Investigations in Geologic Carbon Sequestration: Multiphase Flow of CO_2 and Water in Reservoir Rocks

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Abstract

Understanding the multiphase flow properties of CO_2 and water in porous media is essential for successful large-scale geologic CO_2 storage. Optimizing the design and operation of injection projects will depend on knowledge of injectivity, trapping capacity, distribution of CO_2 in the subsurface and overall areal extent of the subsurface plume. It is increasingly recognized that the capillarity, capillary heterogeneity, and accurate relative permeability curves of CO_2-water systems are essential for accurate simulations ranging from the sub-core to the basin scale. Our research program aims at improving the fundamental understanding on the dynamics of multiphase flow of CO_2 and brine in porous media. The research work integrates experimental investigations and numerical simulations, thus allowing for a direct link between observations in the lab and the physics of the multiphase displacement process. In this context, current available models can be validated and/or extended so as to include a more accurate description of the physical processes involved. Highlights of this year's effort include the following:

- Publication of a complete analysis of the multiphase flow properties of four sandstone rocks with distinct rock lithologies: a Berea sandstone and three reservoir rocks from formations into which CO_2 injection is either currently taking place or is planned. Included in the analysis are drainage relative permeability curves, residual gas saturations over a wide range of initial CO_2 saturations with accompanying Land coefficients, and capillary pressure curves (Krevor et. al., 2012, *Water Resources Research*).
- Publication about the influence of capillary barriers on residual trapping of CO_2. This paper provides the first experimental evidence showing the role of geological heterogeneity on increasing the extent of trapping of CO2 (Krevor et al., 2011, *Geophysical Review Letters*).
- Publication of a new method for making in situ capillary pressure measurements at representative reservoir pressure and temperatures that is significantly faster and more accurate than existing methods, and can also provide sub-core scale measurements of capillary heterogeneity (Pini et al., 2012, *Advances in Water Resources*).
- Publication about multiphase flow and mass transfer processes important for remediation of CO_2 from shallow aquifers (Esposito and Benson, 2012, *International Journal of Greenhouse Gas Control*).
- Publication of a new semi-analytical solution quantifying the influence of gravity, interfacial tension, capillarity, flowrate, and geometry on the multiphase flow of CO_2 and brine (Kuo and Benson, 2012, SPE Conference Paper).
- Publication on approaches for numerical simulation of CO_2 migration in the presence of capillary heterogeneity (Li et. al., 2012, Computational Methods in Water Resources, University of Illinois at Urbana, conference paper).
- Publication of the use of high-precision, mobile CO_2 and CO_2 isotopes for characterization of CO2 fluxes from coal fires (Krevor et al., 2011, *Environmental Science and Technology*).
- Continued improvements in the method we developed to make accurate sub-core scale permeability maps.
- Identification of a new method to enhance oil recovery using exsolution of CO_2 from carbonated water to plug thief zones and improve sweep efficiency of trapped oil and sequester CO_2.

1. Introduction

Carbon dioxide capture and sequestration (CCS) in deep geological formations has emerged over the past fifteen years as an important component of the portfolio of options for reducing greenhouse emissions. Our research focuses on the fundamental science underpinning sequestration in saline aquifers and multiphase flow of CO_2, brine and to a lesser degree, oil. Saline aquifers have the largest sequestration capacity, as compared to oil and gas reservoirs or deep unminable coal beds. Saline aquifers are also more broadly distributed and thus, closer to more emission sources. However, unlike oil and gas reservoirs with proven seals that have withstood the test of time, saline aquifers must be carefully characterized to assure that CO_2 will achieve high retention rates. Improved fundamental understanding of multi-phase flow and trapping in CO_2-brine systems will be needed to take advantage of this large storage capacity of saline aquifers. Important questions remain to be answered, such as, what fraction of the pore space will be filled with CO_2, what will be the spatial extent of the plume of injected CO_2, how much and how quickly will CO_2 dissolve in brine, and how much CO_2 will be trapped by capillary forces when water imbibes back into the plume and to what extent is capillary trapping permanent? How quickly and by which methods can CO_2 leakage into shallow drinking water aquifers be remediated? What are the necessary properties of seals? And, if CO_2 is leaking how can detect this, either deep in the surface or at the land surface. Here we are developing new experimental data and carrying out simulations to improve our ability to answer these questions. As our research progresses, we will assess which, if any, modifications to currently accepted multiphase flow theory are needed and to develop approaches for reliably predicting field-scale performance.

