Title: Investigation of uncertainty in CO₂ reservoir models: a sensitivity analysis of relative permeability parameter values

Authors: Nozomu Yoshida^{1,2}, Jonathan S. Levine³, Philip H. Stauffer¹

¹Earth and Environmental Sciences Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

²Department of Petroleum Engineering, Texas A&M University, College Station, TX, 77843, USA

³U.S. Department of Energy, National Energy Technology Laboratory, Pittsburgh, PA 15236, USA

Emails:

Nozomu Yoshida, noz.yoshida@tamu.edu

Jonathan Levine, jonathan.levine@netl.doe.gov

Philip Stauffer, stauffer@lanl.gov

Corresponding author during refereeing:

Jonathan Levine

jonathan.levine@netl.doe.gov

412-386-7534

Graphical abstract

Abstract

Numerical reservoir models of CO₂ injection in saline formations rely on parameterization of laboratory-measured pore-scale processes. We performed a parameter sensitivity study and Monte Carlo simulations to determine the normalized change in total CO₂ injected using the finite element heat and mass-transfer code (FEHM) numerical reservoir simulator. Experimentally measured relative permeability parameter values were used to generate distribution functions for parameter sampling. The parameter sensitivity study analyzed five different levels for each of the relative permeability model parameters. All but one of the parameters changed the CO₂ injectivity by <10%, less than the geostatistical uncertainty that applies to all large subsurface systems due to natural geophysical variability and inherently small sample sizes. The exception was the end-point CO₂ relative permeability, k_{r,CO_2}^0 , the maximum attainable effective CO₂ permeability during CO₂ invasion, which changed CO₂ injectivity by as much as 80%. Similarly, Monte Carlo simulation using 1000 realizations of relative permeability parameters showed no relationship between CO₂ injectivity and any of the parameters but k_{r,CO_2}^0 , which had a very strong ($R^2=0.9685$) power law relationship with total CO₂ injected. Model sensitivity to k_{r,CO_2}^0 points to the importance of accurate core flood and wettability measurements.

Keywords: carbon sequestration; reservoir modeling; relative permeability; sensitivity analysis

1 Introduction

Geological carbon sequestration (GCS), the injection of carbon dioxide into the deep subsurface, requires accurate numerical reservoir models to predict CO₂ injection rates and quantities. Accurate reservoir modeling has importance to a variety of stakeholders, each having their own information needs. At the national and state scales, policy makers require accurate storage resource estimates to have confidence that sufficient pore space is available for GCS to address national or state greenhouse gas emissions (Goodman et al. 2011). At the field and reservoir scales, commercial operators need accurate reservoir models to predict operational parameters such as number of injection wells, pore space requirements, leakage risks, and areas of review (Dai et al. 2014). The United States Department of Energy (DOE) has a stated goal to “support industry’s ability to predict CO₂ storage capacity in geologic formations to within $\pm 30\%$ ” (US-DOE NETL 2012). Improvements in our understanding of the relative importance of physical parameters will help reduce uncertainty in storage capacity estimates by allowing resources to be focused on making the most cost-effective measurements.

The relative permeabilities of brine and CO₂ are key parameters that set the injectivity of CO₂ into deep saline aquifers. Relative permeability is a multiphase rock-fluid property accounting for the

fractional reduction in fluid conductivity due to blockage of fluid paths by the presence of one or more additional fluid phases. Relative permeability values therefore range between 0 and 1. Relative permeability is a strong function of the phase saturation as well as a function of rock properties such as pore size distribution and wettability (Lake 2010).

The wetting state, e.g., as measured by the contact angle, of a fluid-fluid-rock system can predict the relative permeability curve. More specifically, the relative permeability of each fluid phase is determined by pore- and sub-pore- scale fluid distributions of each continuous fluid phase, e.g. (Willhite 1986, Anderson 1987). At the sub-pore scale, fluids aim to minimize surface energy; thus rock-wetting fluids maximize rock surface contact area and occupy the smallest pores and coat rock surfaces. By comparison, nonwetting fluids will have minimized contact with rock surfaces and therefore will occupy the largest pores and the center of pores. The resulting capillarity-controlled fluid distributions produce significantly different relative permeabilities when measured at core and reservoir scales, with nonwetting fluids having much higher relative permeabilities than wetting fluids for the same saturation values. E.g. the end-point relative permeability, the maximum achievable relative permeability of an invading phase, is related to the wetting state, with values below 0.2 typical of strongly wetting fluids and values greater than 0.7 for strongly nonwetting fluids. The end-point relative permeability can therefore be used to predict the wetting state. This connection between rock surface chemistry, rock-fluid and fluid-fluid surface energies, capillarity, pore scale fluid occupancy, fluid saturations, and relative permeability is the theoretical core of pore- and reservoir-scale multiphase fluid properties and petrophysics. Additional details and a complete literature review on this subject with application to GCS can be found in Levine (2011) and Levine et al. (2014).

Iglauer et al. (2015) provided an excellent and thorough review of the problematic and entirely unresolved state of GCS contact angle experiments and thus wettability. As a result, theoretical wettability-derived relative permeability curves cannot be assigned, and instead GCS models rely on experimentally measured relative permeability values.

To quantify the relative permeability relationship between brine and CO₂ for different types of formations, experimental measurements have been conducted under extensive experimental conditions. In an early work, Dria et al. (1993) conducted steady-state three-phase CO₂/oil/brine relative permeability experiments with a carbonate core. Bennion and Bachu (2005) published unsteady-state experimental results of supercritical CO₂ injection into cores from sandstone and carbonate formations in western Canada. More extensive experimental results were presented in a series of publications: Bennion and Bachu (2008), carbonate formations in Bennion and Bachu (2010) and sandstone formations in Bachu (2013). Perrin et al. (2009) conducted steady-state CO₂-brine core flooding experiments using x-ray CT to

determine saturation distribution. These experimental results show low values of end-point CO₂ relative permeability and high values of residual water saturation under drainage condition.

Levine (2011) and Levine et al. (2014) report experimental measurements of end-point CO₂ relative permeability using the method proposed by Ramakrishnan and Cappiello (1991), which does not require the achievement of a uniform saturation within the core. Both Levine (2011) and Levine et al. (2014) contain reviews of experimental artifacts in CO₂ core flood experiments, including insufficiently elevated capillary pressures to achieve endpoint relative permeability, capillary end effects, and gravitational/buoyancy-induced segregation in horizontal core flood experiments. Unlike previous experiments, this dataset was solely focused on measuring end-point drainage CO₂ relative permeability, with particular attention paid to avoiding experimental artifacts. CO₂ end-point permeabilities in this dataset were clustered around 0.35-0.4.

Krevor et al. (2012) suggested that residual saturations persist irrespective of significant heterogeneities and in the absence of significant gravity segregation. They also reported that the maximum observed saturations in their experiment were limited by the maximum capillary pressure possible given the experimental setup. Pini and Benson (2013) further argued that the low end-point CO₂ relative permeability was caused by the low viscosity of CO₂ relative to water rather than by the rock heterogeneity. Most of the experimental core flood data sets suggest that the end-point relative permeability to CO₂ is low (<0.5) and that the residual water saturation is high (>0.4), though further investigation is required. Additional CO₂ relative permeability experiments have since been reported but were not available in time for the initiation of the present effort.

Relative permeability depends on a variety of parameters. Pressure, temperature and brine salinity are treated as primary independent variables, while the viscosity ratio and interfacial tension (IFT) are secondary variables (Bachu and Bennion 2008, 2009). Multiphase interference is indicated by the curvature of the relative permeability curve, and less-concave relative permeability curves are obtained in experiments with lower IFT (Bennion and Bachu 2006b) or higher porosity and permeability due to higher median pore size (Bennion and Bachu 2010). Middleton et al. (2012) showed that IFT variation can impact estimates of reservoir capacity by about 10%. In carbonate reservoirs, higher permeability units caused by channeling and bypassing of tighter portions of the pore system can contribute to increased macro porosity and reduction of the irreducible CO₂ saturation (Bennion and Bachu 2010). Drainage/imbibition hysteresis also affects the shape of the relative permeability function and is related to interfacial areas between fluids (Juanes et al. 2006, Bennion and Bachu 2008, Porter et al., 2010).

