



Research paper

Impact of the capillary pressure-saturation pore-size distribution parameter on geological carbon sequestration estimates



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ABSTRACT

Cost estimates for geologic carbon sequestration (GCS) are vital for policy and decision makers evaluating carbon capture and storage strategies. Numerical models are often used in feasibility studies for the different stages of carbon injection and redistribution. Knowledge of the capillary pressure-saturation function for a selected storage rock unit is essential in applications used for simulating multiphase fluid flow and transport. However, the parameters describing these functions (e.g. the van Genuchten m pore size distribution parameter) are often not measured or neglected compared to other physical properties such as porosity and intrinsic permeability. In addition, the use of average instead of point estimates of m for numerical simulations of flow and transport can result in significant errors, especially in the case of coarse-grained sediments and fractured rocks. Such erroneous predictions can pose great risks and challenges to decision-making. We present a comparison of numerical simulation results based on average and point estimates of the van Genuchten m parameter for different porous media. Forward numerical simulations using the STOMP code were employed to illustrate the magnitudes of the differences in carbon sequestration predictions resulting from the use of height-averaged instead of point parameters. The model predictions were converted into cost estimates and the results indicate that varying m values in GCS modeling can cause cost differences of up to hundreds of millions dollars.

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1. Introduction

Anthropogenic emissions of CO₂ are rapidly changing the gaseous composition of the atmosphere, and contributing to global climate change due to the demands for power production. Assuming the continued use of fossil fuels, geologic carbon sequestration (GCS) is considered to be the most promising storage option for the CO₂ produced, due to its high potential volume of sequestration, combined with manageable life cycle cost (Herzog, 2001; Ennis-King & Paterson, 2002; Herzog & Golomb, 2004; Pacala & Socolow, 2004; Bickle, 2009; Szulczeński, MacMinn, Herzog, & Juanes, 2012; Rogelj, McCollum, Reisinger, Meinshausen, & Riahi, 2013; Scott, Gillan, Markusson, Chalmers, & Haszeldine, 2013). The main geologic storage units are confined saline aquifers,

coal beds, depleted oil reservoirs, shales, and other reactive rocks that facilitate carbonate precipitation. Scientists and engineers around the world from countries including Australia, Canada, Japan, China, the United Kingdom, and many other member states of the European Union, are working on pilot projects and conducting research to develop economically acceptable full-scale facilities in the near future (Clean Energy Ministerial, 2012).

The Carbon Storage Program of the U.S. Department of Energy (DOE) aims to develop and advance technologies that will significantly improve the efficacy of GCS, reduce the cost of implementation, and lay the foundations for widespread commercial deployment between 2020 and 2030. Over the past few years, DOE alone has committed millions of dollars in funding to research in order to evaluate, improve, and engineer GCS for future full-scale operations (DOE, 2009; DOE, 2010; DOE, 2011; DOE, 2012). The first demonstration-scale test to inject 1 million metric tons (~32 kg/s) of carbon dioxide at a depth of 7000 feet in a saline formation over a three-year period began in Illinois in November 2011. The target saline formation, Mt. Simon Sandstone, is the

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thickest and most widespread saline reservoir in the Illinois Basin, which has an estimated CO₂ storage capacity of 11–151 billion metric tons (Illinois State Geological Survey, 2011).

