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Closed-form Analytical Approaches to Constrain Fraction of Injected CO₂ Dissolving in Brine During CO₂ Storage in Saline Aquifers

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Abstract

CO₂ dissolution in the storage aquifer's brine is one of the main CO₂ immobilization mechanisms in the subsurface. During injection, CO₂ dissolution is primarily governed by the contact area between the CO₂ and brine. Accordingly, when using finite-difference numerical simulation tools to model the CO₂-brine displacement, the model discretization size affects the level of dissolution given that it controls the contact area. Grid size should be carefully selected to avoid unrealistic dissolution estimations. It is important to obtain an independent estimate of the dissolution using discretization-free analytical approaches. In this study, the author presents two analytical approaches to estimate the range of CO₂ dissolution. The first method assumes gravity-dominated flow, with a limited role for the capillary force in the vertical direction, resulting in vertical equilibrium modeling approach. The second assumes viscous-dominated flow with no role for gravity force, maximizing the CO₂-brine contact area and dissolution rate through the fractional flow modeling approach. The two approaches are applied to an example CO₂ injection case and their resulting ranges are compared with estimates from numerical simulations considering different spatial discretizations. Numerical simulations show that large discretizations can result in unrealistic dissolution rates outside the analytically-driven range.

Introduction

Various trapping mechanisms enable immobilization of the injected CO₂ in the subsurface in Geologic CO₂ storage (GCS) projects. Given the relatively substantial dissolution of CO₂ in the brine, CO₂ dissolution is one of the major trapping mechanisms in GCS in deep saline aquifers. Over long term, CO₂ dissolution may be governed by convective mixing due to the higher density of the CO₂-dissolved brine compared with brine with no dissolve CO₂. However, during CO₂ injection, CO₂-brine mass transfer is primarily governed by the surface contact area of the two phases. CO₂-brine displacement is often modelled using finite-difference numerical simulation tools that spatially discretize the storage aquifer

domain. However, the fraction of dissolved CO₂ can be a strong function of the level of spatial discretization of the model. Larger grid size results in larger CO₂ dissolution given that it gives larger CO₂-brine contact area to the CO₂ displacing the brine. As such, numerical simulations may result in unrealistic and unphysical values of CO₂ dissolution.

Evaluating the fraction of injected CO₂ that gets dissolved in the brine, independent of the numerical simulation and space discretization is therefore required. In this work, analytical-based closed-form equations are presented to estimate the range over which the CO₂ dissolution fraction (DCF) may vary.

CO₂ dissolution analytical models

We consider two end scenarios for the CO₂-brine displacement to obtain the dissolution range. First, we consider the flow to be gravity dominant with limited role for the capillary forces in vertical direction. This approach is known as the vertical equilibrium model (VEM) which results in a sharp interface separating the free-phase CO₂ from the brine. This approach results in minimized contact area between the injected CO₂ and the in-situ reservoir brine.

Therefore, it results in minimum amount of CO₂ dissolution. To obtain the higher limit of dissolution, we assume that the flow is viscous dominant with negligible role for gravity. This approach results in the fractional flow model (FFM) which maximizes the contact area and thus, the CO₂ dissolution amount. Schematic of CO₂ and brine saturation distributions considering VEM and FFM are shown in Figure 1.

In VEM, CO₂ comes into contact with the aqueous phase only above the CO₂-brine interface where the aqueous phase saturation is fixed at the irreducible water saturation, S_{wi} . Moles of free-phase CO₂ (m_f) are given by:

$$m_f = \rho_c(1 - S_{wi})V_p \quad (1)$$

where V_p denotes the pore volume contacted by CO₂ and ρ_c is the molar density of CO₂ at reservoir conditions. Moles of dissolved CO₂ (m_d) are given by:

$$m_d = \frac{\text{Moles of dissolved CO}_2}{\text{Volume of aqueous phase}} S_{wi}V_p = x_c \rho_a S_{wi}V_p \quad (2)$$

Where x_c is mole fraction of CO₂ dissolved in brine and ρ_a is aqueous phase density. Therefore, the mole ratio of dissolved CO₂ over the total injected CO₂ is given by:

$$\text{DCF} = \frac{m_d}{m_d + m_f} = \frac{1}{1 + \frac{\rho_c(1 - S_{wi})}{x_c \rho_a S_{wi}}} \quad (3)$$

In FFM, CO₂ front propagation is evaluated based on the fractional flow of the gaseous phase (f_g) versus gaseous phase saturation (S_g) (Burton et al. 2009, Zeidouni et al. 2009, Azizi and Cinar 2013):