We use a combination of laboratory experiments, numerical methods and analytical solutions to address these issues as shown in the schematic below (Figure 1). Interaction and iteration between these four approaches improves our ability to quickly identify and test new phenomena and approaches for accurately capturing them in quantitative models.

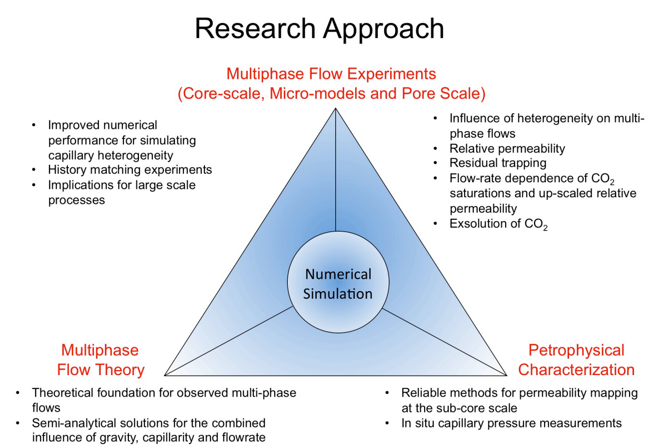

Figure 1. Illustration of the 4 interrelated components of our approach to study multi-phase flow and trapping of CO_2.

2. Core-scale multiphase flow properties of CO_2 and water in sandstone rocks at reservoir conditions

2.1. Enhanced Trapping Due to Capillary Barriers

Simulation studies suggest that substantial amounts of CO_2 can be trapped within permeable sections of a reservoir by capillary forces and intra reservoir heterogeneities [Sadaatpoor et al., 2009], but there is little experimental observation of these phenomena. We have now provided direct experimental evidence for trapping due to capillary heterogeneity, based on the results of CO_2 core flooding experiments conducted at high pressure and temperature to investigate the impact of natural capillary heterogeneity in a sandstone rock on CO_2 saturation buildup and trapping. Carbon dioxide and water were injected through a Mt. Simon sandstone core at 9 MPa pore pressure and 50°C. The core had two regions of distinct capillarity: An upstream 10 cm long region of the core consisted of a relatively high permeability and homogenous sand. A downstream 3 cm long region of the core consisted of a low permeability region characterized by significant cross-bedding and a high capillary entry pressure for CO_2. During a drainage process of CO_2 displacing water, CO_2 builds up upstream of the capillary barrier (see Figure 2). Once in place, CO_2 on the upstream side of the barrier cannot be displaced during 100% water flooding leading to trapped saturations that are a factor 2– 5 times higher than what would be expected from residual trapping alone.

A 2D simulation was designed to further investigate the cause of the saturation buildup. The model was constructed using TOUGH2 and the ECO2N module. In the model, the bulk of the core (Region A and B in Figure 2) is assigned a capillary pressure curve similar to the results of the MICP tests with the lowest entry pressures and porosity of .23. A 1 mm thin barrier between regions A and B is assigned a capillary pressure curve similar to the results of the MICP tests with the highest entry pressure. The relative permeability of both regions was assigned a curve fit to the data from the upstream region. The permeabilities of both the bulk core (regions A and B) and the 1 mm barrier region were varied so the CO_2 saturation profile and the pressure drop across the core were matched for both 100% water flooding and 50% CO_2 flooding.