Numerical simulations are important tools in evaluating the feasibility of GCS, and the design and operation of future storage systems (Birkholzer, Zhou, & Tsang, 2009; Gherardi, Xu, & Pruess, 2007; Han, Kim, Esser, Park, & McPherson, 2011; Jiang, 2011; Juanes, Spiteri, Orr, & Blunt, 2006; Nordbotten, Celia, Bachu, & Dahle, 2005; Obi & Blunt, 2006; Oldenburg & Doughty, 2011; Pruess & Garcia, 2002; Pruess et al., 2002; Wigand, Carey, Schutt, Spangenberg, & Erzinger, 2008). Among the four trapping mechanisms of CO₂ in the subsurface (structural, capillary, solubility, and mineral trappings), capillary or residual-phase trapping of CO₂-rich fluids in pores is particularly important for geologic sequestration in dipping aquifers that do not have structural closure (Hesse, Orr, & Tchelepi, 2008; MacMinn, Szulczevski, & Juanes, 2011). After CO₂ injection stops, plumes of CO₂ gas begin to migrate into the adjacent water/brine saturated rocks, where the hysteresis of relative permeability can be important (Juanes et al., 2006; Oostrom, White, Porse, Krevor, & Mathias, 2016). Predicting the behavior of these CO₂ plumes depends upon our knowledge of the petrophysical parameters for CO₂ gas displacing water or brine in geologic materials (Doughty, 2007). However, estimates of these input parameters are limited and can vary across a wide range depending on the rock type of the target reservoir. Researchers and policy makers need accurate parameter estimates to evaluate the total amount of carbon that can be stored in a particular rock formation and to predict the redistribution of CO₂ following injection (Cheng et al., 2013; Doughty, 2007; Schnaar & Digiulio, 2009). Along with total porosity and intrinsic permeability, parameters describing the functional relationship between liquid saturation, *S*, and capillary pressure, *P_c*, are essential for modeling gas-liquid displacements (i.e. the prediction of the relative permeability functions for the wetting and non-wetting phases) in porous media. Because of their dependence upon interfacial tension and contact angle, these parameters vary depending on the particular combination of displacing and displaced fluids under different temperatures and pressures, and can change drastically.

2. Materials and methods

Capillary pressure-saturation function parameters are essential for modeling the geological storage of carbon dioxide, where brine is the wetting phase, and the supercritical CO₂ is the non-wetting phase. Parameter estimates need to be obtained by fitting equations to experimentally-determined data (Doughty, 2007; Heath, Kobos, Roach, Dewers, & McKenna, 2012). The van Genuchten (VG) (van Genuchten, 1980) and Corey equations (Brooks & Corey, 1996; Corey, 1954; Mualem, 1976) are commonly used in fitting experimental data and extracting the saturation function parameters (White & Oostrom, 2003):

$$S^* = \left(1 + \alpha(P_c)^{1/(1-m)}\right)^m \quad (1)$$

$$K_{rw} = (S^*)^{0.5} \left\{ 1 - \left[1 - (S^*)^{(1/m)} \right]^m \right\}^2 \quad (2)$$

$$K_{rn} = [1 - (S^*)^2] \cdot [1 - (S^*)^2] \quad (3)$$

where *S^{*}* is the normalized water saturation, *m* is an empirical pore-size distribution parameter, *α* is the inverse of the entry pressure for the non-wetting fluid, *K_{rw}* is the relative permeability of the wetting phase, and *K_{rn}* is the relative permeability of the non-

wetting phase. Estimates of the parameters in Eq. (1) depend upon the height of the column used in the experiments. As a result, inverse modeling must be used to extract unique parameters for a physical point (Cropper, Perfect, van den Berg, & Mayes, 2011). The uncorrected parameters are non-unique and are referred to as height-averaged. Significant difference in the cumulative drainage of a sample column can be observed for variably-saturated flow simulations in response to step changes in applied pressure with point and average capillary pressure-saturation parameters (Fig. 1). Variations in the van Genuchten *m* pore-size distribution parameter on the capillary pressure-saturation and relative permeation functions can be very significant as shown in Fig. 2.

To the authors' knowledge, only a few experimental *S* versus *P_c* data sets are available for CO₂ gas or supercritical CO₂ displacing water or brine in porous media. Chalbaud, Lombard, Martin, and Robin (2007) used X-ray imaging in carbonate rock to quantify water saturation profiles during multi-rate unsteady flow experiments, and then employed an inverse history matching approach to estimate *P_c* at all locations where *S* was measured yielding multiple point estimates of the drainage capillary pressure-saturation curve. The *S* versus *P_c* curves for CO₂ differed from those obtained for N₂ displacing water depending on the wettability of the rocks. Plug and Bruining (2007) used unconsolidated quartz sand with a pressure cell combined with dynamic flow conditions to measure the average saturation at different fluid pressures and temperatures during CO₂ injection and imbibition. The resulting hysteretic average saturation versus capillary pressure curves were compared to those obtained using immiscible N₂ and exhibited a pronounced pressure dependency. Their data suggests that the indirect approach can be applied to predict drainage curves, but that rewetting curves do not scale according to the interfacial tension when temperature and pressure change. The Stanford University group has used X-ray CT scanning to measure relative permeability functions for CO₂ and water systems, but no capillary pressure-saturation parameters were reported (Krevor, Pini, Li, & Benson, 2011; Pini, Krevor, & Benson, 2012).

