ENERGY AND THE SUBSURFACE: MODELING HYDRAULIC FRACTURING AND GEOLOGICAL CARBON STORAGE

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Hydraulic fracturing, shale gas, and geological carbon storage are key topics involving subsurface fluid flow that relate to the energy and climate challenge. In this dissertation, I advance the current understanding of these topics using numerical modeling of porous media fluid flow, the development new numerical models, and economic and policy analysis, all supported by a comprehensive collation of data.

In Chapter 2, I investigate the fate of hydraulic fracturing fluids injected into shale formations through numerical modeling of two-phase water and gas flow. My simulations match water and gas production data closely and show that most injected water is imbibed into the shale and retained there by capillary forces. Capillary pressure is a key factor governing the movement of water in shales, and data show that capillary pressure in shales is strongly hysteretic. Therefore, in Chapter 3, I develop a modified numerical capillary pressure hysteresis model that is robust and computationally efficient for simulating flows in shale formations compared with existing models that perform poorly under equivalent conditions.

In Chapter 4, I consider geological carbon storage in shale gas formations. I develop a numerical model of single-phase, two-component flow of methane and carbon dioxide in shales, including adsorption effects, in order to assess geological carbon storage capacity. Application of the model to three major shale gas regions shows that carbon dioxide can only be injected at low rates into individual wells, that individual well capacity is
relatively small, and, therefore, that large-scale carbon storage in shales is unlikely to be economically favorable.

Finally, in Chapter 5, I broaden the scope beyond modeling shale gas formations by conducting an integrated engineering, economic, and policy analysis of potential carbon capture, utilization, and storage deployment in the United States as a result of new tax credits enacted in February 2018. I focus on the pipeline infrastructure required to enable the system. I find that a large-scale system capturing carbon dioxide from ethanol biorefineries in the Midwest and delivering it for use in enhanced oil recovery in the Permian Basin, Texas, could be feasible with low-cost government financing of the pipeline network.
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Chapter 1

Introduction

1.1 Energy, Climate, Environment, and the Subsurface

Anthropogenic emissions of greenhouse gases to the atmosphere have led to approximately one degree Celsius of warming of the Earth’s climate since significant emissions began with the industrial revolution in the 1800s (USGCRP, 2017). The majority (65%) of anthropogenic greenhouse gas emissions are carbon dioxide resulting from the combustion of fossil fuels to fulfill societal energy needs (IPCC, 2014). If the past trend of emissions growth over time (shown in Figure 1.1) continues in the coming decades, we are likely to see much more warming and climate disruption that would present substantial risks to human civilization (IPCC, 2013; USGCRP, 2017). In response to these risks the Paris Climate Agreement was signed in 2015, in which the nations of the Earth set a goal to limit global warming to two degrees Celsius above the pre-industrial average. This goal requires global emissions to begin decreasing in the near future and net greenhouse gas emissions to the atmosphere to be reduced to zero before
the end of this century, as shown in Figure 1.1 (IPCC, 2014). The imperative to rapidly and drastically reduce emissions, combined with the continuing and growing need for energy to power our societies, constitutes the energy and climate problem.

![Figure 1.1 – Global anthropogenic CO₂ emissions (billion tonnes of CO₂ per year) including historical emissions (CDIAC, 2017), the emission pathway implied by the current Paris Agreement pledges (MIT, 2015), and a two degree Celsius emissions pathway (RCP2.6) (IIASA, 2011).](image)

The energy and climate problem is an enormous challenge facing humanity. The physical scale of the challenge and its timescales are daunting, and it encompasses many aspects of our societies: its causes and solutions lie at the intersection of science, engineering and technology, economics, sociology, and politics. This dissertation relates to two important topics in energy, climate, and the environment involving subsurface fluid flow: the advent of hydraulic fracturing and shale gas and geological carbon storage. I present subsurface fluid flow simulation studies investigating key questions related to both of these topics. I also present an integrated engineering, economics, and policy analysis examining a pathway for near-term large-scale deployment of geological carbon storage.
Hydraulic fracturing, combined with horizontal drilling, enabled the boom in shale gas and oil production that has changed the global energy landscape over the past decade. The economic production of large quantities of natural gas from deep shale formations was pioneered in the United States during the mid-2000s, beginning in the Barnett Shale in Texas. Shale gas production has subsequently extended to shale formations throughout the country and grown rapidly from a negligible fraction of United States gas production in 2006 to around 60% in 2017 (EIA, 2018a). The shale gas boom has provided a huge, cheap, new natural gas resource that has led to substantial switching from coal-fired electricity generation to gas-fired electricity generation. Natural gas generation has risen from around 20% of United States electricity generation in 2006 to 34% in 2017, while coal has fallen from around 50% in 2006 to 31% in 2017 (EIA, 2018a). Natural gas-fired electricity generation emits approximately half the carbon dioxide emissions per unit of energy compared with coal-fired generation and a small fraction of the air pollutants. Coal to gas switching has been a major contributor to the approximately 25% decrease in United States electricity sector carbon dioxide emissions since 2006 (EPA, 2018b). Gas-fired electricity generators are also typically more flexible than coal-fired generators and are able to vary their output relatively quickly in time to help balance the growing share of intermittent renewable sources in the electricity grid. Therefore, if guided by appropriate policy measures, shale gas could play an important role in reducing greenhouse gas emissions and enabling the transition to a zero-emissions energy system. While the shale boom has brought energy security and greenhouse gas emissions benefits, potential local environmental impacts from shale gas development have raised significant concerns and become a contentious and prominent public issue world-wide.
(EPA, 2016; Vengosh et al., 2014; Vidic et al., 2013). Many of these concerns are centered on the practice of hydraulic fracturing. The potential for migration of injected hydraulic fracturing fluids, formation brines, and gas in the subsurface and consequent contamination of drinking water aquifers has been of particular interest and prompted a number of modeling studies aimed at assessing the risk (Birdsell et al., 2015a, 2015b; Flewelling & Sharma, 2014; Gassiat et al., 2013; Kissinger et al., 2013; Lange et al., 2013; Myers, 2012; O'Malley et al., 2016; Reagan et al., 2015). I present numerical modeling work that advances upon the previous efforts investigating the fate of hydraulic fracturing fluid injected into shale gas formations in Chapters 2 and 3 of this dissertation.

Another potential role for shale gas development in energy and climate solutions is the use of depleted shale gas formations for geological storage of carbon dioxide (Godec et al., 2013a; Nuttall et al., 2005; Tao & Clarens, 2013). Climate change mitigation assessments consistently find that carbon capture and storage (CCS) is a crucial technology needed to reduce emissions of carbon dioxide to the atmosphere sufficiently to limit warming to the two degree Celsius target of the Paris Agreement (IEA, 2017a; IPCC, 2014). These studies also conclude that the system-wide cost of decarbonizing the energy system will be lower with CCS as part of the solution. CCS, when combined with bio-energy or direct air capture, is also an important option among negative emissions technologies that may be needed to remove carbon dioxide from the atmosphere (Fuss et al., 2014; Sanchez et al., 2015).