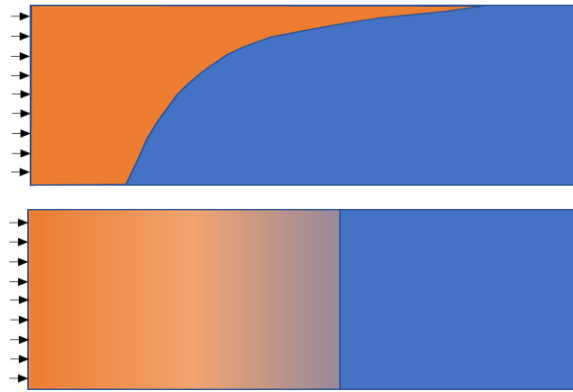


Figure 1. Schematic of CO₂ (orange) and brine (blue) saturation distribution considering vertical equilibrium model (top) versus fractional flow model (FFM).

$$\frac{r_f^2}{t} = \frac{q}{\pi h \phi} \left. \frac{df_g}{dS_g} \right|_{S_g^f} \quad (4)$$

where r_f is the plume radius, q is the volumetric injection rate at reservoir sandface, t is the injection period, h is the reservoir thickness, and ϕ is porosity. S_g^f is the gaseous phase saturation at the plume front which is obtained by drawing a tangent line from the origin on the $f_g - S_g$ graph. Neglecting the water vaporization (and thus, the dry-out effect), the average saturation of the gaseous phase within the CO₂ plume can be approximated by:

$$\bar{S}_g = \frac{1}{\left. \frac{df_g}{dS_g} \right|_{S_g^f}} \quad (5)$$

The total moles of injected CO₂ is $qt\rho_c$. The brine contacted by this injected CO₂ gets saturated by CO₂ at x_c mole fraction. Accordingly, total moles of dissolved CO₂ is $\pi r_f^2 h \phi (1 - \bar{S}_g) x_c \rho_a$ where ρ_a is the aqueous phase molar density. Combining this expression with Eqs (4) and (5), we get:

$$\text{DCF} = \frac{x_c \rho_a}{\rho_c} \left(\left. \frac{df_g}{dS_g} \right|_{S_g^f} - 1 \right) \quad (6)$$

As noted above, the dry-out effect has been neglected in deriving Eq. (6). Accounting for dry-out would change the results slightly at the cost of complicating the final solution which becomes (derivation omitted for brevity):

$$\text{DCF} = \frac{\rho_a x_c}{\rho_c} \left(\left. S_a^f \frac{df_g}{dS_g} \right|_{S_g^f} - S_a^d \left. \frac{df_g}{dS_g} \right|_{S_g^d} + (f_g^f - f_g^d) \right) \quad (7)$$

In this equation, superscript d denotes the dry-out front characteristics.

Model application

In this section, the above analytical approaches are applied to an example case with operational and reservoir conditions of a relatively shallow storage formation. The analytical approaches provide a range for the DCF which is then compared with the numerical simulation results obtained for various spatial discretizations.

For the example case, CO₂ is injected at fixed rate of 2100 m³/day (bottomhole condition) into a 30-m thick aquifer for 10 years. The aquifer porosity and permeability are 0.3 and 100 mD, respectively. The aquifer top depth is 1200 m where the initial pressure is 12 MPa. Reservoir temperature is 41.7 degC. The relative permeability curves are given based on power-law relationships. For the aqueous phase at saturation S_a , the aqueous phase relative permeability is $k_{ra} = \left(\frac{S_a - S_{wi}}{1 - S_{wi}} \right)^2$ and the gaseous CO₂-rich phase relative permeability is $k_{rg} = 0.8 \left(\frac{1 - S_a}{1 - S_{wi}} \right)^2$. Brine density and CO₂ density are 1000 and 715 kg/m³, respectively at initial reservoir conditions. The mole fraction of CO₂ dissolved in brine is 0.022 at reservoir initial conditions. Brine viscosity is 0.635 cp and its compressibility is 5e-7/kPa. Rock compressibility is 1e-6/kPa. The reservoir model is radial with 20 km external radius.

Closer look at Eqs (3) and (6) proves that S_{wi} is the main reservoir parameter controlling DCF. According to Eq. 3, increasing S_{wi} decreases the denominator and therefore increases DCF. The same observation

can be derived from Eq. (6). Increasing S_{wi} increases the slope of the tangent line drawn on the S-shape f_g - S_g graph (i.e. $df_g/dS_g|_{S_g^f}$) because it pushes the curve to the left resulting in increased DCF (see Figure 2a). For the modeled case herein, S_{wi} is set at 0.25, 0.35, and 0.45. Applying the VEM approach given by Eq. (3), the corresponding DCF values are 0.023, 0.037, and 0.055, while using the FFM-based analytical approach given by Eq. (6), DCF values are 0.109, 0.131, and 0.158. which are much lower than those obtained based on FFM, as expected. These results are illustrated in solid lines in Figure 2b. Using Eq. (7) increases the estimated DCF values slightly resulting in 0.109, 0.131, and 0.158 for S_{wi} values of 0.25, 0.35, and 0.45, respectively.