As the number of projects evaluating GCS has grown, journal publications on numerical modeling of GCS are appearing at a rate of approximately 7–9 per month in comparing to 5–6 per month before 2013. Searching the Web of Science with the keywords "carbon dioxide", "geological", "storage", and "modeling" returned 777 records (2011: 82; 2012: 73; 2013: 108; 2014: 93; 2015: 84;

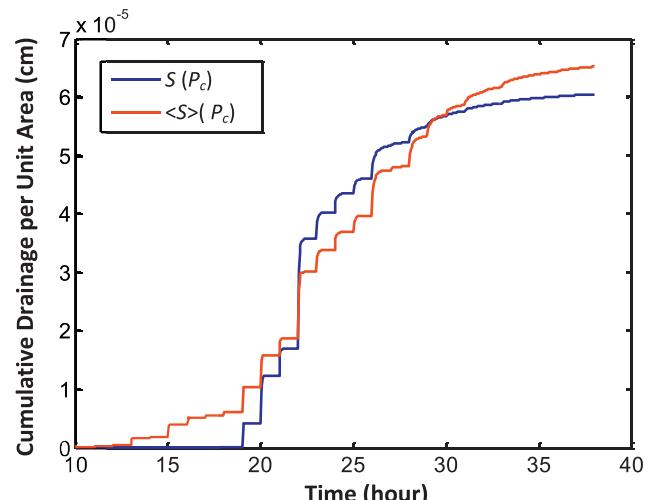


Fig. 1. Cumulative drainage of sample column in response to step changes in applied pressure simulated using both point, *S*(*P_c*), and average, *<S>*(*P_c*), capillary pressure-saturation parameters in the STOMP numerical model for variably-saturated flow.

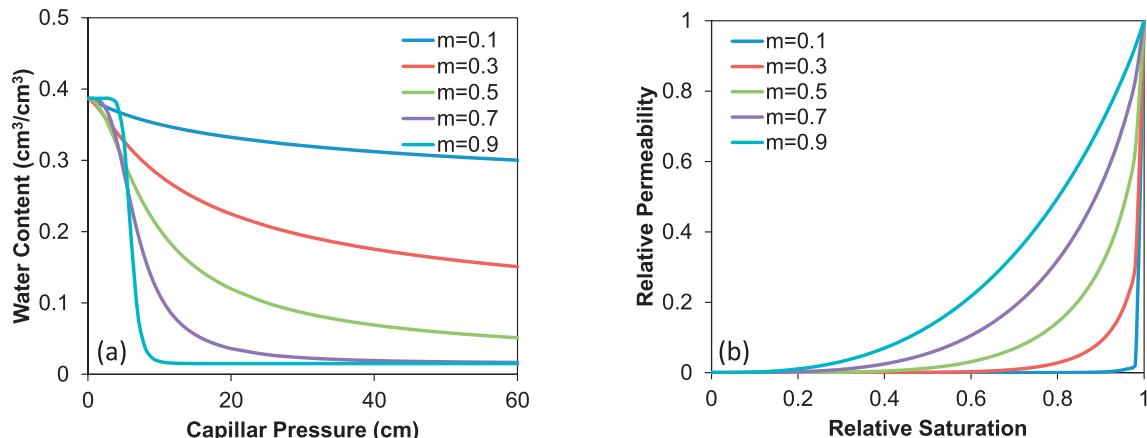


Fig. 2. Variations in (a) predicted capillary pressure-saturation [using Eq. (1)] and (b) wetting phase relative permeability functions [using Eq. (2)] with different m values based on the van Genuchten (1980) equation.