Compared to CO₂ injection for enhanced oil recovery, GCS sites often have less detailed information about reservoirs where CO₂ will be injected (Stauffer et al. 2009; Dai et al., 2014). For these cases, uncertainty quantification on CO₂ injectivity and storage capacity has been performed on parameters

related to geologic heterogeneity (Deng et al. 2012, Heath et al. 2012). Burton et al. (2009) investigated the importance of uncertainty in relative permeability on CO₂ injectivity and found a four-fold variation in injectivity with different relative permeability curves. Burton et al. (2009) also report that the end-point CO₂ relative permeability and the curvature of the aqueous phase relative permeability curve can change the injection rate by 20%. Kopp et al. (2009a) performed dimensional analysis using the capillary and gravity numbers to investigate CO₂ storage capacity in saline aquifers. They found that the relative permeability-saturation relationships significantly influence plume evolution, velocity, and average CO₂ saturation, with a still larger influence on storage capacity. Further investigation by Kopp et al. (2009b) indicated that the CO₂ relative permeability associated with the residual brine saturation is a major influence on the average CO₂ saturation in reservoirs, whereas the brine relative permeability has a major influence on the geometric capacity coefficient. Kopp et al. (2009b) concluded that relative permeability relationships influence the capacity estimates to a similar extent as the entire range of reservoir properties such as geothermal gradient and depth.

Mathias et al. (2013) investigated the uncertainty of the relative permeability data on the estimation of CO₂ injectivity for brine aquifers with 25 published relative permeability datasets. Their work employed a semi-analytical solution that assumes negligible capillary pressure, constant fluid properties, and vertical pressure equilibrium. They showed that for the 25 samples examined, there is no clear relation between the relative permeability data and porosity, permeability, IFT or lithology. They evaluated CO₂ injectivity through the assessment of the maximum constant CO₂ injection rate that can be sustained for 30 years without exceeding hydrofracture pressure. Finally, they concluded that while uncertainty in relative permeability relations can have a substantial effect on injectivity estimation for open aquifers, for closed aquifers, the effects associated with formation compressibility play a more important role. In a related work, González-Nicolás et al., (2015) use another semi-analytical model (with similar limiting assumptions) to explore global sensitivity of CO₂ injection and leakage to changes in a set of parameters that include brine residual saturation and CO₂ end-point relative permeability. Their findings showed second order effects for brine residual saturation (and the functionally linked CO₂ end-point relative permeability). However, the global parameter set includes degrees of freedom that are unrelated to the relative permeability functions, thus making it potentially more difficult to extract information on individual impacts of the relative permeability functions alone. For these reasons, relative permeability remains a source of considerable uncertainty in numerical simulations of geologic CO₂ sequestration.

The objective of this work is to evaluate the importance of relative permeability relationships on geologic CO₂ sequestration injectivity in more detail than previous studies, including a robust statistical analysis of the sensitivity of injectivity to each relative permeability model parameter for simulations with full thermodynamic coupling of the mass and energy equations, including impacts of capillary pressure,

variable fluid properties, and non-equilibrium vertical pressure gradients. After a brief review of a relative permeability model and available published experimental datasets, we present numerical simulations to investigate the importance of each parameter in the relative permeability model on CO₂ injectivity. This study focuses on uncertainty resulting from a select set of parameters (e.g. relative permeability parameters). While there are many other uncertain parameters in a GCS system that can be varied such as intrinsic permeability, and reservoir heterogeneity, we focus on exploring uncertainty in the fluid retention parameters. To this end, we consider the case of supercritical CO₂ injection into a 2D cylindrical saline reservoir using the finite-element heat- and mass-transfer code (FEHM) developed by Zyvoloski et al. (2011). A parameter sensitivity study is performed with a relative permeability model based on the distribution of experimental data. Further investigation is conducted using a Monte Carlo simulation method with statistically-estimated properties of each relative permeability parameter.

2 Methods

2.1 Relative permeability for CO₂–brine

The multiphase Darcy equation used in FEHM is:

$$\mathbf{v}_l = -\frac{kk_{r,l}}{\mu_l}(\nabla p_l - \rho_l \mathbf{g}) \quad (1)$$

where $\mathbf{v}_l$ is the volumetric flux of phase l ($l = \text{CO}_2$ or brine), k is permeability, $k_{r,l}$ is the relative permeability of phase l , μ_l is the viscosity of phase l , p_l is phase pressure, and $\mathbf{g}$ represents the acceleration due to gravity (Zyvoloski et al. 2011). The relative permeability model (Lake 2010) used in this work is a variation of the Corey equations:

$$k_{r,\text{CO}_2} = k_{r,\text{CO}_2}^0 \left(\frac{S_{\text{CO}_2} - S_{\text{CO}_2,ir}}{1 - S_{\text{CO}_2,ir} - S_{\text{brine},ir}} \right)^m \quad (2)$$

and

$$k_{r,\text{brine}} = k_{r,\text{brine}}^0 \left(\frac{S_{\text{brine}} - S_{\text{brine},ir}}{1 - S_{\text{CO}_2,ir} - S_{\text{brine},ir}} \right)^n \quad (3)$$

where $k_{r,l}^0$ is the end-point relative permeability of phase l , S_l is the saturation of phase l , $S_{l,ir}$ is the residual or irreducible saturation of phase l , and m and n are the curvature exponents of CO₂ and brine, respectively. This work only considers the drainage process, therefore $k_{r,\text{brine}}^0$ and $S_{\text{CO}_2,ir}$ are set to 1 and 0, respectively. Therefore, the relative permeability model is simplified as:

$$k_{r,\text{CO}_2} = k_{r,\text{CO}_2}^0 \left(\frac{S_{\text{CO}_2}}{1 - S_{\text{brine},ir}} \right)^m = k_{r,\text{CO}_2}^0 \left(\frac{1 - S_{\text{brine}}}{1 - S_{\text{brine},ir}} \right)^m \dots\dots\dots (4)$$

and

$$k_{r,\text{brine}} = \left(\frac{S_{\text{brine}} - S_{\text{brine},ir}}{1 - S_{\text{brine},ir}} \right)^n \dots\dots\dots (5)$$

The remaining parameters, k_{r,CO_2}^0 , $S_{\text{brine},ir}$, m and n , are determined by regression analysis with measured experimental data. Formation unit, source of the data, lithology, porosity, and experimental conditions (pressure, temperature and IFT) are summarized in

TABLE 1, and the corresponding experimental results and their statistical parameter values are summarized in **TABLE 2**. These tables are a summary of the published experimental results. Bennion and Bachu (2008, 2010) and Bachu (2013) used the variation of the Corey equations for fitting their experimental data. Mathias et al. (2013) conducted least-square fittings of the Perrin and Benson (2010) and Krevor et al. (2012) data to aid comparison with Bennion and Bachu (2008, 2010). We use the estimated relative permeability parameters in Mathias et al. (2013) for the data of Perrin and Benson (2010) and Krevor et al. (2012). The relative permeability curves given by the experimental work are shown in **Figure 1** as well.

TABLE 1

Summary of formation unit, source of the data, lithology, porosity, and experimental conditions (pressure, temperature and IFT) from the published literature.