Simulation results are shown in Figure 2. The characteristic saturation buildup and good matches for the pressure drop across the core could be obtained if the bulk core and barrier had permeabilities of 7.5 Darcy and .01 mD respectively. While this is an exceedingly simple representation of the core, the results indicate that the rock is characterized by bulk regions of high permeability and low capillary entry pressures and thin bedding planes with low permeabilities and high capillary entry pressures. The measured permeabilities are a result of the combined contribution of these regions. While there was clearly a large barrier at the start of the downstream cross-bedded region of the core, here were likely also several low permeability planes in the upstream region (see clear bedding planes at x = .5 in Figure 2). This is probably in part the cause of the nonmonotonic saturation profile upstream of the major barrier.

Hesse and Woods [2010], Green and Ennis- King [2010], and Mouche et al. [2010] have developed methods for upscaling the impact of small-scale heterogeneities on the buoyant flow and residual trapping of CO2 in a brine saturated system to the reservoir scale [Hesse and Woods, 2010; Green and Ennis- King, 2010; Mouche et al., 2010]. They show that among other factors, saturation buildup beneath intra-reservoir flow barriers is highly dependent on the lateral extent and vertical distribution of heterogeneities. The observations made in this study suggest that this analysis should be extended beyond the consideration of mudrock distribution to the impact that depositional structures and diagenetic horizons will also play in the movement and steady-state distribution of a CO_2 plume. In addition, where buoyant CO_2 exists beneath the spill point of intra-reservoir heterogeneities, it should be considered immobile in the absence of recharge from continued injection.