2016: 111; 2017: 49). More than two-thirds of these papers used a particular m value of 0.457 (as shown in Table 1) (Andre, Audigane, Azaroual, & Menjoz, 2007; Audigane, Gaus, Czernichowski-Lauriol, Pruess, & Xu, 2007; Birkholzer et al., 2009; Gherardi et al., 2007; Pruess & Garcia, 2002; Pruess, 1997; Pruess, Xu, Apps, & Garcia, 2003). We have been unable to trace how this m value (0.457) was first measured and what porous medium it was estimated for. Furthermore, Cropper et al. (2011) indicated that the VG m parameter showed a major curvilinear deviation when different methods (height averaged versus point) are used to estimate it. He indicated that the height averaging method systematically underestimates the m parameter relative to the point-based estimates leading to an over prediction of the breadth of the pore size distribution. Cheng et al. (2013) conducted sensitivity analyses on a suite of petrophysical parameters for GCS and found that the m parameter ranked second after intrinsic permeability in terms of its influence on the cost per ton of injected CO_2 . Use of biased or assumed m parameter values could lead to erroneous predictions in GCS and impose great risks and challenges to decision-making. Given the dearth of direct measurements on CO_2 -water/brine-rock systems, we conducted a sensitivity analysis, including cost estimates, to determine the impact of the VG m parameter in model simulations of subsurface carbon sequestration.

3. Results and discussion

3.1. The impact of the m parameter on numerical simulations

The main purpose of this technical note is to emphasize the importance and impacts of having accurate estimates of the VG m parameter for inputs to numerical models used to predict

multiphase flows associated with geologic carbon sequestration. Forward numerical simulations using STOMP (Subsurface Transport Over Multiple Phases) (White & Oostrom, 2003) were employed to illustrate the magnitudes of the errors in flow and transport predictions resulting from the use of height-averaged instead of point estimates of m for different materials. The model chosen was radial flow from a CO_2 injection well in a 100 m thick brine aquifer, which is an example problem used in a model inter-comparison study (Pruess et al., 2002).

Values of the VG m parameter for Berea sandstone (a sedimentary rock), Glass beads (45–70 μm diameter, GB), disturbed Hanford sand (HL), and undisturbed sediments from the upper coarse layer (UCL), medium fine layer (MFL), and lower coarse layer (LCL) at Hanford were obtained from Cropper et al. (2011). This small but diverse group of porous media permits a rigorous evaluation of the sensitivity of GCS simulations to m , including: consolidated (Berea) versus unconsolidated (GB, HL, UCL, MFL, and LCL), sieved and repacked (GB and HL) versus undisturbed (UCL, MFL, LCL), high porosity (HL) versus low porosity (Berea), coarse texture (LCL) versus fine texture (GB), and wide pore-size distribution (Berea) versus narrow pore-size distribution (GB). Cropper et al. (2011) also investigated the effects of the estimation method (point versus height averaged) on the values of m . Point and height-averaged VG m parameter values for the 6 different porous media were obtained from Cropper et al. (2011) using the relationship $m = 1 - 1/n$. In each case the average of all 3 replicates were used. The resulting VG m values used in STOMP ranged from 0.426 to 0.862. The widely-used VG m value of 0.457 (as discussed previously) was used as our baseline in the model. All of the other parameters in the model remained unchanged while the VG m values were varied (Pruess et al., 2002).

Table 1

List of highly cited journal papers in GCS modeling and the m value used (as of August 2017).

| Authors | Year | Title | m value | Citation |
|-------------------|------|---|---------------|----------|
| Pruess and Garcia | 2002 | Multiphase flow dynamics during CO_2 disposal into saline aquifers | 0.457 | 186 |
| Birkholzer et al. | 2009 | Large-scale impact of CO_2 storage in deep saline aquifers: A sensitivity study on pressure response in stratified systems | 0.46 | 238 |
| Pruess et al. | 2003 | Numerical modeling of aquifer disposal of CO_2 | 0.457 | 108 |
| Andre et al. | 2007 | Numerical modeling of fluid-rock chemical interactions at the supercritical CO_2 -liquid interface during CO_2 injection into a carbonate reservoir | 0.60 | 129 |
| Gherardi et al. | 2007 | Numerical modeling of self-limiting and self-enhancing caprock alteration induced by CO_2 storage in a depleted gas reservoir | 0.457 | 107 |
| Audigane et al. | 2007 | Two-dimensional reactive transport modeling of CO_2 injection in a saline aquifer at the Sleipner site | 0.65 −0.75 | 157 |
| Pruess | 1997 | On vaporizing water flow in hot sub-vertical rock fractures | 0.457 | 21 |

The STOMP simulations indicated that when the VG m value increases in brine aquifers, less carbon mass is stored in the aqueous phase, while more carbon mass occurs in the gas phase. The VG m parameter is related to the pore-size distribution; the smaller the m value, the wider the range of pore sizes present. Therefore, aquifers with smaller VG m values should provide greater solubility trapping. However, smaller m values can also cause pressure to build up within the reservoir due to the presence of smaller pores. As gas pressure builds up at the injection point, it can increase operation and maintenance costs on infrastructure.