Formation Unit	Source	Lithology	Porosity (%)	Pressure (MPa)	Temperature (°C)	IFT (mN/m)
Otway	Perrin and Benson (2010)	Sandstone	18.2	12.40	63	30.0
Berea #1	Perrin and Benson (2010)	Sandstone	20.3	12.40	50	30.0
Berea #2	Krevor et al. (2012)	Sandstone	22.1	9.00	50	32.0
Paaratte	Krevor et al. (2012)	Sandstone	28.3	9.00	50	32.0
Mt. Simon	Krevor et al. (2012)	Sandstone	24.4	9.00	50	32.0
Tuscaloosa	Krevor et al. (2012)	Sandstone	23.6	9.00	50	32.0
Cardium #1	Bennion and Bachu (2008)	Sandstone	15.3	20.00	43	19.8
Cardium #2	Bennion and Bachu (2008)	Sandstone	16.1	20.00	43	19.8
Viking #1	Bennion and Bachu (2008)	Sandstone	12.5	8.60	35	32.1
Viking #2	Bennion and Bachu (2008)	Sandstone	19.5	8.60	35	32.1
Viking #3	Bachu (2013)	Sandstone	17.2	8.60	35	32.1
Ellerslie #1	Bennion and Bachu (2008)	Sandstone	12.6	10.90	40	32.5
Ellerslie #2	Bachu (2013)	Sandstone	29.0	10.90	40	32.5
Basal Cambrian #1	Bennion and Bachu (2008)	Sandstone	11.7	27.00	75	27.0
Basal Cambrian #2	Bachu (2013)	Sandstone	11.6	27.00	75	35.6
Basal Cambrian #3	Bachu (2013)	Sandstone	11.9	18.00	58	29.5
Basal Cambrian #4	Bachu (2013)	Sandstone	11.9	20.33	60	30.2
Basal Cambrian #5	Bachu (2013)	Sandstone	12.5	27.00	75	35.6
Clearwater	Bachu (2013)	Sandstone	33.1	2.00	20	56.3
Rock Creek	Bachu (2013)	Sandstone	14.5	15.40	75	32.9
Halfway	Bachu (2013)	Sandstone	17.7	21.80	74	34.6
Belloy	Bachu (2013)	Sandstone	23.6	13.50	56	33.1
Graminia	Bachu (2013)	Sandstone	31.6	9.60	39	35.1
Gilwood	Bachu (2013)	Sandstone	11.5	16.80	46	35.7
Deadwood #1	Bachu (2013)	Sandstone	17.6	15.24	47	36.1
Deadwood #2	Bachu (2013)	Sandstone	16.2	23.61	65	30.3
Deadwood #3	Bachu (2013)	Sandstone	19.3	22.30	61	31.9
Granite Wash	Bachu (2013)	Sandstone	14.8	14.66	54	37.0
Webamun #1	Bennion and Bachu (2008)	Carbonate	7.9	11.90	41	29.5
Webamun #2	Bennion and Bachu (2008)	Carbonate	14.8	11.90	41	29.5
Webamun #3	Bennion and Bachu (2010)	Carbonate	15.4	11.90	41	29.5
Nisku #1	Bennion and Bachu (2008)	Carbonate	9.7	17.40	56	34.6
Nisku #2	Bennion and Bachu (2010)	Carbonate	10.4	17.40	56	34.6
Nisku #3	Bennion and Bachu (2010)	Carbonate	10.9	17.40	56	34.6
Grosmont	Bennion and Bachu (2010)	Carbonate	11.8	11.90	41	34.2
Morinville Leduc	Bennion and Bachu (2010)	Carbonate	11.6	11.40	40	33.1
Redwater Leduc	Bennion and Bachu (2010)	Carbonate	16.8	9.20	36	35.1
Cooking Lake #1	Bennion and Bachu (2008)	Carbonate	9.9	15.40	55	35.7
Cooking Lake #2	Bennion and Bachu (2010)	Carbonate	16.7	15.50	55	35.7
Slave Point	Bennion and Bachu (2010)	Carbonate	9.9	18.80	43	29.5
Winnipegosis	Bennion and Bachu (2010)	Carbonate	14.8	8.73	36	45.3

TABLE 2

Summary of experimental results with regression analysis for the power law relative permeability model.

Formation Unit	Lithology	Permeability (md)	$S_{brine,ir}$	k_{r,CO_2}^0	m	n
Otway	Sandstone	45	0.4370	0.6594	6.5	2.5
Berea #1	Sandstone	430	0.5890	0.0700	1.3	2.5
Berea #2	Sandstone	914	0.4438	0.3948	2.6	3.2
Paaratte	Sandstone	1156	0.3894	0.3284	3	4.6
Mt. Simon	Sandstone	7.5	0.4371	0.4929	1.6	6
Tuscaloosa	Sandstone	220	0.7030	0.0767	3.2	4.7
Cardium #1	Sandstone	0.356	0.1970	0.5260	1.7	1.3
Cardium #2	Sandstone	21.17	0.4250	0.1290	1.3	1.2
Viking #1	Sandstone	2.7	0.5580	0.3319	3.2	2.9
Viking #2	Sandstone	21.72	0.4230	0.2638	2.8	1.7
Viking #3	Sandstone	1558.65	0.6010	0.0973	4.34	1.33
Ellerslie #1	Sandstone	0.376	0.6590	0.1156	2.2	2.1
Ellerslie #2	Sandstone	3812.36	0.3820	0.5735	4.79	1.18
Basal Cambrian #1	Sandstone	0.081	0.2940	0.5446	5	1.8
Basal Cambrian #2	Sandstone	0.0057	0.5690	0.2105	3.89	1.45
Basal Cambrian #3	Sandstone	252.50	0.4900	0.1562	1.35	1.63
Basal Cambrian #4	Sandstone	157.80	0.6510	0.2100	3.74	4.54
Basal Cambrian #5	Sandstone	0.030	0.2750	0.3255	5.48	1.21
Clearwater	Sandstone	0.0164	0.3430	0.4939	1.60	1.24
Rock Creek	Sandstone	65.03	0.4790	0.0434	1.90	2.19
Halfway	Sandstone	54.23	0.4660	0.2733	3.48	3.12
Belloy	Sandstone	536.60	0.6530	0.0762	5.22	1.67
Graminia	Sandstone	133.90	0.4420	0.1461	4.98	1.42
Gilwood	Sandstone	0.749	0.5655	0.5454	3.73	1.75
Deadwood #1	Sandstone	103.66	0.4897	0.1062	7.00	1.80
Deadwood #2	Sandstone	69.11	0.5959	0.0941	4.00	1.50
Deadwood #3	Sandstone	137.90	0.6540	0.2597	6.57	1.20
Granite Wash	Sandstone	70.13	0.5789	0.4050	1.81	1.15
Webamun #1	Carbonate	0.018	0.5950	0.5289	5.6	1.4
Webamun #2	Carbonate	66.98	0.5690	0.1883	2.1	1.4
Webamun #3	Carbonate	54.3	0.8520	0.1015	5.78	2.76
Nisku #1	Carbonate	45.92	0.3300	0.1768	1.1	2.8
Nisku #2	Carbonate	21.02	0.4920	0.0999	4.6	2.7
Nisku #3	Carbonate	74.4	0.3970	0.1078	2.93	2.27
Grosmont	Carbonate	153.9	0.5200	0.1101	5.19	1.73
Morinville Leduc	Carbonate	371.9	0.5300	0.0746	3.67	1.82
Redwater Leduc	Carbonate	353.6	0.6650	0.0476	4.78	1.57
Cooking Lake #1	Carbonate	65.3	0.4760	0.0685	5.6	1.4
Cooking Lake #2	Carbonate	4.87	0.5963	0.0940	5.44	1.83
Slave Point	Carbonate	0.217	0.5460	0.5037	1.16	1.56
Winnipegosis	Carbonate	3.09	0.2108	0.6117	4.5	2.4
Mean	—	268.0	0.502	0.260	3.676	2.159
Standard Deviation	—	655.3	0.1367	0.1908	1.697	1.109
Variance	—	4.294×10^5	0.0187	0.0364	2.881	1.231

Figure 1—Relative permeability curves from the available literature (solid line denotes brine relative permeability and dashed line denotes CO₂ relative permeability)

Figure 1—Relative permeability curves from the available literature (solid line denotes brine relative permeability and dashed line denotes CO₂ relative permeability) (continued)

Figure 2 shows histograms of log permeability and relative permeability parameters ($S_{brine,ir}$, k_{r,CO_2}^0 , m and n), and scatter plots generated from the published experimental data. The green and red data points denote the results given by the sandstone and carbonate samples, respectively. These scatter plots show no obvious relations between the variables, though the dataset generates some clusters and $S_{brine,ir}$ and k_{r,CO_2}^0 seem to correlate slightly; it is also noted that the sandstone and carbonate results do not show a clear difference, in agreement with observations made by Mathias et al., (2013). In the following sections, we assume the intrinsic permeability and the relative permeability parameters as independent variables based on these observations. It is noted that assuming statistical independence may lead to overestimating the interval of variability of CO₂ mass storage, an impact that is more pronounced for strongly correlated variables.

Figure 2—Scatter plots and histograms of intrinsic permeability and relative permeability model parameters generated from published experimental data

2.2 Numerical simulation model

We employ a fully coupled heat and mass conservation formulation in our simulations to preserve conservation of the full energy balance including changes in gravitational potential (Stauffer et al. 2014). In this work, a 2-D cylindrical, homogeneous and isotropic saline reservoir is considered. The reservoir is overlain by a cap rock and underlain by a bed rock formation with low permeability (10^{-19} m^2). FEHM is used to simulate injection of supercritical CO_2 into the reservoir under non-isothermal conditions. Supercritical CO_2 is injected into the reservoir for 4 years with a constant injection pressure and temperature (16.5 MPa, 36 °C) at the bottom of the wellbore. Because the simulation is fully coupled, viscosity, density, and pressures continue to change throughout the simulations resulting in a variable injection rate. As in Levine et al. (2013), the modeled processes are essentially time independent meaning that the results scale with the CO_2 mass injected. The wellbore diameter is 0.275 m, and the well is

perforated across the entire 20 m thick permeable injection reservoir. The reservoir fluids are assumed to be mutually saturated, i.e. dissolution between the CO₂ and water phases is ignored. The CO₂ and water thermophysical properties of density, viscosity and enthalpy are estimated based on data from the NIST chemistry webbook (Lemmon et al. 2005). Other parameters used in the simulation are summarized in **TABLE 3**, and schematics of the model geometry are shown in **Figure 3**. The reservoir outer radius and thicknesses of the caprock, reservoir and bedrock are 20 km, 40 m, 20 m and 40 m, respectively. The reservoir is discretized into 40401 nodes with a logarithmically-spaced grid in the r -direction and uniform spacing (0.5 m) in the z -direction. Constant pressure and temperature conditions were set for the top and bottom boundaries of the model: pressure is initial hydrostatic equilibrium with the reservoir and upper and lower boundary temperatures of 36 °C and 37 °C, respectively. Pressure and temperature are set to an initial hydrostatic constant at the reservoir's 20 km radial boundary to simulate an open connection to an infinite aquifer.