The premise for geological carbon storage in shales is that the many existing shale wells could be used to inject carbon dioxide (rather than drilling new wells), that storage in shales may be more secure than other formations, and that carbon dioxide injection could
be used for enhanced gas recovery from shales. Previous studies have shown large static capacity estimates for carbon storage in shale gas formations (Godec et al., 2013a; Godec et al., 2014; Nuttall et al., 2005). In Chapter 4, I develop a model to investigate the dynamic capacity of shale formations for geological carbon storage.

In February 2018, the United States Congress enacted the most significant financial incentives for CCS in the world so far (Future Act, 2017). The new tax credits are sufficient to make carbon capture from low-cost industrial sources economically favorable in the near-term. Therefore, in Chapter 5, I extend my consideration of geological carbon storage more broadly and present an economic and policy analysis that examines the potential for large-scale CCS deployment as a result of the tax credits.

1.2 The Shale Gas System

Most of the work in this dissertation focuses on questions related to shale gas. Therefore, I present here a brief introductory overview of the shale gas system to give the reader a broad context of the system before the more focused modeling investigation chapters that follow. This overview is the introduction section of my paper published in the journal *Water Resources Research*, which presents collated data for the shale system that I collected throughout my dissertation research (Edwards & Celia, 2018b).

Shale gas formations are organic-rich fine-grained rocks that contain a significant amount of natural gas. (Shale formations can also contain oil, but gas-rich formations are the focus of this dissertation.) However, the shale formations have extremely low permeability and therefore hydraulic fracturing is required to create fractures that
enhance the permeability of the rocks and enable economic flow rates of gas to be extracted through wells.

Shale gas geological formations have heterogeneous mineralogy and structure (Vega et al., 2015). The rocks are characterized by extremely low permeability, low porosity, nanometer-scale pore sizes, and high organic matter content (Chalmers et al., 2012a; Clarkson et al., 2013). Gas fills the majority of the pore space and is also adsorbed to organic matter in the shales (Zhang et al., 2012). Initial water saturations are low and the resident brines are very saline (EPA, 2016). A large proportion of the rock porosity may be within the organic matter, which is often unevenly distributed in the shales (Loucks et al., 2012; Milliken et al., 2013). Gas shales therefore exhibit heterogeneous and mixed wettability, and bulk samples typically spontaneously imbibe both oil and water (Singh, 2016; Xu & Dehghanpour, 2014). The shales have very high capillary pressure due to the very small pore sizes and low water content (Engelder et al., 2014). Capillary pressure is likely the dominant driver of water imbibition in shales, although clay water absorption and osmosis may also contribute (Roshan et al., 2016b).

Gas is produced through deep wells drilled horizontally through the “pay zone” of the target shale formation. Details of drilling and completion techniques vary among different operators and locations, but are generally similar (King, 2012; NETL, 2011; J. W. Thompson et al., 2011). The horizontal well section is usually oriented normal to the expected hydraulic fracture plane of the rock. The horizontal well is perforated at regular intervals in a series of stages. In each stage, a large volume of hydraulic fracturing fluid is injected into the well at high pressure to induce hydraulic fractures in the rock. An illustrated cross-section of a typical shale gas well is shown in Figure 1.2.
Figure 1.2 – Illustration of the shale gas system showing typical quantities for system geometry parameters. Not drawn to scale. The parameter letter labels shown here are also listed for the corresponding parameters in the data tables in Appendix A.

The hydraulic fracturing fluid is typically predominantly water (so-called “slickwater”) containing proppant (usually sand) to hold fractures open, as well as a number of chemical additives that modify the fluid properties (GWPC & IOGCC, 2017; Stringfellow et al., 2014). Hydraulic fracture geometry may range from planar to complex networks, and induced fractures may connect with and re-activate existing fractures (Fisher & Warpinski, 2012; Gale et al., 2014). Hydraulic fractures are commonly monitored by microseismic methods (Fisher & Warpinski, 2012; Hammack et al., 2014), which locate fracture events in the rock and can be used to estimate the geometry, height and horizontal length of fracture networks. While microseismic monitoring is the best tool available for locating fractures, it is not perfect and may not
always capture the full extent of fracture deformation (Das & Zoback, 2013a, 2013b). Microseismic and other evidence indicate that a significant fraction of the created fractures are not effectively propped open (Kam et al., 2015; Sharma & Manchanda, 2015). These un-propped fractures close within a relatively short period of time and do not contribute significantly to gas production.

Shale gas wells are often “shut-in” for varying lengths of time, typically days to weeks, following the hydraulic fracturing operation (Bertoncello et al., 2014). Following shut-in, production of gas and water from the well commences. The production is initially dominated by returning fracturing fluid during the “flowback” period, but water flow rates typically decrease rapidly until very little water is produced (Clarkson & Williams-Kovacs, 2013). A large fraction of the injected water volume usually does not return from the formation, although the return varies among different wells and formations (Nicot et al., 2014; Vidic et al., 2013). Flowback recovery has been observed to decrease with increasing length of shut-in time (Q. Zhou et al., 2016). Gas production rates decline more rapidly compared with conventional oil and gas wells. Shale wells are therefore expected to have shorter productive lives compared with conventional wells, but lifespans remain uncertain given the relatively young age of most shale gas wells (Patzek et al., 2013).

I have collated a substantial amount of data on the shale gas system through the course of my investigations. These data are summarized in Appendix A.
1.3 Dissertation Overview

Chapters 2 to 4 of this dissertation focus on the shale gas system. Chapters 2 and 3 concern the fate of hydraulic fracturing fluid in shale gas formations. Chapter 4 transitions to investigating the potential for geological storage of carbon dioxide in shale gas formations. Finally, Chapter 5 considers geological carbon storage more broadly with an analysis of near-term pathways for scaling up carbon capture and storage in the United States.

In Chapter 2, I implement a numerical model of two-phase water and gas flow in a shale gas formation in order to test the hypothesis that the remaining water is imbibed into the shale rock by capillary forces and retained there indefinitely. This work advances upon previous efforts to use numerical models to investigate the fate of hydraulic fracturing fluid in two ways. Firstly, my model includes the essential physics required to model both water and gas flow within the shale formations and, therefore, is able to consider gas production and the fate of fracturing fluid and as inter-related processes in one system. Secondly, I apply the model to simulate water and gas production for wells from a specific location where there is sufficient available data to build and test the model. The simulations match production data from the wells remarkably closely and show that all the injected water can be accounted for within the shale system, with most imbibed into the shale rock matrix and retained there for the long term by capillary pressure forces.

The work presented in Chapter 2 was published in the journal *Environmental Science and Technology* (Edwards et al., 2017). I presented different stages of this work at the American Geophysical Union Fall Meeting, San Francisco, CA, in December 2015 and

In Chapter 3, I develop a model of capillary pressure hysteresis that is suitable for implementation in simulations of two-phase water and gas flow in shale gas formations. Experimental data show that capillary pressure in shale rocks is strongly hysteretic. The hydraulic fracturing and production processes in shale gas wells drive rapid and large changes in water saturation and capillary pressure in both time and space in shale rocks. Capillary pressure hysteresis is therefore important for accurately simulating flows in shales. However, existing capillary pressure hysteresis models do not perform well under the complex conditions in shales; they impose impractical computational restrictions and produce physically implausible capillary pressure behavior. I developed a modified version of the existing Killough (1976) model that is robust and computationally efficient for simulating flows in shale formations, and which produces very similar simulation results to the existing models under appropriate parameter selections.