To better demonstrate the practical effects of m on GCS, mean build-up pressures at the injection well resulting from the different m values were calculated as a function of the injection rate and compared to the conditions in the base model (injection rate of 12.5 kg/s, VG m parameter of 0.457, gas pressure at 2.0×10^7 Pa at the injection point). Corresponding injection rates for the different m values were then computed and compared at the same back pressure as the base model (Fig. 3). Variations in these corresponding injection rates reflect the ease with which CO₂ can be injected into the reservoir. Based on Fig. 3, the smaller the VG m parameter, the lower the corresponding rate of CO₂ injection that can be achieved while at the same baseline pressure. As shown, variations in the corresponding injection rates, caused solely by differences in the m parameter (0.426 v.s. 0.772), ranged over several kilograms per second (11.5 kg/s and 19.2 kg/s), which is equivalent to a total difference of 0.2 million metric tons (200 million kilograms) of CO₂ sequestered per year.

3.2. The impact of the m parameter on cost estimates

A major concern for GCS is the huge uncertainties in estimated costs (Herzog, 2001). Policy and decision making rely greatly on these estimated costs. Many studies have been conducted to investigate the economic analysis of GCS (Allinson, Nguyen, & Bradshaw, 2003; Bock et al., 2003; Azar, Lindgren, Larson, & Mollersten, 2006; Friedmann, Dooley, Held, & Edenhofer, 2006; USEPA, 2008; Eccles, Pratson, Newell, & Jackson, 2009; McCoy & Rubin, 2009; Vidas, Hugman, & Clapp, 2009; Eccles, Pratson, Newell, & Jackson, 2011). Eccles et al. (2009) applied an analytical model with three major components (i.e. maximum storage potential, maximum injection rates, and cost per ton stored) and

concluded that, because the properties of natural reservoirs vary substantially, storage costs vary on a case-by-case basis, sometimes by orders of magnitude. Eccles et al. (2011) further explored the cost of storage using their previous model for 15 key deep-saline sandstone aquifers in the United States. They showed that cost estimates for storage span three orders of magnitude and average more than \$100/ton of CO₂. Variations in the pore-size distribution of the target formation were not explicitly considered in any of the above engineering and economic analyses.

To better demonstrate the possible impacts of the VG m parameter of different media on costs in GCS, simple cost estimates have been performed. Heath et al. (2012) reported that the annualized costs of a 7000-foot deep (2134 m) well injection is about \$876,483 per annum in 2010 US dollars (\$917,803 per annum in 2012 US dollars based on an inflation factor of 1.023 per year). The minimum and maximum VG m parameters that we investigated (0.426 and 0.772, respectively) corresponded to injection rates of 11.5 and 19.2 kg/s, respectively. Based on the dollar amount reported by Heath et al. (2012), the costs of GCS operations in reservoirs with these two different m values were estimated to be between \$2.53/ton to \$1.52/ton, respectively in 2012 dollars. The impacts solely due to variations in the VG m parameter values in the numerical simulations resulted in approximately one million dollars difference per million metric tons of CO₂ injected. The Mt. Simon Sandstone, for example, has an estimated CO₂ storage capacity of 11–151 billion metric tons (11,000 to 151,000 billion kilograms). In this setting, the differences between values of the m parameter could lead to billions of dollars of difference over the course of the whole GCS operation. Other petrophysical properties, such as intrinsic permeability and porosity, can result in even greater variations in estimated costs (Heath et al., 2012).