TABLE 3
Reservoir parameters used in this work.

Parameter	Value
Cap/bed rock permeability, m ²	10 ⁻¹⁹
Saline reservoir permeability, m ²	10 ⁻¹³
Porosity, -	0.1
Rock bulk density, kg/m ³	2563
Rock bulk compressibility, 1/MPa	1.0 × 10 ⁻⁹
Rock specific heat, J/(kg·K)	1010
Rock thermal conductivity, W/(m·K)	1.0
Wellbore diameter, m	0.275
Initial reservoir pressure, MPa	11
Initial reservoir temperature, °C	36
Reservoir thickness, m	20

Figure 3—Schematics of reservoir geometry

The capillary pressure model used in this work is the van Genuchten model (1980):

$$P_c = P_0 \left[(S^*)^{-\frac{1}{\lambda}} - 1 \right]^{1-\lambda} \quad (6)$$

where λ is the pore size distribution index, and the normalized brine saturation, S^* , is given by:

$$S^* = \frac{S_{brine} - S_{brine,ir}}{1 - S_{brine,ir}} \quad (7)$$

Input parameter values of this model are summarized in **TABLE 4** and are taken from benchmarking problem #3 in Pruess et al. (2002). The capillary pressure curve is fixed in this work to focus on the effects by the parameters of the relative permeability model.

TABLE 4
Input parameters for capillary pressure model.

Parameter	Value
P_0 , kPa	19.61
$S_{brine,ir}$, -	0
λ , -	0.457

2.3 Setup for parameter sensitivity study and stochastic analysis

The relative permeability model used in this work has four parameters, k_{r,CO_2}^0 , $S_{brine,ir}$, m and n . Values of these parameters were estimated by statistical fitting of the data from the 41 experiments in **TABLE 2** to a truncated normal/lognormal distribution. The influence on CO₂ injectivity of each of these parameters is investigated in this sensitivity study. P50 values of the cumulative probability distribution functions (CDF) are chosen for the base case values of each parameter, and total injected CO₂ is calculated from a numerical reservoir model simulation run. Then, one of the parameters is changed from the base case value, and the change of the total injected CO₂ is used to determine the sensitivity of injectivity to that parameter. The relative permeability parameters are varied independently because they were shown to be uncorrelated in **Figure 2**. This process is repeated for all of the parameters to determine normalized sensitivities.

Histograms of the normalized frequencies of each parameter's experimental data overlain with the fitted probability distribution function (PDF) and fitting parameters are contained in **Figure 4** and bottom of **TABLE 2**, respectively. $S_{brine,ir}$ and the k_{r,CO_2}^0 are physical parameters bound between 0 and 1. The CO₂ and brine curvature exponents (m and n) are bounded between 1 and 10, corresponding to a linear drainage relative permeability curve and the upper bound of the experimental data, respectively. A

truncated normal distribution is fitted to the experimental data of $S_{brine,ir}$ and m , and a truncated lognormal distribution is fitted to that of k_{r,CO_2}^0 and n due to asymmetric shapes of their histograms. **Figure 5** shows the comparisons of the CDFs of each parameter given by those experimental results and fitted distributions. **Figure 6** shows Q-Q plots of the experimental results and the fitted distributions. These plots show reasonable fitting of the distributions against experimental data. In addition, the two-sample Kolmogorov-Smirnov test was performed on the experimental data sets and the samples drawn from the fitted CDFs via a rejection sampling method (100,000 samples of each parameter are used to construct the theoretical CDF). **TABLE 5** shows p -values given by the two-sample Kolmogorov-Smirnov test, and they are larger than 0.05 (5% of significance level). The Kolmogorov-Smirnov tests were performed after the data sets were standardized.

Parameter values at a 10% cumulative probability, P10, as well as the P30, P50, P70, and P90 values are also shown, and are reported in **TABLE 6**. **Figure 7** shows the relative permeability curves generated from these values. In this sensitivity study, numerical simulations are run for 17 relative permeability curves: the base case using P50 values, and four non-P50 percentiles for each of the four parameters.

Figure 4—Normalized frequency and probability density function for the parameters estimated using published experimental data

Figure 5—Cumulative distribution function by experimental data (empirical CDF) and fitted distribution (theoretical CDF)

(a) Brine residual saturation

(b) End-point CO₂ relative permeability

(c) CO₂ exponent (m)

(d) Brine exponent (n)

Figure 6—Q-Q plots of the experimental data and the fitted distributions

TABLE 5

p -values estimated using the two-sample Kolmogorov-Smirnov Test on experimental data sets and fitted distributions.

Parameter	p-value
$S_{brine,ir}$	0.978
k_{r,CO_2}^0	0.492
CO ₂ exponent (m)	0.936
Brine exponent (n)	0.327

TABLE 6

Selected CDF percentiles estimated using the experimental data and truncated normal distributions as PDFs.

Parameter	CDF Percentiles				
	P10	P30	P50	P70	P90
$S_{brine,ir}$	0.3266	0.4301	0.5017	0.5733	0.6770
k_{r,CO_2}^0	0.0684	0.1252	0.1897	0.2858	0.5016
CO ₂ exponent (m)	1.929	2.977	3.799	4.650	5.908
Brine exponent (n)	1.263	1.642	2.017	2.497	3.421

Figure 7—Relative permeability curves for the parameter sensitivity study

The relative permeability parameter sensitivity study was performed to clarify the relative effect of each parameter on CO₂ injectivity. While the parameter values used in the sensitivity study are estimated from experimental data, values of individual parameters and their combinations still have uncertainty. These parameters depend on a variety of reservoir conditions, and, even for the same formation, e.g. the Basal Cambrian shown in **Figure 1 (c)**, they vary significantly between samples. Monte Carlo simulation is performed to account for the uncertainty in each individual parameter as well as combinations of parameters. Latin Hypercube Sampling (LHS) is used to reduce the number of realizations in an efficient but rigorous manner (McKay et al. 1979). LHS divides the distribution into many intervals, each with the same probability, with the number of intervals corresponding to the number of simulations. Samples are then randomly selected for each reservoir model parameter with each interval sampled only once. The continuous CDFs in **Figure 5** were used in this work. The CDFs were computed from sample means and variances, creating as an artifact very small and very large values not measured in experiments. We consider 1000 realizations of the relative permeability curves with LHS. To assess the reliability of the estimated uncertainty (variance) of the total injected CO₂ from the 1000 realizations, the stabilities of the estimated mean and variance are checked against cases of 100 and 500 realizations following the work of Ballio and Guadagnini (2004) on convergence assessment for numerical Monte Carlo simulations. When these values are similar between different realization sizes, the number of realizations can be considered to be sufficient for reliable estimation of the uncertainty.

3 Results

3.1 Parameter sensitivity study

Seventeen simulations were conducted with the relative permeability curves shown in **Figure 7** under constant injection pressure for 4 years to clarify the influence of each individual parameter on CO₂ injectivity. **Figure 8** shows the simulation results of injected CO₂ with time. The amount of injected CO₂ shows a linear relationship with time; the slope, however, depends on the reservoir rock and fluid conditions. It is observed in **Figure 8** that total injected CO₂ is highly sensitive to k_{r,CO_2}^0 compared to the other parameters. Higher k_{r,CO_2}^0 values produce higher injection rates. It is also noted that $S_{brine,ir}$ and k_{r,CO_2}^0 show positive correlations to the total injected CO₂, while m and n have negative correlations; i.e. increases in $S_{brine,ir}$ and k_{r,CO_2}^0 increase injectivity, while increases in m and n reduce injectivity.

Figure 9 shows the saturation distribution of the supercritical CO₂, i.e. the spread of the CO₂ plume, after 4 years of injection for the minimum, base, and maximum cases (P10, P50, and P90 values) for each parameter. For the mutually-saturated CO₂-brine displacement simulated here (which is equivalent to immiscible displacement), the shape of the CO₂ plume depends on the shock front of the supercritical CO₂.