A manuscript based on the work presented in Chapter 3 is currently in preparation for submission to a peer-reviewed journal (Edwards et al., 2018).

In Chapter 4, I develop a numerical model of single-phase, two-component flow of methane and carbon dioxide in a shale gas formation and apply it to assess the capacity for geological carbon storage in shales. Previous studies found large total storage capacity estimates in shale formations. However, the dynamics (rates and timescales) of carbon dioxide injection into individual shale wells, which are crucial to the practical and economic feasibility of carbon storage in shales, has not been thoroughly considered. My
model is based on shale gas production data and the associated models consistent with those data. The model is much simpler and more readily applied than complex reservoir simulators that may otherwise be used for such an evaluation. Application of the model to three shale gas-producing regions shows that carbon dioxide can only be injected at low rates into individual wells, that individual well capacity is relatively small, and, therefore, that large-scale carbon storage in shales is unlikely to be economically favorable.

The work presented in Chapter 4 was published in the journal *Environmental Science and Technology* (Edwards et al., 2015). I presented this work at the American Geophysical Union Fall Meeting, San Francisco, CA, in December 2014, and also at the Scottish Carbon Capture and Storage (SCCS) Conference, Edinburgh, United Kingdom, in October 2015.

Through the course of my research on the shale gas system in Chapters 2 to 4 of this dissertation, I collected a wide range of data from a diverse array of sources. The lack of comprehensive publicly available data is an impediment for modeling efforts on the shale gas system. Therefore, I published my collated data in the journal *Water Resources Research* to make this resource available to other researchers (Edwards & Celia, 2018b). These data are included as Appendix A of this dissertation.

In Chapter 5, I broaden my scope beyond shale gas formations with a techno-economic and policy analysis of the potential for near-term deployment of carbon capture and storage in the United States. The United States Congress enacted new and substantial tax credit incentives for carbon storage in February 2018. I analyze the pipeline infrastructure that could be deployed to maximize the impact of the tax credits for enabling and
accelerating large-scale carbon capture deployment. My methodology combines assessment of carbon emissions capture opportunities with pipeline network and economic optimization analysis, integrated with policy analysis. I demonstrate a significant potential for near-term CCUS deployment by targeting the lowest-cost existing capture opportunities. The carbon dioxide pipeline transport network would serve near-term oil industry demand for carbon dioxide for enhanced oil recovery while also connecting multiple prospective long-term dedicated geological storage resources. I identify the challenges for implementing this network and discuss potential paths forward.

At the time of writing this dissertation, a manuscript based on the work in Chapter 5 has been submitted to the journal *Proceedings of the National Academy of Sciences* and is under review (Edwards & Celia, 2018a).
Chapter 2

The Fate of Hydraulic Fracturing Fluid in Shales

This chapter is a modified version of the article Edwards et al. (2017) published in Environmental Science and Technology. The article was modified to change the writing from the plural first person (we) to singular first person (I). A new paragraph was added to the modeling methodology section (Section 2.2.3) detailing the connection between Chapter 2 and Chapter 3. Supporting information for this chapter can be found in the supporting information of the published article, except for the model parameters which are also included in Appendix B of this dissertation.

2.1 Introduction

The combination of high-volume hydraulic fracturing (HF) and horizontal drilling has enabled a boom in natural gas (and oil) production from organic-rich shales in North America over the past decade (Martineau, 2007). HF involves the injection of large volumes of aqueous HF fluid (typically around $2 \times 10^4$ m$^3$ per shale gas well) into the target shale formation at high pressure in order to fracture the rock (Gallegos et al., 2015; Jackson et al., 2015; Nicot et al., 2014; Q. Zhou et al., 2016). Figure 2.1 shows a diagram of a typical horizontal shale gas well. The HF fluid used to fracture the shale is
predominantly water (so-called “slickwater”), but also contains sand proppant to hold the created fractures open as well as chemical additives to modify the fluid properties (Vidic et al., 2013). Much of the injected HF fluid does not return from the well as “flowback” or “produced water” after gas production begins. The proportion of HF fluid that returns is commonly between 10–30% but varies among wells and formations, from as low as 5% in the Marcellus Shale to as high as 100% in the Barnett Shale (Clark et al., 2013; EPA, 2016; NETL, 2011; Nicot et al., 2014; Vidic et al., 2013; Xu et al., 2015; Q. Zhou et al., 2016). Uncertainty over the fate of the HF fluid that remains underground has driven both public concern about potential environmental contamination and interest from the oil and gas industry to determine the impact of the remaining HF fluid on gas production (Cheng, 2012; EPA, 2016; Fakcharoenphol et al., 2016; Vidic et al., 2013).
Figure 2.1 – Illustration of a horizontal shale gas well (top) with the model representation shown below it. Hydraulic fractures are created at regular intervals along the horizontal well. The fractures are represented in the model as rectangular prism fractures perpendicular to the well. The model fracture representation includes the dark gray propped inner region of the hydraulic fracture that contributes to gas flow, and the light gray un-propped outer region of the hydraulic fracture that is open during HF injection but closes afterward. The dashed orange box shows the numerical model domain. The red arrows represent gas flow out of the shale matrix during production, and the blue arrows represent HF fluid flow into the shale matrix during hydraulic fracturing. Not to scale.
From an environmental perspective, there is concern that HF fluid could migrate upward and contaminate drinking water aquifers (EPA, 2016; Gassiat et al., 2013; Kissinger et al., 2013; Lange et al., 2013; Myers, 2012; Vengosh et al., 2014; Vidic et al., 2013). Chemical additives typically comprise 0.5–2% of the HF fluid, and while most of the commonly used chemicals are biodegradable and/or non-toxic, many others are of unknown toxicity, and some are toxic and/or confirmed or suspected carcinogens (EPA, 2016; GWPC & IOGCC, 2017; Konschnik & Dayalu, 2016; Stringfellow et al., 2014, 2017; Wattenberg et al., 2015). Once injected into the shales, the HF fluid mixes with the resident brines that often contain metals, salts, and radionuclides (Chapman et al., 2012; EPA, 2016; Haluszczak et al., 2013; Parker et al., 2014). Most North American shale gas formations are deep (the mean HF depth is 2,500 meters) with substantial vertical separation distance between the top of the hydraulic fractures and drinking water aquifers (often 1,000 meters or more) (Davies et al., 2012; EPA, 2016; Fisher & Warpinski, 2012; Hammack et al., 2014; Jackson et al., 2015). Observational and modeling studies indicate that HF fluid is very unlikely to migrate upward directly through such a substantial geological barrier (Birdsell et al., 2015a; Flewelling & Sharma, 2014; Hammack et al., 2014; Kissinger et al., 2013; Lange et al., 2013). However, natural subsurface features, such as faults and fractures, and man-made features, such as oil and gas wells, could potentially provide permeable pathways for upward flow, especially if intersected by hydraulic fractures.