The VG m parameter values selected reflect differences in both the type of porous medium and estimation method (point versus height averaged). Fig. 4 indicates that costs per ton of injected CO₂ were highest for materials with small m values, or broad pore-size distributions (e.g. Berea) and lowest for materials with large m values or narrow pore-size distributions (e.g. glass beads, GB). The estimation method can also have a significant impact, with m values from the height averaging method significantly overestimating costs compared to the more accurate point estimates of m (Fig. 4). Note that the largest value of m investigated (the 0.862

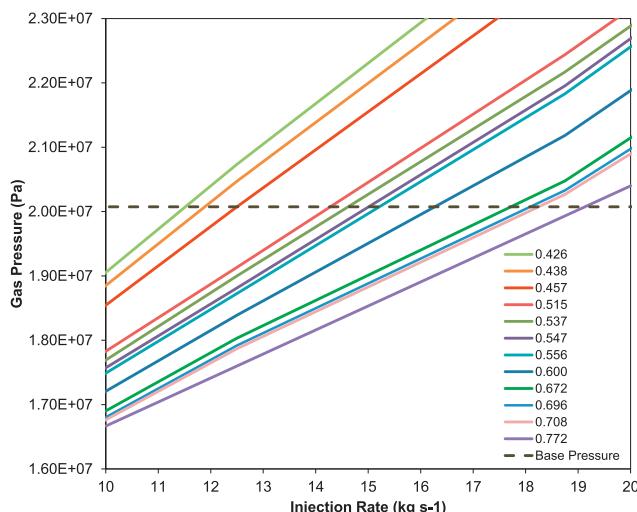


Fig. 3. Mean gas pressure in the brine reservoir versus injection rate for different m parameter values. The base pressure line-designates the gas pressure associated with the base model ($m = 0.457$, injection rate = 12.5 kg/s).

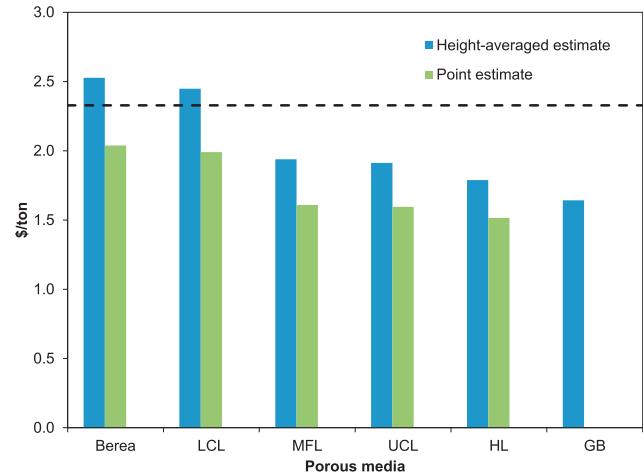


Fig. 4. Variation in injection costs for geologic carbon sequestration due to the m values of different porous media (LCL: lower coarse layer Hanford sediments; MFL: middle fine layer; UCL: undisturbed upper coarse layer; HL: disturbed upper coarse layer; GB: glass beads), and their estimation method (point versus height averaged). Dashed line: \$2.33/ton for base model (one ton equals 1000 kg).

point estimate for glass beads) gives a step function-like capillary pressure saturation curve with gas entry occurring over a very narrow range. This combination of features resulted in the model failing to converge. Therefore, results for this simulation were not included in Figs. 3 and 4. The environmental conditions in reservoirs such as temperature and pressure while the CO₂ is injected can also influence capillary pressure-saturation function parameters. Therefore, estimates of petrophysical parameters for a particular porous medium and fluid pair should be based on actual experimental data collected under relevant environmental conditions. Such input data is vital for accurate model simulations and can have a significant impact on cost estimates for GCS applications. It will help society to continue to use fossil fuels while sustain mining in energy sources and maintain the high quality of the environment at the same time.

4. Conclusions

Numerical modeling can be a powerful tool for evaluating different well designs and CO₂ injection strategies when accurate input parameters are available. Petrophysical properties of the porous medium are important and can be critical to GCS injection predictions. Large numbers of GCS modeling studies have assumed a single value for the van Genuchten capillary pressure-saturation parameter (m), which is related to the width of the pore-size distribution. Numerical simulations of CO₂ injection into a brine aquifer were shown to be quite sensitive to variations in measured values of this parameter for different porous media. The simulations also illustrated that the magnitudes of the errors in flow and transport predictions resulting from the use of height-averaged instead of point values of m on GCS were significant. Estimated costs due to the influence of the m parameter alone can be in range of millions of dollars over the lifetime of GCS operations. As petrophysical properties of the porous medium in the target reservoir are critical to GCS injections, direct measurements of these properties for the relevant fluid pair at appropriate environmental conditions are essential.

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