Berg and Ott (2012) investigated the stability of CO₂–brine immiscible displacement considering the shock-front mobility ratio and two dimensionless numbers: the capillary number and the gravity number. The shock-front mobility ratio is the product of the viscosity ratio, $\mu_{\text{CO}_2}/\mu_{\text{brine}}$, and the relative permeability ratio, $k_{r,\text{CO}_2}(S_{\text{shock}})/k_{r,\text{CO}_2}(S_{\text{brine}} = 1)$. The macroscopic capillary number defined as the ratio of the macroscopic viscous force (pressure drop) to macroscopic capillary pressure is used in the description of the stability of the shock front. The gravity number is defined as the ratio of the viscous force to the gravity force, e.g. Hilfer and Øren (1996). Berg and Ott (2012) and Ott and Berg (2013) pointed out that in a gravity-dominated system where the gravity number is much higher than 1, the simple 1D Buckley-Leverett shock-front argument is no longer valid, and the gravity effect should be included in the calculation of the capillary number and shock-front mobility ratio. Kopp et al. (2009b) discussed the effect of these parameters on CO₂ storage capacity and that a low ratio of gravitational to viscous forces or a high ratio of capillary to viscous forces can lead to higher CO₂ storage capacities.

Figure 9 shows similar changes in the CO₂ plume shape from changes in $S_{\text{brine},ir}$ and k_{r,CO_2}^0 ; the CO₂ plume spreads more as the values of $S_{\text{brine},ir}$ and k_{r,CO_2}^0 increase. In this work, the capillary pressure curve is fixed and does not change with the change of parameters used in the sensitivity study. Similarly, the gravity number is nearly unchanged between sensitivity study cases because fluid density differences are indirectly and negligibly affected by changes to relative permeability parameters. The two dimensionless numbers are therefore principally affected by the convective force, which is described by the characteristic velocity defined as the saturation front propagation velocity in Kopp et al. (2009b). The characteristic velocity is approximated by the Buckley-Leverett shock-front velocity expressed as:

$$v_{S_{\text{CO}_2}} = \frac{q_t}{A\phi} \frac{df_{\text{CO}_2}}{dS_{\text{CO}_2}} \bigg|_{S_{\text{CO}_2}} \dots\dots\dots (8)$$

where q_t is the total volumetric flow rate, A is the cross-sectional area in the flow direction, ϕ is porosity, f_{CO_2} is the fraction of volumetric flow that is the CO₂ phase, and S_{CO_2} is the saturation of the CO₂ phase (Dake 1995). As shown in **Figure 8 (b)**, increases in k_{r,CO_2}^0 increase the flow rate, resulting in the advancement of the CO₂ plume shown in **Figure 9 (b)**. By comparison, changes in injection rate due to changes in $S_{\text{brine},ir}$ are relatively small (**Figure 8**). **Figure 8 and Figure 9** show negative correlations of CO₂ injectivity and plume spread with the curvature exponents, i.e. increased multiphase interference effects decrease both injectivity and plume spread.

In **Figure 10**, we show absolute and normalized changes to the total injected CO₂ with changes in the parameter values from the base case including both positive/negative correlations and differences in magnitudes of changes between parameters. While normalized changes of injected CO₂ are within a $\pm 10\%$ range for the entire range of $S_{\text{brine},ir}$, m , and n values, changes due to variations in k_{r,CO_2}^0 are $+80/-$

60%. The absolute and normalized radial CO₂ extents (chosen as $S_{\text{CO}_2} \geq 0.01$) after four years injection are shown in **Figure 11**. Again, variations in k_{r,CO_2}^0 cause comparatively large changes, +60%/-45%, while the curvature exponents cause similarly small changes, with all but the P10 value of the CO₂ curvature exponent causing changes that are within $\pm 10\%$ of the base case. However, $S_{\text{brine},ir}$ causes +35/-15% changes in the plume extent due to volumetric rearrangement of similar amounts of injected CO₂; i.e. a voxel at $S_{\text{brine},ir}$ or any other normalized saturation will have more (or less) CO₂, requiring the plume to contract (or expand) proportionally.

The parameter sensitivity study quantitatively shows that k_{r,CO_2}^0 has the dominant role in determining the total mass of injected CO₂ compared to the other parameters. The magnitude of the total injected CO₂ is impacted most strongly by k_{r,CO_2}^0 (50-60%), while the other relative permeability model parameters cause smaller changes, at most $\pm 10\%$. It is noted that the parameter range of k_{r,CO_2}^0 is wider than that of the other parameters in **Figure 10**; the experimental value of k_{r,CO_2}^0 has higher uncertainty compared to the others (compare the ratios of P10 and P90 values in **Table 6**). In addition, changes to $S_{\text{brine},ir}$ and k_{r,CO_2}^0 also affect the curvature of the relative permeability curve (**Figure 7**). The sensitivity study highlights the need for more rigorous analysis to generalize our observations related to uncertainty in the experimental measurements.

(a) Brine residual saturation

(b) End-point CO₂ relative permeability

(c) CO₂ exponent (m)

(d) Brine exponent (n)

Figure 8—Total injected CO₂ history resulting from this sensitivity study

Figure 9—CO₂ plume after 4 years injection with different parameter values

Figure 10—Changes in the total injected CO₂ after four years of injection from changes in parameter values from the base case

Figure 11—Changes in the radial extent of CO₂ ($S_{CO_2} \geq 0.01$) after four years injection due to normalized changes in parameter values from the base case

3.2 Monte Carlo Simulation

In this section we present a comprehensive investigation of the relative permeability parameters to more fully explore the uncertainty in the experimental measurements. The continuous CDFs in **Figure 5****Error! Reference source not found.**, which assume a truncated normal/lognormal distribution on each parameter in the relative permeability model, are used for Latin Hypercube Sampling. LHS reduces the number of realizations to achieve a reasonable but accurate random distribution as applied to retention curve uncertainty analysis in the work by Stauffer and Lu (2012). In the current study, 1000 realizations of the relative permeability model are generated. These realizations are plotted in **Figure 12**. As noted by Mathias et al. (2013) there is no clear relation between the relative permeability data and porosity, permeability, IFT or lithology in the measured data. Thus, our approach of spanning the parameter space with the observations should yield meaningful behavior expected from a wide range of reservoir rocks.

Figure 12—1000 Realizations of brine and CO₂ relative permeability curves of brine drainage by CO₂

We conducted 1000 numerical reservoir simulations for the previously described 2D cylindrical reservoir case (**Figure 3** and **Table 3**) using FEHM. **Figure 13****Error! Reference source not found.** shows total injected CO₂ mass as a function of time for 1000 realizations (gray lines). This figure also shows six curves representing the median (blue), mean (red) and confidence intervals. The green and orange solid curves show the mean $\pm 1\sigma$ (68% probability), and the green and orange dotted curves show the mean $\pm 2\sigma$ (95% probability). The mean value of total injected CO₂ amount at four years injection is 0.730 Mt, and the standard deviation is 0.370 Mt, which is 50.7% of the mean value.

Figure 13—Total injected CO₂ vs. time (1000 realizations)

The reliabilities of the estimated mean and standard deviation were checked by comparing these values against the cases of 100 and 500 realizations. **TABLE 7** summarizes the estimated mean, standard deviation, and coefficient of variation of the total injected CO₂ mass after 4 years of injection for different numbers of Monte Carlo realizations. While the mean value has a negligible change between the cases, the standard deviation and coefficient of variation changed between 100 and 500 realizations and the change gets smaller between 500 and 1000 realizations. Therefore, 1000 realizations are sufficient to estimate the sample mean and standard deviation.

TABLE 7

Changes of statistical properties of the Monte Carlo simulation results with different numbers of realizations (100, 500 and 1000).