The three cases above (for $S_{wi}=0.25, 0.35, \text{ and } 0.45$) were also modelled using numerical simulation (CMG-GEM 2024) considering different spatial grid sizes. The grid size is set to 2, 20, and 200 m. The reservoir is vertically discretized into 15 layers each with 2-m thickness. The resulting DCFs from different numerical simulations are shown in Figure 2b in comparison with the analytically driven values. The 200-m grid size is clearly resulting in overestimation of DCF.

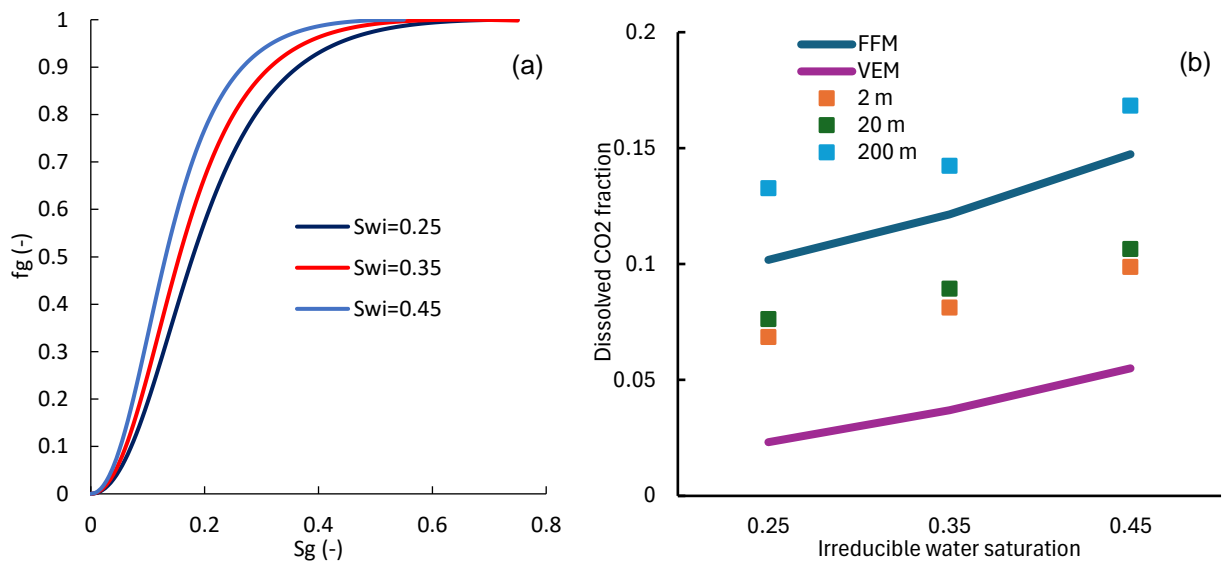


Figure 2. (a) f_g - S_g graph illustrating that increasing S_{wi} result in increased slope of the tangent line drawn from origin, and (b) Analytically-driven DCF estimates (shown by lines) versus those obtained by numerical simulations (shown by squares) with different grid sizes.

Conclusions

In GCS operations, a fraction of the injected CO_2 gets dissolved into the storage aquifer's brine. The dissolved CO_2 fraction (DCF) is often evaluated through numerical modeling simulations where the level of model discretization can significantly affect the estimated DCF. In this study, the author presented two analytical discretization-free approaches to determine the range within which the CO_2 dissolution fraction in the storage aquifer's brine may vary. Values outside of this range may hint that the numerical simulation results are not reliable.

The two approaches were applied to an example CO_2 injection case, and numerical simulations performed to demonstrate the impact of spatial discretization on dissolution rates. For this example case, DCF values obtained by the analytical approaches range between 2.3% and 15% considering irreducible water saturation ranging between 0.25 to 0.45. Numerical simulations with three values of spatial discretization (2, 20, and 200 m) showed that 200-m grid size results in overestimation of DCF. This observation underscore the importance of augmenting numerical simulations findings with analytical methods to provide more consistent and physically realistic estimates of CO_2 dissolution.

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References

- Azizi, Ehsan and Cinar, Yildiray. 2013. Approximate analytical solutions for CO₂ injectivity into saline formations. *SPE Reservoir Evaluation & Engineering* **16** (02): 123-133.
- Burton, McMillan, Kumar, Navanit, and Bryant, Steven L. 2009. CO₂ injectivity into brine aquifers: Why relative permeability matters as much as absolute permeability. *Energy Procedia* **1** (1): 3091-3098.
- CMG-GEM. Computer Modelling Group CMG-GEM Version 2024user's guide. *Computer Modelling Group Ltd.*, <http://www.cmgl.ca/software/gem.htm>.
- Zeidouni, M., Pooladi-Darvish, M., and Keith, D. 2009. Analytical solution to evaluate salt precipitation during CO₂ injection in saline aquifers. *International Journal of Greenhouse Gas Control* **3** (5): 600-611.