Several numerical modeling studies have found that HF fluid could migrate upward along permeable pathways from deep shale formations to drinking water aquifers under certain conditions (Gassiat et al., 2013; Kissinger et al., 2013; Lange et al., 2013; Myers, 2012).
Those studies have been notably criticized for unrealistic model conditions because, among other reasons, they are one-phase models that do not include the two-phase flow process of capillary water imbibition into the shales (Cohen et al., 2013; Engelder, 2012; Flewelling & Sharma, 2014, 2015; Saiers & Barth, 2012). Gas-producing shales are typically water-wet, have low water saturation, and have very small pores with associated very high capillary pressure, which suggests that water injected into fractures in those rocks would spontaneously imbibe into the shale rock “matrix” (Engelder, 2012; Engelder et al., 2014; Ghanbari & Dehghanpour, 2015; Roshan et al., 2016a; Xu & Dehghanpour, 2014; L. Zhou et al., 2016; Z. Zhou et al., 2016c). One prominent hypothesis proposes that capillary imbibition draws the injected HF fluid into the shale, that the capillary pressure will retain the HF fluid in the shale for the long term, and that imbibition explains the low proportion of HF fluid return (Engelder, 2012; Engelder et al., 2014; Ghanbari & Dehghanpour, 2016; Singh, 2016). Thus, in order to evaluate the potential for HF fluid to migrate, it is critical to understand and quantify imbibition behavior. If all the injected HF fluid is imbibed and retained in the shale formations, it will not be free to migrate elsewhere.

HF fluid imbibition is also of great interest from the perspective of maximizing gas production from shale wells. Imbibed water near the fracture-shale matrix interface occupies pore space and impedes the flow of gas into the fractures (sometimes called “water blocking” in the oil and gas industry) (Bertoncello et al., 2014; Holditch, 1979; Mahadevan & Sharma, 2005; Z. Zhou et al., 2016c). While there is a large body of oil and gas industry-focused literature investigating the imbibition of water into shales, these studies mainly concentrate on the implications of imbibition for hydraulic fracturing
design and gas production rather than the questions of migration potential and the ultimate fate of the HF fluid (Bertoncello et al., 2014; Binazadeh et al., 2016; Cheng, 2012; Dehghanpour et al., 2012, 2013; Ewy, 2014; Fakcharoenphol et al., 2016; Gdanski et al., 2009; Ge et al., 2015; Ghanbari & Dehghanpour, 2015, 2016; Holditch, 1979; Liu, et al., 2016; Mahadevan & Sharma, 2005; Mahadevan et al., 2007; Makhanov et al., 2014; Ren et al., 2016; Roshan et al., 2015, 2016b; Roychaudhuri et al., 2013; Singh, 2016; Wang & Rahman, 2016; Xu & Dehghanpour, 2014; Yan et al., 2015; L. Zhou et al., 2016; Z. Zhou et al., 2016a, 2016b, 2016c). Several studies show a large amount of water could be stored in the fractures or matrix of shale formations, but they do not consider how mobile that water will be, particularly in the long term (Birdsell et al., 2015b; Ghanbari & Dehghanpour, 2016; Makhanov et al., 2014; O'Malley et al., 2016; Ren et al., 2016; Xu et al., 2015). In addition to capillary pressure, clay water absorption/hydration and osmosis have been investigated as additional drivers of imbibition (Binazadeh et al., 2016; Dehghanpour et al., 2012, 2013; Ewy, 2014; Fakcharoenphol et al., 2016; Ge et al., 2015; Z. Zhou et al., 2016c), but capillary pressure appears to be the main driver of imbibition and the most likely to be prevalent under in-situ conditions (Roshan et al., 2015, 2016b).

Birdsell et al. (2015b) quantified capillary imbibition of HF fluid into shales using a one-dimensional, incompressible, semi-analytical model of imbibition from a fracture face into the shale matrix. Their study found that a large proportion of the injected HF fluid could imbibe, assuming typical shale formation and HF fluid parameters. However, the Birdsell et al. (2015b) imbibition model has significant simplifications limiting its ability to represent the flow of water and gas within shale gas formations. Their simulations only
consider imbibition during a 5-day shut-in period after HF injection, and the constant-saturation fracture-face boundary condition is not able to represent the condition in the fracture during shut-in and the subsequent production period, as water flows out of the fracture. Furthermore, the assumption of incompressible fluids means that the model cannot simulate gas compressibility during high-pressure HF injection, or gas flow and pressure drawdown during production. These factors can have significant effects on the movement of HF fluid in the shale formation, and their omissions make the model of limited use in answering questions about both the initial imbibition of the HF fluid into the shale matrix, and particularly the long-term fate of the HF fluid within the shale formation.

In the work reported in this Chapter, I implement a model of two-phase water and gas flow in a shale gas formation including fluid compressibility and fracture water dynamics. I aim to determine the long-term distribution of injected HF fluid in the shale, and whether it is plausible that all injected HF fluid remains in the shale. I consider gas production and HF fluid flow behavior as inter-related processes within one physical system, and compare my model simulations with externally observable data from that system. I apply the model to shale wells in the Horn River Basin in British Columbia, Canada, for which high-quality water and gas production data are available. I investigate the following questions: how much HF fluid could be imbibed into the shale, and over what timescale? What is the long-term fate of imbibed HF fluid after the initial injection period? Is it plausible that all the HF fluid remains in the shale formation? Is the hypothesized capillary imbibition of HF fluid compatible with observed water and gas production from shale gas wells?
2.2 Methods

2.2.1 Modeling Approach

I implement a two-phase (water and gas) numerical model of a shale gas formation and apply the model to simulate wells from a specific well pad in the Horn River Basin where there is sufficient information available to build and test the model. The aim of my modeling is to quantitatively and qualitatively explain the long-term behavior of injected HF fluid within the shale formation while being consistent with the externally observable water and gas production data. I develop a model containing the simplest justifiable geometry, physical processes, and parameters. I constrain the system geometry and select parameters using directly observed well and geological formation data from the Horn River Basin. I also calibrate unknown and uncertain parameters while being constrained by the Horn River data, as well as corresponding data from similar shale formations.

2.2.2 Model Data

My study uses data for 12 wells located on one well pad in the Horn River Basin. The wells are drilled into the adjacent Muskwa and Otter Park members of the Horn River Formation (Abbasi, 2013; Chen & Hannigan, 2016; Kam et al., 2015). Early-time (first 2-6 weeks) water and gas production data with hourly time resolution, in addition to long-term (4 years) gas production data with monthly time resolution, are available for the wells (Abbasi, 2013; BCOGC, 2016; Ghanbari & Dehghanpour, 2016). The raw short-term data are noisy and have been smoothed by a weighted average for presentation in the figures. Additional available well data include depth, horizontal well lateral length, fracture (well perforation) spacing, hydraulic fracturing injection pressure, injection flow
rate, total injected fluid volume, and well pressure during production. Geological formation data include thickness, porosity, initial water saturation, gas composition, gas pressure, and temperature. More information about the data and sources is included in Appendix A and in the supporting information of Edwards et al. (2017).