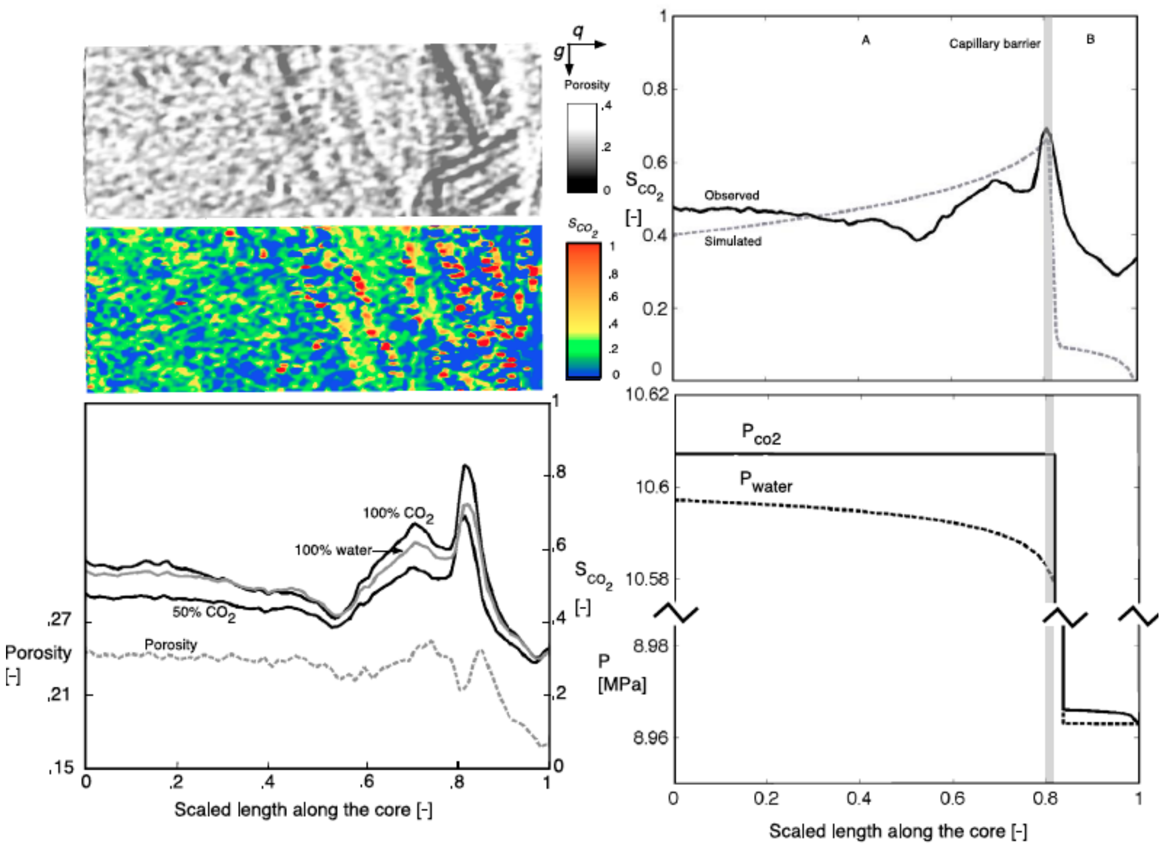

Figure 2. (left, top) A porosity map on a plane through the center of the core shows significant cross-bedding at the downstream end. (left, middle) A map of CO_2 saturation on the same plane during 100% water flooding following 100% CO_2 flooding. (left, bottom) Plots of average porosity and average CO_2 saturation along the length of the core during different fractional flows of CO_2 and during 100% water flooding. (right, top) A comparison of simulated and observed CO_2 saturation along the length of the core during 50% CO_2 flooding. (right, bottom) Water and CO_2 pressure along the length of the rocks for the simulation results.

2.2. In situ Measurements of Capillary Pressure and Capillary Heterogeneity

A novel method has been developed to measure drainage capillary pressure curves both at the core and sub-core scale using CO_2 and water at reservoir conditions. The experimental configuration is very similar to the one used for traditional steady-state relative permeability experiments. Capillary pressure measurements are made at the inlet face of the sample by successively increasing the flow rate of the non-wetting phase while measuring the saturation with a medical X-ray Computed Tomography (CT) scanner (Figure 3). The method requires that the wetting phase pressure is uniform across the core and can be measured in the outlet end-cap. A capillary pressure curve is obtained in less than two days, as compared to weeks for existing methods that use porous plates.

We have demonstrated this technique by measuring drainage capillary pressure curves of CO_2 and water for two sandstones rock cores with different lithology and pore size distribution (Figure 4). Experiments are carried out at 25 and 50 ^{o}C and at 9 MPa pore pressure, while keeping the confining pressure on the core at 12 MPa. There is excellent agreement between the new method and data from mercury intrusion porosimetry; beside providing confidence in the new technique, such comparison allows for an estimate of the wetting and interfacial properties of the CO_2/water system.

X-ray CT scanning allows for precise imaging of fluid saturations at a resolution of about (2.5 x 2.5 x1) mm^3, thus enabling quantification of sub-core scale capillary pressure curves (Figure 5). These measurements provide independent confirmation that sub-core scale capillary heterogeneity plays a critical role in controlling saturation distributions during multiphase flow.

The implications of small-scale capillary heterogeneity on CO_2 plume movement, solubility trapping and capillary trapping are being investigated. However, in order to carry out these studies we need to develop improved computational tools for carrying out this research. Progress in this regard was reported last year and results have now been submitted as a conference publication (Li et al., 2012).

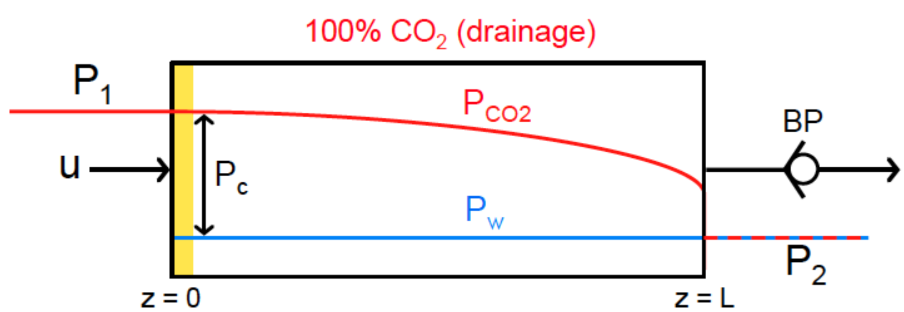

Figure 3. Schematic showing the configuration of the experimental setup for the capillary pressure measurements.