Number of Realizations	Mean (Mt)	Standard Deviation (Mt)	Coefficient of Variation (-)
100	0.726	0.360	0.496
500	0.730	0.375	0.514
1000	0.730	0.370	0.507

Figure 14 (a) shows the histogram of the total injected CO₂ mass. The cumulative normalized frequency of the total injected CO₂ mass is calculated and compared in **Figure 14 (b)** with a normal distribution using the sampled mean and standard deviation values. **Figure 14 (b)** also shows a box plot using the statistical values associated with the minimum and the maximum value of the simulation results. The box plot also shows the confidence interval of $\pm 1\sigma$ with 68% probability after four years of injection, corresponding to the green and orange solid curves shown in the **Figure 13**. We assessed for the normality of the cumulative normalized frequency following Ballio and Guadagnin (2004). The Shapiro-

Wilk test for normality gives a p-value less than 0.05. Thus, statistically, the normalized frequency curve is not a normal distribution; however, the deviation from the normal distribution is not large near the mean (P50), as shown in **Figure 14 (b)**. Furthermore, **Figure 15** shows a normal Q-Q plot of the total injected CO₂ after 4 years of injection given by 1000 realizations. Deviation from the normal distribution is mainly apparent outside of the range of $\pm 1\sigma$, and the deviation becomes larger around the lower and upper quantiles of total injected CO₂; it is therefore reasonable to estimate the probability of the uncertainty in the standard deviation within the range of $\pm 1\sigma$ using a normal distribution for the total injected CO₂ amount. Cautions should be taken when quantifying the probability of the uncertainty using the standard deviation outside range of the $\pm 1\sigma$ since the distribution deviates somewhat from a normal distribution.

Figure 14—Statistical distribution of total injected CO₂ (1000 realizations)

Figure 15—Normal Q-Q plot of total injected CO₂ given by 1000 realizations

We also quantified the correlation of the total injected CO₂ amount to individual parameters in the relative permeability model. **Figure 16** shows variations in the total injected CO₂ after four years of injection associated with changes to the parameters in the relative permeability model for the 1000 realizations. It is observed that k_{r,CO_2}^0 shows strong correlation to the total injected CO₂, while $S_{brine,ir}$, m and n do not show any correlation. **Figure 17** shows rank variations in the total injected CO₂ after four years injection to rank changes of the parameters in the relative permeability model to investigate the Spearman rank correlation coefficient (Iman et al. 1981). It is more clearly observed that the rank of k_{r,CO_2}^0 shows strong correlation to the rank of the total injected CO₂ (rank correlation coefficient is 0.9881), while the ranks of $S_{brine,ir}$, m and n show no correlation.

(a) Brine residual saturation

(b) End-point CO₂ relative permeability

(c) CO₂ exponent (m)

(d) Brine exponent (n)

Figure 16—Total injected CO₂ to the parameters of the relative permeability model (1000 Realizations)

(a) Brine residual saturation

(b) End-point CO₂ relative permeability

(c) CO₂ exponent (m)

(d) Brine exponent (n)

Figure 17—Rank of total injected CO₂ to rank of the parameters of the relative permeability model (1000 Realizations)

In **Figure 18 (a)** and **(b)**, k_{r,CO_2}^0 is plotted versus the total injected CO₂ mass in both Cartesian and log-log scales to highlight the power law relationship; the power law regression lines, equations, and R^2 values are also shown on the figure. The R^2 value is 0.9685 (correlation coefficient is 0.9841): nearly all variation in CO₂ injectivity can be assigned to variations in the value of k_{r,CO_2}^0 raised to the 0.7224 power. Although the exponent of this function is specific to the geometry and physical state of this study, the underlying physical processes leading to this result should hold for other similar systems. The decrease in slope seen in **Figure 18 (a)** implies that injectivity is more sensitive to end-point relative permeability changes at the lower end of the measured range. In **Figure 18 (a)** and **(b)**, two vertical dotted lines are also drawn to show the experimentally measured range of k_{r,CO_2}^0 values (**TABLE 2**). The gray shaded

zones are regions with the k_{r,CO_2}^0 outside of the measurements; they are generated from the continuous CDF used in this study. The deviations from the regression curve are mainly seen in these regions.

Figure 18—Total injected CO₂ mass versus end-point CO₂ relative permeability with a regression line showing the power law relationship

The model regression fit to the Monte Carlo simulations is explained by only one parameter, k_{r,CO_2}^0 , but provides excellent estimation. In **Figure 19 (a)** and **(b)**, total injected CO₂ is plotted against k_{r,CO_2}^0 including the regression line and several error-envelopes ($\pm 10\%$, $\pm 20\%$ and $\pm 30\%$). As shown in **Figure 19 (a)**, the simulation results are largely within the $\pm 20\%$ range. **Figure 19 (b)** shows the same plot with a log-log scale (excluding the data with $k_{r,CO_2}^0 < 0.01$). As discussed in **Figure 18**, the results given by **Figure 18** outside the range of experimental measurement show significant deviation from the regression line. However, these values of k_{r,CO_2}^0 are created by use of the LHS with continuous CDFs. In terms of practical site-selection of CO₂ sequestration, we can ignore the deviation in these regions. Within experimentally measured range, results are clustered along the regression line and are distributed within the $\pm 30\%$ range, with only five exceptions out of 1000 realizations; i.e., 99.5% of model runs were within $\pm 30\%$ of a regression line relying solely on one variable: end-point CO₂ relative permeability.

Figure 19—Total injected CO₂ mass versus end-point CO₂ relative permeability including error envelopes from the regression line

These results support the conclusion of the sensitivity study that k_{r,CO_2}^0 has the dominant role in the determination of the total injected CO₂ mass when compared to other parameters in the relative permeability model. This conclusion highlights the importance of the work done by Levine (2011) to measure end-point CO₂ relative permeability and quantifies similar assertions made therein regarding the sensitivity of injectivity and storage capacity to end-point CO₂ relative permeability. Quantification of reservoir model sensitivity plays an important role in informing where limited experimental resources can achieve the most cost-effective result. Because end-point relative permeability is a function of wetting state, accurate determination of CO₂-brine wetting state through contact angle and core flooding experiments is necessary to help achieve the DOE's stated goal to "support industry's ability to predict CO₂ storage capacity in geologic formations to within $\pm 30\%$ ".

4 Summary and Conclusion

Numerical reservoir models rely on functions that are parameterized to avoid numerical instabilities and to achieve computational efficiency. Relative permeability between brine and carbon dioxide (CO₂) is a crucial parameter for determining CO₂ injectivity in CO₂ reservoir models. In this work, the FEHM reservoir simulator was used to determine the sensitivity of CO₂ injectivity to relative permeability model parameters.

A numerical sensitivity study uses a truncated normal/lognormal distribution fit to published experimental results to set a base case and relative permeability model parameter variation range. The

sensitivity study probes the effect of variations in the values of individual relative permeability model parameters at 10%, 30%, 70% and 90% cumulative probability values in comparison to a base case using 50% values for all the parameters. Variations in experimentally measured end-point CO₂ relative permeability from the base case cause +80/-60% changes in the total mass of CO₂ injected over 4 years and +60/-45% changes in the radial extent of the injected CO₂ (with a CO₂ saturation cutoff of 0.01). Variations in residual brine saturation cause +35/-15% changes to the CO₂ radial extent, but otherwise changes due to variations in residual brine saturation and either CO₂ or brine curvature exponents are within 10% of the base case, less than variations due to geological uncertainty, and in agreement with results obtained in previous semi-analytical studies.

A more robust Monte Carlo simulation method was used to determine the statistical sensitivity of CO₂ injectivity to changes in individual parameters while varying all model parameters. Latin Hypercube Sampling was used to efficiently but reliably perform Monte Carlo analysis using 1000 FEHM simulations. The results show that the total injected CO₂ mass varies by $\pm 50.7\%$ within 1σ and $\pm 101.4\%$ within 2σ for a range of shapes of relative permeability curves. The Monte Carlo simulation results show that total CO₂ injected is statistically correlated to only the end-point CO₂ relative permeability, with no correlation to residual brine saturation or the CO₂ or brine curvature exponents (m and n). Total injected CO₂ and end-point CO₂ relative permeability have a power law relationship with an exponent of 0.7224 and the relationship had an R^2 of 0.9685, i.e. nearly complete predictive power. Deviation from the regression line is within $\pm 30\%$ for 99.5% of the model runs.

Both the individual parameter sensitivity study and the Monte Carlo simulations show the importance of end-point CO₂ relative permeability measurements for accurate reservoir modeling. However, the results are only as good as the experimental data used to create the parameter distribution functions and the majority of reported measurements of end-point CO₂ relative permeability are known to suffer from significant experimental artifacts (Levine 2014). The end-point relative permeability and fluid-fluid-rock wetting state are directly related by well-studied petrophysical processes. Unfortunately, a recent review (Iglauer et al 2015) highlighted the significant uncertainty in CO₂-brine contact angle experimental measurements of the wetting state in GCS systems, with a nearly-complete lack of reproducibility between laboratories. In the absence of in-situ injection tests that can be used to estimate field properties, numerical modeling of geological carbon sequestration reservoirs relies on laboratory scale experimental measurements. High pressure CO₂-brine contact angle and core flooding experiments are difficult and expensive, but comprehensive and reliable datasets are needed. Models can only ever be as good as the input assumptions as determined by laboratory experimental measurement.