### 2.2.3 Modeling Methodology

Shale gas production data from thousands of wells in several major formations have been modeled remarkably accurately by simple one-dimensional, one-phase, continuum-scale, diffusive (Darcy flux) natural gas flow in a finite domain (Edwards et al., 2015; Male et al., 2014, 2016; Patzek et al., 2013, 2014; Silin & Kneafsey, 2012). The implied conceptual model for the shale gas formation is a homogeneous shale matrix between parallel, vertical, planar hydraulic fractures spaced at regular intervals along the horizontal well (illustrated in Figure 2.1). This planar fracture model was shown by others to most closely match production data (in comparison to other fracture representations) for Horn River Formation wells nearby to those considered in this study (Kam et al., 2015). I adopted the homogeneous shale matrix, planar fracture conceptual model and extended it to two-phase (water and gas) flow.

I began my modeling by using the one-dimensional, one-phase gas flow model to obtain the unknown shale formation parameters by history-matching simulated gas production with the observed production data. The unknown parameters (effective matrix permeability and hydraulic fracture area) were also constrained by permeability, formation thickness, and fracture size data for the Horn River Formation (Ahmed & Ehlig-Economides, 2013; BCOGC, 2016; Chen & Hannigan, 2016; Kam et al., 2015;
All other required model parameters were known from the available well and formation data (Abbasi, 2013; BCOGC, 2016). Each well on the well pad has nearly identical characteristics except for the spacing between hydraulic fracturing perforations along the horizontal wells: some of the wells have 25-meter spacing between fractures, while others have 40-meter spacing (Abbasi, 2013).

Cumulative natural gas production data for each of the wells is shown in Figure 2.2 with the history-matched one-phase model simulations. Cumulative production data from the wells clearly fall into two groups of higher and lower-producing wells. The excellent history-match of the one-dimensional model simulations to both groups of production data were achieved by varying only the fracture spacing (25-meter and 40-meter) while holding all other well and formation parameters constant. Although production data for thousands of wells across different shale formations have been matched closely using the one-dimensional model representation, there are some other wells for which the one-dimensional model appears to be too simple (Edwards et al., 2015; Male et al., 2014, 2016; Patzek et al., 2013, 2014; Silin & Kneafsey, 2012). These include 6 wells on the same Horn River well pad that are drilled into the underlying Evie member of the Horn River Formation. The production data from the Evie wells were able to be matched using a more complex three-dimensional geometrical model representation; further details are included in the supporting information of Edwards et al. (2017). The following work focuses on the Muskwa and Otter Park member wells.
The history-matched formation parameters from the one-phase modeling (effective matrix permeability and hydraulic fracture area) were used as input for the two-phase model. I used the two-phase model to simulate the shale gas well completion and production process. The process includes three distinct periods: (1) the hydraulic fracturing period, where HF fluid is injected into the well at high pressure; (2) the shut-in period, when the well is closed for a period of time following HF; and (3) the production period, when the well is opened, the pressure inside the wellbore decreases, and water and gas flow into the wellbore (Abbasi, 2013; BCOGC, 2016; Ghanbari & Dehghanpour, 2016; Merkle et al., 2013). The two-phase numerical modeling was performed using the open-source MATLAB Reservoir Simulation Toolbox (MRST) automatic differentiation two-phase black oil model (Krogstad et al., 2015; Lie, 2016). Although the HF fluid and
resident formation brine have some differing properties, they are miscible and represented by a single water phase in the model (described interchangeably as “water” or “HF fluid”). I assume water is initially near residual saturation within the shale.

The hydraulic fracture was represented in the model as a rectangular prism (a three-dimensional extension of a plane) with a defined aperture and resolved as grid cells with volume, porosity, and permeability. Explicitly resolving the fractures was important for the system behavior because storage and flow of water within the fracture volume is significant in the water dynamics. The numerical model domain included half of the aperture of one hydraulic fracture and the adjacent shale matrix region extending to the plane of symmetry with the neighboring fracture (illustrated in Figures 2.1 and 2.3). Fracture volume in the model was static (i.e. initiation, growth, and closure were not modeled). The implicit assumption is that fractures are created instantaneously at the beginning of injection (justified by microseismic data showing that fractures are mostly created early in the HF injection period), and that the fracture volume does not change with time (Ciezobka et al., 2016). An illustration of the numerical grid structure used in the simulations is shown in Figure 2.3, with the fracture and matrix grid cells identified. There were no-flow boundaries on all sides of the model. Fluids were injected and extracted from the model domain through a pressure-dependent source/sink in the central hydraulic fracture cell representing the well (see Figure 2.3). Water was injected through the well cell at high pressure during the HF period, and the cell was a low-pressure sink that drew in both water and gas during the production period.
Figure 2.3 – Schematic illustration of the model domain and hydraulic fracture model representation and discretization. The illustration is not to scale and does not show the actual number or spacing of grid cells, but it is representative of the structure. More detailed information about the model discretization is included in the supporting information of Edwards et al. (2017).

A substantial amount of literature indicates that the created area of hydraulic fractures is much larger than the area that is “propped” open by sand proppant and able to effectively contribute to gas production (Ahmed & Ehlig-Economides, 2013; Cipolla & Wallace, 2014; Kam et al., 2015; Merkle et al., 2013; Sardinha et al., 2014; Sharma & Manchanda, 2015). The remaining “un-propped” fracture area closes after HF injection and does not contribute significantly to production (Merkle et al., 2013; Sharma & Manchanda, 2015). I interpret the hydraulic fracture area that was history-matched for the one-phase gas production modeling as the propped fracture area. The larger un-propped fracture area (and volume), however, is important for water flow and imbibition during HF injection. I therefore modified the conceptual model of the shale gas formation for the two-phase model by increasing the area and volume of the initially created hydraulic fractures. Fracture size was increased in the simplest way: by extending the rectangular prism
fractures with a narrower-aperture un-propped fracture region outside of the inner propped fracture region (illustrated in Figures 2.1 and 2.3). The permeability of the un-propped fracture region was decreased immediately at the end of the HF injection period (with the implicit assumption being that the fractures close immediately), based on evidence that the fracture closure time-scale is short compared to my simulation duration, and measurements of closed un-propped fracture permeability for the Horn River Formation (supported by data from other formations) (Cipolla & Wallace, 2014; Kam et al., 2015; Merkle et al., 2013; Sharma & Manchanda, 2015; Wu et al., 2017; Zhang et al., 2014). The permeability of the closed un-propped fractures is sufficiently low that the outer region effectively does not contribute to flow during the production period. Propped fracture permeability was also decreased, more moderately, to account for the fracture walls closing on the proppant. The simple fracture representation in the model was effective for capturing the long-term behavior of the system that is the focus of my study, but created limitations for matching the very early-time (initial days) behavior where fracture dynamics are important. These limitations are discussed further in the results and the supporting information of Edwards et al. (2017).