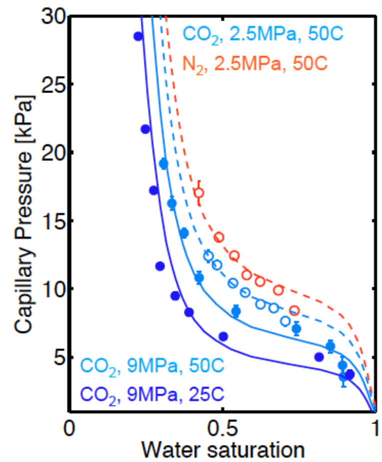

Figure 4. Capillary pressure measurements made for CO_2/water and N_2 water at a range of in situ conditions. The circles are the measurements made using this new technique. The solid lines show the curves measured using mercury injection and normalized to the CO_2/brine system using the interfacial tension based on surface tension data from the literature.

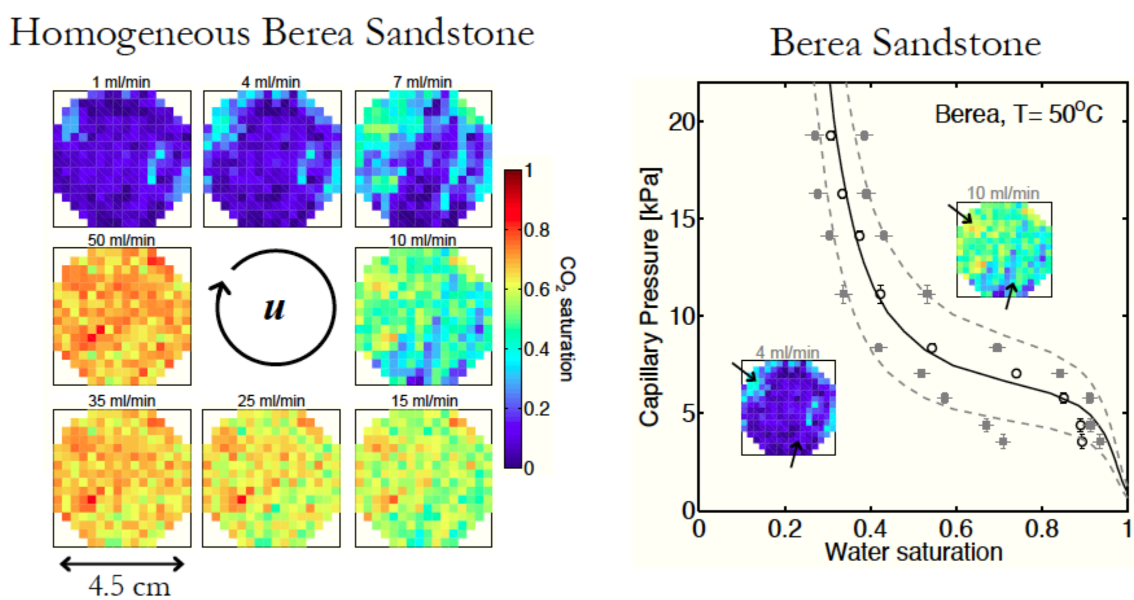

Figure 5. Illustration of the data used for constructing sub-core scale capillary pressure curves and the capillary pressure curves for two pixels.