Our work focuses on uncertainty resulting from relative permeability parameters with experimentally determined parameter ranges. The parameter uncertainty is modeled using statistics of relative

permeability data obtained from several disparate formations. This might lead to an over estimation of the variability CO₂ storage capacity, but this is a practical approach to estimate range of uncertainty in the absence of field data. Once field data are available at a given site, history matching and model calibration can reduce model uncertainty and increase the predictive power of site specific models. In such efforts, core-scale relative permeability models are up-scaled to field-scale relative permeability models (Lomeland et al. 2012), but non-unique relationships between intrinsic and relative permeability models may result from such model calibration (Mishra et al. 2014). In addition, the 2D homogeneous model used in this work represents average intrinsic permeability of the reservoir, but heterogeneity in intrinsic permeability may attenuate the effects of relative permeability uncertainty reducing the variance of the CO₂ storage capacity.

Acknowledgement

This work is part of the Big Sky CO₂-EOR/Storage Project that is supported by the U.S. Department of Energy and managed by the National Energy Technology Laboratory under contract number DE-FE0000397. This research was supported in part by an appointment to the National Energy Technology Laboratory Research Participation Program, sponsored by the U.S. Department of Energy and administered by the Oak Ridge Institute for Science and Education.

References

- Anderson, W. G. 1987. Wettability Literature Survey Part 5: The Effects of Wettability on Relative Permeability. *Journal of Petroleum Technology* **39** (11): 1453-1468. doi: 10.2118/16323-PA.
- Bachu, S. 2013. Drainage and Imbibition CO₂/Brine Relative Permeability Curves at in Situ Conditions for Sandstone Formations in Western Canada. *Energy Procedia* **37** (0): 4428-4436. doi: 10.1016/j.egypro.2013.07.001.
- Bachu, S., Bennion, D. B. 2008. Effects of in-situ conditions on relative permeability characteristics of CO₂-brine systems. *Environmental Geology* **54** (8): 1707-1722. doi: 10.1007/s00254-007-0946-9.
- Bachu, S., Bennion, D. B. 2009. Interfacial Tension between CO₂, Freshwater, and Brine in the Range of Pressure from (2 to 27) MPa, Temperature from (20 to 125) °C, and Water Salinity from (0 to 334 000) mg·L⁻¹. *Journal of Chemical & Engineering Data* **54** (3): 765-775. doi: 10.1021/jc800529x.
- Ballio, F., Guadagnini, A. 2004. Convergence Assessment of Numerical Monte Carlo Simulations in Groundwater Hydrogy. *Water Resources Research* **40** (4): 729-774. doi: 10.1029/2003WR002876.
- Bennion, D. B., Bachu, S. 2005. Relative Permeability Characteristics for Supercritical CO₂ Displacing Water in a Variety of Potential Sequestration Zones. Paper SPE-95547-MS presented at SPE Annual Technical Conference and Exhibition, 9-12 October, Dallas, Texas. doi: 10.2118/95547-MS.
- Bennion, D. B., Bachu, S. 2006a. The Impact of Interfacial Tension and Pore Size Distribution/Capillary Pressure Character on CO₂ Relative Permeability at Reservoir Conditions in CO₂-Brine Systems. Paper SPE-99325-MS presented at SPE/DOE Symposium on Improved Oil Recovery, 22-26 April, Tulsa, Oklahoma, USA. doi: 10.2118/99325-MS.

- Bennion, D. B., Bachu, S. 2006b. Dependence on Temperature, Pressure, and Salinity of the IFT and Relative Permeability Displacement Characteristics of CO₂ Injected in Deep Saline Aquifers. Paper SPE-102138-MS presented at SPE Annual Technical Conference and Exhibition, 24-27 September, San Antonio, Texas, USA. doi: 10.2118/102138-MS.
- Bennion, D. B., Bachu, S. 2008. Drainage and Imbibition Relative Permeability Relationships for Supercritical CO₂/Brine and H₂S/Brine Systems in Intergranular Sandstone, Carbonate, Shale, and Anhydrite Rocks. *SPE Reservoir Evaluation & Engineering* **11** (03): 487-496. doi: 10.2118/99326-PA.
- Bennion, D. B., Bachu, S. 2010. Drainage and Imbibition CO₂/Brine Relative Permeability Curves at Reservoir Conditions for Carbonate Formations. Paper SPE-134028-MS presented at SPE Annual Technical Conference and Exhibition, 19-22 September, Florence, Italy. doi: 10.2118/134028-MS.
- Benson, S., Pini, R., Reynolds, C., Krevor, S. 2013. Relative Permeability Analysis to Describe Multiphase Flow in CO₂ Storage Reservoirs. Global CCS Institute Targeted Report (2013/08/07).
- Berg, S., Ott, H. 2012. Stability of CO₂-brine immiscible displacement. *International Journal of Greenhouse Gas Control* **11** (0): 188-203. doi: 10.1016/j.ijggc.2012.07.001.
- Burton, M., Kumar, N., Bryant, S. L. 2009. CO₂ injectivity into brine aquifers: Why relative permeability matters as much as absolute permeability. *Energy Procedia* **1** (1): 3091-3098. doi: 10.1016/j.egypro.2009.02.089.
- Chalabaud, C. A., Lombard, J.-M. N., Martin, F., Robin, M., Bertin, H. J., Egermann, P. 2007. Two Phase Flow Properties of Brine-CO₂ Systems in a Carbonate Core: Influence of Wettability on P_c and k_r. Paper SPE-111420-MS presented at SPE/EAGE Reservoir Characterization and Simulation Conference, 28-31 October, Abu Dhabi, UAE. doi: 10.2118/111420-MS.
- Dai, Z., P. H. Stauffer, J.W. Carey, R.S. Middleton, Z. Lu, J.F. Jacobs, K. Hnottavange-Telleen, L.H. Spangler. 2014. Pre-site characterization risk assessment for commercial-scale carbon sequestration, *Environ. Sci. Technol* **48** (7): 3908-3915. doi: 10.1021/es405468p.
- Dake, L. P. 1995. *Fundamentals of Reservoir Engineering*, Elsevier Science B.V. (Reprint).
- Deng, H., Stauffer, P. H., Dai, Z., Jiao, Z., Surdam, R. C. 2012. Simulation of industrial-scale CO₂ storage: Multi-scale heterogeneity and its impacts on storage capacity, injectivity and leakage. *International Journal of Greenhouse Gas Control* **10** (0): 397-418. doi: 10.1016/j.ijggc.2012.07.003.
- Dria, D. E., Pope, G. A., Sepehrnoori, K. 1993. Three-Phase Gas/Oil/Brine Relative Permeabilities Measured Under CO₂ Flooding Conditions. *SPE Reservoir Engineering* **8** (2): 143-150. doi: 10.2118/20184-PA.
- Egermann, P., Chalabaud, C. A., Duqueroix, J., Le Gallo, Y. 2006. An Integrated Approach to Parameterize Reservoir Models for CO₂ Injection in Aquifers. Paper SPE-102308-MS presented at SPE Annual Technical Conference and Exhibition, 24-27 September, San Antonio, Texas, USA. doi: 10.2118/102308-MS.
- González-Nicolás, A., Baù, D., Cody, B.M., and Alzraiee, A. 2015. Stochastic and global sensitivity analyses of uncertain parameters affecting the safety of geological carbon storage in saline aquifers of the Michigan Basin. *International Journal of Greenhouse Gas Control* **37**: 99-114. doi: 10.1016/j.ijggc.2015.03.008.
- Goodman, A., Hakala, A., Bromhal, G., Deel, D., Rodosta, T., Frailey, S., Small, M., Allen, D., Romanov, V., Fazio, J., Huerta, N., McIntyre, D., Kutchko, B. and Guthrie, G. 2011. U.S. DOE methodology for the development of geologic storage potential for carbon dioxide at the national and regional scale. *International Journal of Greenhouse Gas Control* **5** (4): 952-965. doi: 10.1016/j.ijggc.2011.03.010.
- Heath, J. E., Kobos, P. H., Roach, J. D., Dewers, T., McKenna, S. A. 2012. Geologic Heterogeneity and Economic Uncertainty of Subsurface Carbon Dioxide Storage. *SPE Economics & Management* **4** (01): 32-41. doi: 10.2118/158241-PA.
- Hilfer, R., Øren, P. E. 1996. Dimensional analysis of pore scale and field scale immiscible displacement. *Transport in Porous Media* **22** (1): 53-72. doi: 10.1007/BF00974311.