The model of two-phase flow in a shale gas formation implemented for my simulations is a homogeneous shale matrix, rectangular prism fracture model with un-propped and propped hydraulic fracture regions. The model was implemented in MRST to simulate the completion and production procedure for the specific wells I am modeling: a 4-hour HF injection, followed by a 60-day shut-in, and then indefinite gas production (Ahmed & Ehlig-Economides, 2013; BCOGC, 2016; Ghanbari & Dehghanpour, 2016). Most additional formation parameters required for the two-phase model (capillary pressure,
relative permeability, residual saturations, hydraulic fracture height and width) were
selected based on data for the Horn River Formation and other shale gas formations
(Ahmed & Ehlig-Economides, 2013; Bennion & Bachu, 2008; Chalmers et al., 2012b;
Kam et al., 2015; Sigal, 2013). Hydraulic fracture aperture (volume) and permeability
were the key parameters varied to history-match water injection and production while
also being guided by typical reported values for these parameters (Ezulike et al., 2016;
Kam et al., 2015; Xu et al., 2015; Zhang et al., 2014). The total fracture volume in my
model (2.4–3.9×10⁴ m³) is consistent with independent estimates of total fracture volume
for neighboring Horn River Formation wells (2.8–6.3×10⁴ m³) (Ezulike et al., 2016;
Xu et al., 2015). HF injection pressure and the production bottom-hole well pressure (as a
function of time) are the external driving force parameters for the system, and were
known from measured data (Abbasi, 2013; BCOGC, 2016). The water and gas fluid
phases were compressible, with the non-linear gas pressure-density and pressure-
viscosity relationships prescribed by the known shale formation temperature and gas
composition.

The capillary pressure model was a key component of the overall model that strongly
influences water imbibition behavior. Capillary pressure in shales has been shown to be
strongly hysteretic (Kale et al., 2010; Sigal, 2013). However, I found that the water and
gas production data could be matched more closely using a non-hysteretic capillary
pressure model and therefore decided to use non-hysteretic capillary pressure. The non-
hysteretic model is a conservative case for investigating the fate of hydraulic fracturing
fluid, since hysteresis acts to increase the drainage capillary pressure and therefore retain
water more strongly in the rocks once it imbibes. There are a number of possibilities
related to fracture geometry and properties of the rocks and fracturing fluid that may cause the non-hysteretic capillary pressure model to provide a better match to the data despite the system actually being strongly hysteretic. For example, additives in the hydraulic fracturing fluid break down with time to reduce fluid viscosity and surface tension, enabling greater recovery of the water. This would reduce the drainage capillary pressure and therefore reduce the effect of hysteresis. Hysteretic capillary pressure models are investigated in Chapter 3. For the work in this Chapter, capillary pressure was modeled using the non-hysteretic van Genuchten (1980) equation with parameters matched to shale capillary pressure imbibition data (Chalmers et al., 2012b; Kale et al., 2010; Sigal, 2013).

A full list of parameter values is included in Appendix B. Further information on the two-phase model is included in the supporting information of Edwards et al. (2017), including more information on the data sources, the one-phase model, the parameter history-matching procedure, and parameter sensitivity.

2.3 Results and Discussion

The two-phase model results match the gas and water production data closely. Figure 2.4 shows simulated long-term cumulative gas production compared with data for the individual Horn River Formation wells. The simulated gas production curves are very similar to the one-phase model simulations presented in Figure 2.2. However, the one-phase model required an artificial adjustment to early-time (first two weeks) production rate to account for reduced flow rates due to two-phase flow effects during early-time. The two-phase model simulation needed no such adjustment. Figure 2.5 shows simulated
gas production rate during the first 50 days of production compared with individual well data. Gas production is initially suppressed by water impeding the flow of gas from the shale, but increases with time during the first 10 days of the simulation as water is removed. The simulated initial production rate is lower than the data, but the simulation reproduces two key common characteristics of the data: the peak gas production rate after about 10 days and the subsequent rate of decline in production. The close agreement of the model to gas production rate data is more evident in Figure 2.6, which compares the simulation result with less noisy, longer-term, monthly time resolution data (in addition to the early-time, hourly resolution data).

Figure 2.4 – Cumulative natural gas production (billion standard cubic feet) versus time since the beginning of gas production for individual wells (orange lines) compared with two-phase model simulated production with 25-meter fracture spacing (black line) and 40-meter fracture spacing (green line).
Figure 2.5 – Natural gas production rate (thousand standard cubic feet per day) versus time since the beginning of gas production for individual wells (yellow-orange-red lines) compared with two-phase model simulated gas production with 25-meter fracture spacing (black line) and 40-meter fracture spacing (green line) for the first 50 days of production.

Figure 2.6 – Natural gas production rate (thousand standard cubic feet per day) versus time since the beginning of gas production for individual wells, showing early-time hourly resolution data (yellow-orange lines) and longer-term monthly resolution data (red lines) compared with two-phase model simulated gas production with 25-meter fracture spacing (black line) and 40-meter fracture spacing (green line) for the first two years of production.
Simulated water production rate during the first 50 days of production is shown in Figure 2.7 along with individual well data. After an initial pulse of water within the first day, simulated water production rate increases for the first 5 days of production, followed by a peak, and then a continuously decreasing rate. Similar to the gas production, simulated water production matches the qualitative and quantitative characteristics of the data except at very early time. The very early-time gas and water behavior could be matched under some parameter scenarios, but not without compromising the amount of water that could be injected and the longer-term quantity of water production. Since the focus of my modeling is to determine the plausibility of all water being retained within the shale long-term, it was more important to match the water quantities and long-term behavior. This limitation of the model is due to the simplified fracture representation and is further explained in the supporting information of Edwards et al. (2017).
Figure 2.7 – Water production rate (liters per second) versus time since the beginning of gas production for individual wells (light-dark blue lines) compared with two-phase model simulated water production with 25-meter fracture spacing (black line) and 40-meter fracture spacing (green line) for the first 50 days of production. The truncated water rate for the 25-meter spacing simulation at very early time reaches 6.1 liters per second.

The long-term water balance for the simulated shale system is presented in Figure 2.8, which shows the distribution of the injected water volume between the fractures and shale matrix with time throughout the HF injection, shut-in, and production. Total volumes of water injected into the system during the 4-hour HF injection period were $5.7 \times 10^4$ m$^3$ for the 25-meter spacing model and $3.5 \times 10^4$ m$^3$ for the 40-meter spacing model. These volumes were matched to the average of $4.8 \times 10^4$ m$^3$ actually injected into the 12 Horn River Formation wells (Abbasi, 2013). During the injection period, a substantial fraction of the water imbibes into the shale matrix, but the majority (about 70%) remains in the fractures. During the 60-day shut-in period, water continues imbibing from the fractures into the shale matrix. The total amount of water in the system does not change, but its distribution within the system changes. At the commencement of
the production period, water flows from the fractures into the well and the total amount of water in the system declines. About 17% of the injected volume is ultimately produced from the well. There is also a small decrease of water in the shale matrix at the beginning of production due to some water flowing from the matrix back into the fractures driven by the very high pressure gradient toward the fractures. However, after a short period, the pressure gradient reduces sufficiently and capillary-driven imbibition of water from the fractures into the shale matrix resumes (even as gas flow continues in the opposite direction). Within two years, almost all remaining mobile water has imbibed from the fractures into the shale matrix.