2.3 Influence of Capillary Pressure Treatment on Solubility Trapping

Capillary pressure is one of the major driving forces in long-term CO_2 sequestration. Whether to use a plateau or a slope to represent the nonwetting-phase entry distinguishes a convex capillary pressure model (e.g., Brooks-Corey model) from an S-shaped model (e.g., van Genuchten model). It is also an important issue in interpreting the capillary pressure curves measured from mercury porosimetry. Although we use a steep `entry-slope' in the S-shaped model to reduce the dissimilarity between the two models, and indeed such slight difference in entry-pressure representations has little effect on simulations that do not model dissolution, it is not the case in long-term CO_2 sequestration simulations where CO_2 dissolution is included. For example, Figure 6 provides a comparison between size of the plume and the plume volume for 4 different treatments of the entry pressure. As shown, the distance to the tip of the plume varies by nearly a factor of 2, and the plume volume differs by even a greater amount. We have learned that S-shaped capillary pressure models can greatly facilitate the dissolution of the CO_2 plume, especially in deep storage formations that have large vertical permeabilities and small critical gas saturation values.

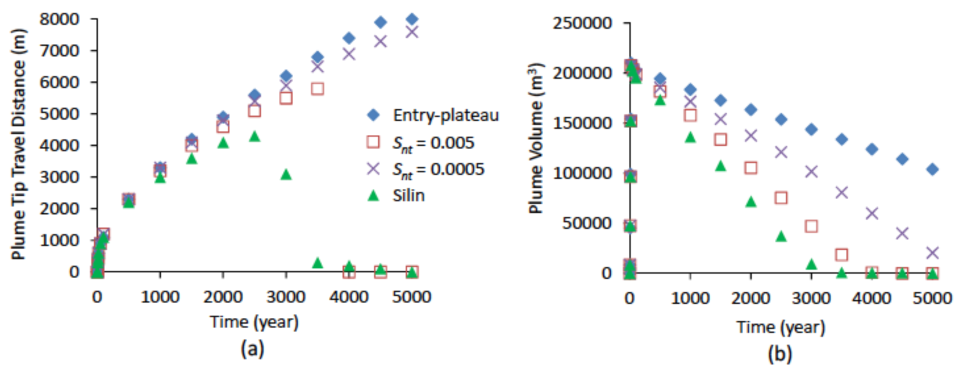

Figure 6. Comparison of the distance to the plume tip and plume volume for 4 different treatments of the capillary entry pressure. Note the wide variation depending on the treatment.

More work is needed to assess the most appropriate treatment for the entry pressure. However, using a convex capillary pressure model is more conservative in estimating the rate that the CO_2 plume will dissolve in the formation water. It also makes the simulation to run faster and reduces the sensitivity of simulation results. In this case, the simulator needs to allow for a "virtual" capillary pressure in wetting-phase-saturated regions. A manuscript describing this work is in preparation (Li et al., 2012).

2.4. Semi-Analytical Solutions for Predicting CO_2 Saturations as a Function of Flowrate, Interfacial Tension, and Core Geometry

We have developed an approximate semi-analytical solution for predicting the average steady-state saturation during multiphase core flood experiments over a wide range of capillary and gravity numbers. Last year we reported on the influences of flow rate, gravity, and sub-core heterogeneity on the brine displacement efficiency using the 3-D simulator TOUGH2. These studies have demonstrated that the average saturation depends on the capillary and gravity numbers in a predictable way. The purpose of this work is to provide a simple approximate semi-analytical solution for predicting the average saturation during core flood experiments, thus avoiding the need for 3-D simulations. A two dimensional analysis of the governing equations for the CO_2/brine multiphase flow system at steady-state is used to develop the approximate semi-analytical solution. We have developed a new criterion to identify the viscous-dominated regime at core scale. Variations of interfacial tension, core permeability, length of the core, and the effects of buoyancy, capillary and viscous forces are all accounted in the theoretical solutions. We have also shown that three dimensionless numbers (N_B, N_{gv}, R_l) and two critical gravity numbers ($N_{gv,c1}$, $N_{gv,c2}$) are required to properly capture the balance of viscous, gravity, and capillary forces. As illustrated for 4 different fractional flows in Figure 7, there is good agreement of the average saturations between the 3-D simulations and the model. This new model can be used to design and interpret multiphase flow core-flood experiments. This work was presented at a conference is and is now under review for publication in a peer reviewed journal (Kuo and Benson, 2012).