- Iglauer, S., Pentland, C. H., and Busch, A. 2015. CO₂ wettability of seal and reservoir rocks and the implications for carbon geo-sequestration. *Water Resources Research* **51** (1): 729-774. doi: 10.1002/2014WR015553.
- Iman, R. L., Helton, J. C., and Campbell, J. E. 1981. An Approach to Sensitivity Analysis of Computer Models, Part 1—Introduction, Input Variable Selection and Preliminary Variable Assessment. *Journal of Quality Technology* **13** (3): 174-183.
- Juanes, R., Spiteri, E. J., Orr, F. M., Blunt, M. J. 2006. Impact of relative permeability hysteresis on geological CO₂ storage. *Water Resources Research* **42** (12): W12418. doi: 10.1029/2005WR004806.
- Kopp, A., Class, H., Helmig, R. 2009a. Investigations on CO₂ storage capacity in saline aquifers: Part 1. Dimensional analysis of flow processes and reservoir characteristics. *International Journal of Greenhouse Gas Control* **3** (3): 263-276. doi: 10.1016/j.ijggc.2008.10.002.
- Kopp, A., Class, H., Helmig, R. 2009b. Investigations on CO₂ storage capacity in saline aquifers—Part 2: Estimation of storage capacity coefficients. *International Journal of Greenhouse Gas Control* **3** (3): 277-287. doi: 10.1016/j.ijggc.2008.10.001.
- Krevor, S. C. M., Pini, R., Zuo, L., Benson, S. M. 2012. Relative permeability and trapping of CO₂ and water in sandstone rocks at reservoir conditions. *Water Resources Research* **48** (2): W02532. doi: 10.1029/2011WR010859.
- Lake, L. W. 2010. *Enhanced Oil Recovery*. Richardson, Texas, Society of Petroleum Engineers (Reprint).
- Lemmon, E.W., McLinden, M.O., Friend, D.G. 2005. "Thermophysical Properties of Fluid Systems" in *NIST Chemistry WebBook, NIST Standard Reference Database Number 69*, Eds. P.J. Linstrom and W.G. Mallard, National Institute of Standards and Technology, Gaithersburg MD, 20899, <http://webbook.nist.gov>
- Levine, J. 2011. Relative Permeability Experiments of Carbon Dioxide Displacing Brine & Their Implications for Carbon Sequestration. PhD Dissertation, Columbia University (2011).
- Levine, J. S., Goldberg, D. S., Lackner, K. S., Matter, J. M., Supp, M. G., Ramakrishnan, T. S. 2014. Relative Permeability Experiments of Carbon Dioxide Displacing Brine and Their Implications for Carbon Sequestration. *Environmental Science & Technology* **48** (1): 811-818. doi: 10.1021/es401549e.
- Levine, J.S., Kelkar, S., Stauffer, P.H. 2013. Hydraulically Induced Shear Failure During CO₂ Injection into Deep Ocean Sediments. In *47th US Rock Mechanics/Geomechanics Symposium*.
- Lomeland, F., Hasanov, B., Ebeltoft, E., and Berge, M. 2012. A Versatile Representation of Upscaled Relative Permeability for Field Applications. Presented at the SPE Europec/EAGE Annual Conference, 4-7 June, Copenhagen, Denmark. SPE-154487-MS. doi:10.2118/154487-MS.
- McKay, M.D., Beckman, R.J., and Conover, W.J. 1979. A Comparison of Three Methods for Selecting Values of Input Variables in the Analysis of Output from a Computer Code, *Technometrics* **21** (2): 239-245. doi:10.2307/1268522.
- Middleton, R.S., G. Keating, P.H. Stauffer, A. Jordan, H. Viswanathan, Q. Kang, B. Carey, M. Mulkey, J. Sullivan, S.P. Chu, and R. Esposito. 2012. The multiscale science of CO₂ capture and storage: From the pore scale to the regional scale. *Energy and Environmental Science*, **5**, 7328. doi:10.1039/C2EE03227A.
- Mishra, S., Oruganti, Y. D., Gupta, N., Ganesh, P. R., McNeil, C., Bhattacharya, I. and Spitznogle, G. 2014. Modeling CO₂ plume migration based on calibration of injection and post-injection pressure response at the AEP Mountaineer Project. *Greenhouse Gas Sci Technol* **4**: 331-356. doi:10.1002/ghg.1434.
- Müller, N. 2011. Supercritical CO₂-Brine Relative Permeability Experiments in Reservoir Rocks—Literature Review and Recommendations. *Transport in Porous Media* **87** (2): 367-383. doi: 10.1007/s11242-010-9689-2.
- Mathias, S. A., Gluyas, J. G., González Martínez de Miguel, G. J., Bryant, S. L., Wilson, D. 2013. On relative permeability data uncertainty and CO₂ injectivity estimation for brine aquifers.

- 681 *International Journal of Greenhouse Gas Control* **12** (0): 200-212. doi:
682 10.1016/j.ijggc.2012.09.017.
- 683 National Energy Technology Laboratory. 2012. 2012 United States Carbon Utilization and Storage Atlas
684 (Atlas IV). (DOE/NETL-2012/1589).
- 685 Ott, H., Berg, S. 2013. Stability of CO₂-Brine Primary Drainage. *Energy Procedia* **37** (0): 4568-4574.
686 doi: 10.1016/j.egypro.2013.06.364.
- 687 Perrin, J.-C., Benson, S. 2010. An Experimental Study on the Influence of Sub-Core Scale
688 Heterogeneities on CO₂ Distribution in Reservoir Rocks. *Transport in Porous Media* **82** (1): 93-
689 109. doi: 10.1007/s11242-009-9426-x.
- 690 Perrin, J.-C., Krause, M., Kuo, C.-W., Miljkovic, L., Charoba, E., Benson, S. M. 2009. Core-scale
691 experimental study of relative permeability properties of CO₂ and brine in reservoir rocks.
692 *Energy Procedia* **1** (1): 3515-3522. doi: 10.1016/j.egypro.2009.02.144.
- 693 Pini, R., Benson, S. M. 2013. Simultaneous determination of capillary pressure and relative permeability
694 curves from core-flooding experiments with various fluid pairs. *Water Resources Research* **49**
695 (6): 3516-3530. doi: 10.1002/wrcr.20274
- 696 Porter, M.L., D. Wildenschild, G. Grant, and J.I. Gerhard. 2010. Measurement and prediction of the
697 relationship between capillary pressure, saturation, and interfacial area in a NAPL-water-glass
698 bead system. *Water Resources Research*, 46(8). .
- 699 Pruess, K., Garcia, J., Kovscek, T., Oldenburg, C., Rutqvist, J., Steefel, C., Xu, T. 2002. Intercomparison
700 of numerical simulation codes for geologic disposal of CO₂. Tech. rep., Lawrence Berkley
701 National Laboratory, Earth Science Division. LBNL-51813.
- 702 Ramakrishnan, T. S., Capiello, A. 1991. A new technique to measure static and dynamic properties of a
703 partially saturated porous medium. *Chemical Engineering Science* **46** (4): 1157-1163. doi:
704 10.1016/0009-2509(91)85109-B.
- 705 Stauffer, P.H., Viswanathan, H.S., Pawar, R.J., Guthrie, G.D., A system model for geologic sequestration
706 of carbon dioxide, *Environ. Sci. & Technol.* **2009**, 43 (3), 565-570.
- 707 Stauffer, P.H., A. Lu. 2012. Quantifying transport uncertainty in unsaturated rock using Monte Carlo
708 sampling of retention curves. *Vadose Zone J* **11**(4). doi:10.2136/vzj2011.0171
- 709 Stauffer, P. H., Lewis, K. C., Stein, J. S., Travis, B. J., Lichtner, P., and Zyvoloski, G. 2014. Joule-
710 Thomson Effects on the Flow of Liquid Water. *Transport in Porous Media* **105** (3): 471-485. doi:
711 10.1007/s11242-014-0379-3.
- 712 van Genuchten, M. T. 1980. A Closed-form Equation for Predicting the Hydraulic Conductivity of
713 Unsaturated Soils1. *Soil Sci. Soc. Am. J.* **44** (5): 892-898. doi:
714 10.2136/sssaj1980.03615995004400050002x.
- 715 Willhite, G. Paul. 1986. *Waterflooding*. Richardson, Texas, USA. Society of Petroleum Engineers.
- 716 Zyvoloski, G.A., Robinson, B.A., Dash, Z.V. and Trease, L.L. Summary of the Models and Methods for
717 the (FEHM) Application -- A Finite-Element Heat- and Mass-Transfer Code. LA-13306-MS, Los
718 Alamos National Laboratory, Los Alamos, New Mexico, (2011).