ \varepsilon \frac{d^2}{dx^2} c(x) + \frac{d}{dx} c(x) = a \]  \hspace{0.5cm} (A.1)

where \( \varepsilon \) is a small Peclet number and \( x \) a dimensionless space coordinate, and a source term. The boundary conditions read \( c(x = 0) = 0 \) and \( c(x = 1) = 1 \).

The exact solution is given by

\[ c(x) = \left(1 - a\right)\left(\varepsilon \frac{x}{\varepsilon} - 1\right) + ax \]  \hspace{0.5cm} (A.2)

The solution away from the boundary can be approximated as the solution obtained with \( \varepsilon = 0 \), i.e., \( \frac{dc_o}{dx} = a \), which only needs to satisfy only the outer boundary condition \( c_o(x = 1) = 1 \). The solution is

\[ c_o(x) = a(x - 1) + 1 \]  \hspace{0.5cm} (A.3)

Using a coordinate transformation \( X = x / \varepsilon \) we can write Eq. (A.1) as

\[ \frac{d^2}{dX^2} c_1(X) + \frac{d}{dX} c_1(X) = a \varepsilon \]  \hspace{0.5cm} (A.4)
The boundary conditions read \( c_i(X = 0) = 0 \) and \( c_i(X = 1 / \varepsilon) = 1 \). The outer boundary condition is only stated for easy reference. Without the outer boundary condition the solution in the limit that \( \varepsilon = 0 \) is any multiple of \( \beta(1 - \exp(-X)) \), i.e., \( \beta(1 - \exp(-X)) \), where \( \beta \) is an arbitrary constant. Imposing the outer boundary condition would give in the limit that \( \varepsilon \to 0 \) a multiplicative factor of unity, but the exact solution shows that this is incorrect. The outer boundary condition must be dropped for the inner solution in the same way that the inner boundary was dropped for the inner solution. Instead the inner solution must be matched to the outer solution using the matching principle, i.e.,

The \( m \) term inner expansion of the \( n \) term outer expansion =

\[
\text{The } n \text{ term outer expansion of the } m \text{ term inner expansion} \quad (A.5)
\]

The \( m \) and \( n \) can be taken as any two integers. By definition the \( m \) term inner expansion of the \( n \) term outer expansion is found by rewriting it in inner variables, expanding asymptotically for small \( \varepsilon \) and truncating the results to \( n \) terms. The inner solution \( (1 - \exp(-X)) \) rewritten in outer variables reads \( \beta(1 - \exp(-x / \varepsilon)) \). This solution truncated at zeroth (or any higher order for that matter) in \( \varepsilon \) reads \( c_i^\varepsilon = \beta \). The outer solution written in terms of inner variables reads \( c_o^\varepsilon = a \left( \varepsilon X - 1 \right) + 1 \). Until zeroth order we find \( \beta = 1 - a \).

\[
c_o(x) = a \left( x - 1 \right) + 1 \quad \text{for the outer solution} \quad (A.6)
\]
\[
c_i(X) = \left( 1 - a \right) \left( 1 - \exp(-X) \right) \quad \text{for the inner solution} \quad (A.7)
\]

The comparison between the inner and outer solutions to the exact equation is given in Figure A.1.

![Figure A.1: Inner solution (Eq. 7) and outer solution (A.6) and exact solution of Eq. (A.2)](image-url)