**Figure 2.8** – Simulated shale system water balance for the 25-meter fracture spacing (40-meter is almost identical), showing water volume as a percentage of total injected water volume versus time. The graph shows the total amount of water in the system (grey) and the distribution of water within the shale system between the fractures (dark blue) and the shale matrix (light blue). The vertical dashed black line shows the transition between the shut-in period (2) and the production period (3). The HF injection period (1) is 4 hours at the start of the simulation, which can only be seen in this graph by the near-vertical increase in water volumes near time zero.
My simulations match the observations from the wells using a model that includes the essential physics of the system but maintains a simple structure. The results also show that all the water injected into the wells can be accounted for within the shale system, with most of the injected water stored in the shale rock matrix in the long term. I observe that the simulated total injected water volume and early-time water production rate are dominated by the storage and flow of water in the hydraulic fractures, and are therefore sensitive to the fracture geometry, volume, permeability, and closure timescale. However, the long-term distribution of water within the system (imbibition into the shale rock matrix) is insensitive to both these hydraulic fracture parameters and different capillary pressure parameters. The parameter sensitivity is discussed further in the supporting information of Edwards et al. (2017). Since the focus of my investigation is on the long-term behavior of the system, I conclude that the model captures the dominant physics of the system, with a strong focus on capillary imbibition, phase pressures, and compressibilities.

I note here that the amount of water injected into the 12 Horn River Formation wells (average $4.8 \times 10^4$ m$^3$) was considerably higher than is typical for the major shale gas plays in the United States (around $2 \times 10^4$ m$^3$ per well) (Gallegos et al., 2015; Jackson et al., 2015; Nicot et al., 2014; Q. Zhou et al., 2016). While the Horn River Formation is particularly thick, the ability to match the unusually high volume adds confidence that typical injected volumes can be accommodated in shale formations. I also note that the simple rectangular prism fracture representation is a conservative model for investigating the plausibility of water remaining in the shale formation because more complex fractures with increased fracture surface area and volume would enable more imbibition and water
storage within the system. Another important note is that the no-flow boundaries of the model meant that water could only remain within the shale system, so my simulations show the plausibility of all water remaining in the shale, rather than the impossibility of water flowing out of the shale. Water could flow out through fractures during the HF period, but the long-term water dynamics within the model suggest that long-term upward water flow is unlikely. Any mobile water present in the fractures will imbibe into the shale matrix. Furthermore, pressure drawdown in the formation due to gas production means that even if fractures extend out of the shale formation and contain mobile water, the direction of water flow will be into the shale, rather than out, until the formation pressure re-equilibrates in the very long-term.

The simulations also revealed important characteristics regarding the driving forces and timescale of water imbibition into the shale matrix. During the 4-hour HF injection period, forced, pressure-driven imbibition dominated due to the high-pressure water injection compressing gas in the shale matrix. About 50% of the total imbibition into the shale was pressure-driven. After HF injection, during shut-in and production, spontaneous capillary-driven imbibition occurred, but at a considerably slower rate than the pressure-driven imbibition during HF injection, and over a much longer time-scale. Most previous studies that quantified water imbibition into the shale matrix used incompressible fluid models and therefore could not capture the important role of pressure-driven imbibition during HF injection (Birdsell et al., 2015a, 2015b; Ren et al., 2016; Wang & Rahman, 2016). Regardless of the driving force of imbibition, capillary forces retain imbibed water in the shale matrix. In particular, the low capillary pressure in the hydraulic fractures compared to the shale matrix (due to the large difference in pore
sizes) results in a strong capillary pressure gradient into the matrix. Once water has imbibed, capillary forces similarly drive the water to move even farther into the shale. Water saturation distribution graphs that illustrate the imbibition processes are included in the supporting information of Edwards et al. (2017).

2.3.1 Environmental Implications

My simulations of two-phase flow in a shale formation support the hypothesis that injected HF fluid is imbibed into the shale matrix and retained there in the long term by capillary forces. HF fluid residing in the fractures at the end of HF will be imbibed into the shale in the long term and will not be able to migrate elsewhere, even if a permeable pathway is present. The model results are robust for a wide range of parameters: even a halving of capillary pressure does not significantly change the system behavior and sees almost all mobile water imbibed into the shale within two years. Other factors besides capillary imbibition also decrease the likelihood that HF fluid will migrate upward in the long term (as has been shown by others), including the lack of strong buoyant drive (compared with gas), a very short-lived pressure drive during HF, and long-term pressure drawdown during gas production that will draw fluids toward the well (Flewelling & Sharma, 2014).

However, the simulation results do not imply that HF fluids cannot flow out of shale formations under any circumstances. The time-scale of capillary imbibition of water into the shale matrix is much longer than the HF injection period. Therefore, the HF injection period, when a high-pressure driving force exists and water in the fractures is mobile, is the highest risk period for water migration. If a direct hydraulic connection is present or
created during HF, water could flow along that pathway. For example, water could migrate along deficient well cement or an adjacent well if the hydraulic fractures intersect that well. Where evidence of HF fluid migrating out of the target formation exists, those pathways have been implicated (DiGiulio & Jackson, 2016; EPA, 2016; ERCB, 2012). Furthermore, while HF fluid may be unlikely to migrate out of the formation, natural gas is both buoyant and not spontaneously imbibed like water, so it is more likely to migrate in the subsurface. Modeling and field observations support this hypothesis (Darrah et al., 2014; Jackson et al., 2013; Molofsky et al., 2013; Osborn et al., 2011; Reagan et al., 2015; Vengosh et al., 2014).

I applied my simulations to one well pad in the Horn River Formation and consequently my results are only directly applicable there. The imbibition behavior of other shales will vary depending on differences in permeability, porosity, fracture network size and geometry, and rock mineralogy and wettability. However, the major shale gas formations in North America, such as the Barnett, Marcellus, Haynesville, and Fayetteville shales, all share characteristics similar to the Horn River Formation shales, including depth, well design, HF techniques, and many shale rock properties (see data tables in the Appendix A) (NETL, 2011; Nieto et al., 2009; Petzet, 2008). In particular, all available literature indicates that shale gas formations have similar pore size, wettability, and capillary pressure characteristics, and therefore capillary imbibition behavior (Chalmers et al., 2012a, 2012b; Clarkson et al., 2013; Dehghanpour et al., 2013; Engelder et al., 2014; Sigal, 2013; L. Zhou et al., 2016; Z. Zhou et al., 2016c). Since my results are robust for a large range of capillary parameters, the implications are therefore generally relevant for similar deep gas shales with large vertical separation between the shale and aquifers.
My analysis was possible due to the public availability of high-quality data for oil and gas operations in British Columbia, Canada. More high-quality, publicly available data in the United States would enable similar analyses to be performed directly for gas shales there, where the most public concern has been raised over potential environmental impacts.
Chapter 3

Modeling Capillary Pressure Hysteresis in Shales

3.1 Introduction

In Chapter 2, I modeled the injection of hydraulic fracturing fluid and subsequent production of water and gas in the shale gas system. I showed that capillary pressure is a key factor determining the movement of water in shale formations. The capillary pressure model is therefore an important element of a simulation of water and gas flow in shale formations. In the simulations in Chapter 2, I found that a simple, non-hysteretic capillary pressure model enabled the closest match of simulated water and gas flow to the data. However, experimental data show that capillary pressure in shales is hysteretic (Kale et al., 2010; Sigal, 2013). Figure 3.1 shows capillary pressure data from samples of shale gas formations. These data are strongly hysteretic. The data show that the primary drainage capillary pressure curves are significantly higher than the imbibition capillary pressure curves, and that the shales have substantial non-wetting phase residual saturations. In this Chapter, I develop a hysteretic capillary pressure model suitable for the computational demands of modeling water and gas flow in shale gas formations. The
model follows from the original ideas of Florian Doster, who is a collaborator and co-author on the work.