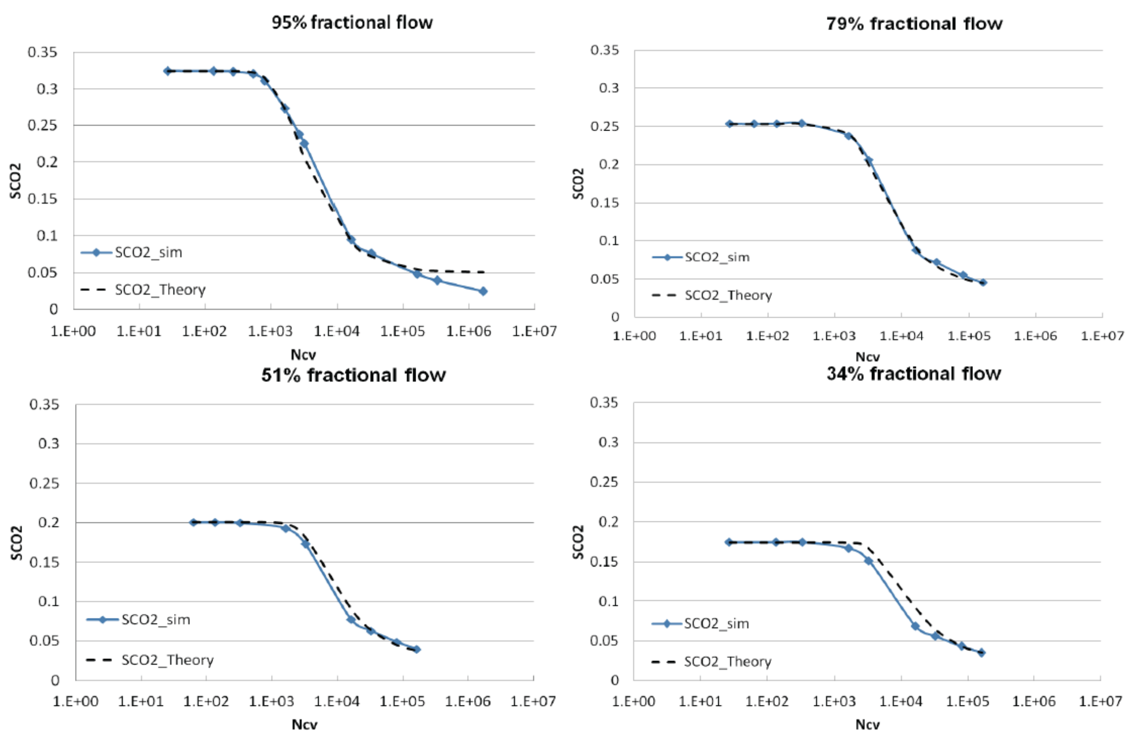

Figure 7. Comparison between the semi-analytical solution and simulated CO_2 saturations over a wide range of capillary numbers and fractional flows.

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Benson, S.M. ,"Recent Advance in CO_2 Storage," U.S.-Norway Science Week, Berkeley, California.

Benson, S.M., "Remediation Methods for CO_2 Leakage," U.S. Department of Energy R&D Workshop on Storage on Saline Aquifers, Pittsburg, Pennsylvania.

Benson, S.M., "Contingency Planning and Methodologies for Intervention," International Petroleum Institute for Environmental and Social Issues (IPIECA) Workshop on Carbon Capture and Storage, Washington, DC.

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