The hydraulic fracturing and subsequent fluid production processes inject and remove significant quantities of water from shale gas formations and drive very large, rapid changes in pressure and water saturation in the shale rocks, particularly close to the hydraulic fracture faces. There are multiple reversals in direction of water saturation change for particular locations within the rocks, and there are also different directions of saturation change at different locations in the rocks. I found that the existing, commonly used capillary pressure hysteresis models, such as those by Kool and Parker (1987) and Killough (1976), do not perform well under these conditions: they require impractically short time-steps and produce physically implausible capillary pressure behavior.

Therefore, I modified the Killough (1976) model to simplify the capillary pressure

![Figure 3.1](image-url)  

**Figure 3.1** – Capillary pressure as a function of water saturation for data measured from the Barnett Shale by Sigal (2013) and Kale et al. (2010). The raw mercury intrusion capillary pressure data were converted to hydraulic fracturing fluid-gas capillary pressure as described in Edwards and Celia (2018b).
hysteresis behavior. The modified model produces very similar simulation results to the existing models, while enabling much faster computational performance and eliminating undesirable capillary behavior.

3.2 Methods

3.2.1 Modeled Shale System

Figure 3.2 shows an illustration of the shale gas system and our model representation of the system. Hydraulic fractures are initiated at regular intervals along the horizontal wells, and the wells are oriented such that the fractures open predominantly in a vertical plane perpendicular to the wells. The model assumes planar, parallel, evenly spaced hydraulic fractures with a homogeneous shale rock matrix between the fractures. The fractures are assumed to have permeability several orders of magnitude higher than the adjacent shale matrix, so pressure is essentially equal at all points within the fractures compared with the shale. Under these conditions, water and gas flow in the shale is one-dimensional, perpendicular to the fracture planes (shown in Figure 3.2). Therefore, water and gas flow can be modeled as one-dimensional, two-phase, continuum-scale, diffusive (Darcy flux) porous media flow in a finite domain. The model has no-flow boundaries at the midpoints between fractures, representing the planes of symmetry. A narrow (2.5 mm) set of cells immediately adjacent to one boundary represent the high-permeability hydraulic fracture, while the rest of the model cells represent the low-permeability shale matrix. Water and gas are injected and extracted through a pressure-dependent source/sink in the hydraulic fracture cells to drive the system.
Figure 3.2 – Illustration of a horizontal shale gas well (top) with the model representation shown below it. Hydraulic fractures are created at regular intervals along the horizontal well. The model fractures are planar, parallel, and perpendicular to the well. Water and gas flow in the shale matrix is modeled as one-dimensional flow perpendicular to the fracture planes (shown by the blue arrows), between a fracture and the plane of symmetry with its adjacent fracture (indicated by the dashed orange plane). Not drawn to scale.

This type of one-dimensional model of fluid flow in shale gas formations has been shown to simulate gas production data from thousands of wells in multiple major shale gas formations remarkably accurately (Edwards et al., 2015; Male et al., 2014, 2016; Patzek et al., 2013, 2014; Silin & Kneafsey, 2012). In Chapter 2, I showed that this model also simulates two-phase water and gas production data accurately. The model presented in
Chapter 2 is three-dimensional, which was needed in order to account for the total volume of hydraulic fracturing fluid injected into the system. In this Chapter, however, I am focused on developing a model of hysteretic capillary pressure behavior rather than matching production data and therefore use the simpler one-dimensional model.

The modeled shale gas system includes a low permeability shale matrix with a low initial water saturation near the residual saturation. The modeled shale well completion and production process includes three distinct periods: (1) a three-hour hydraulic fracturing period, where water is injected into the fracture at high pressure; (2) a one-day shut-in period, when the well is closed and no fluids are injected or extracted; and (3) a one-day production period, when the well is opened, the pressure in the hydraulic fractures decreases, and water and gas flow into the fracture from the shale matrix (Abbasi, 2013; BCOGC, 2016; Ghanbari & Dehghanpour, 2016; Merkle et al., 2013). The simulated duration of the injection period corresponds to typical industry practice (Ahmed & Ehlig-Economides, 2013; BCOGC, 2016; Ciezobka, 2013; Ciezobka et al., 2016; Neuhaus & Miskimins, 2012). The shut-in and production simulation durations are much shorter than typical time periods in practice. These durations were selected to encompass the period of greatest rate of water saturation and capillary pressure change, which are the key intervals for the performance of the hysteretic capillary pressure model. This allows the capillary behavior to be highlighted in the results. The reversals in direction of water saturation change and associated capillary pressure hysteresis are driven by the changes in the driving forces of the system during the different periods; I do not impose imbibition or drainage a priori. The result is a complex pattern of changes from imbibition to drainage.
in both time and space within the shale matrix domain. This represents a stringent test for the capillary hysteresis model.

The two-phase numerical modeling was performed using the open-source MATLAB Reservoir Simulation Toolbox (MRST) automatic differentiation two-phase black oil model (Krogstad et al., 2015; Lie, 2016). The injected hydraulic fracturing fluid and the resident formation brine are represented by a single water phase in the model (described interchangeably as “water” or “hydraulic fracturing fluid”). Both the water and gas phases are compressible. A full list of model parameters is included in Appendix C.

3.2.2 Existing Capillary Pressure Hysteresis Models

Capillary pressure arises in porous media with two or more immiscible phases that have differential preference for wetting the solid surfaces: a pressure differential arises across the interface between the phases, which is called the capillary pressure. Capillary pressure is typically mathematically expressed at the continuum-scale as a function of wetting-phase saturation, \( p_c = p_c(s_w) \), most commonly with equations developed by Brooks and Corey (1964) and van Genuchten (1980). Hysteresis implies the capillary pressure depends not only on the current state of phase saturations in the system, but also on its history. Capillary pressure hysteresis is observed in many porous media (Albers, 2014), including shales. Capillary pressure hysteresis occurs due to the underlying geometry of pores in the porous medium, with the larger pores invaded first under drainage and the smallest invaded first under imbibition. Differences in fluid wetting angles depending on direction of flow, and the disconnection of one fluid phase as it is displaced by another, may also play a role (Albers, 2014). Since the physical phenomena
that cause capillary pressure hysteresis are below the resolution of the continuum-scale
models in which we wish to model hysteretic behavior, up-scaled, parameterized, and
simplified mathematical representations of hysteresis are required.

The fundamentals of capillary pressure hysteresis and hysteretic models are illustrated in
Figure 3.3. The solid red curve is the primary drainage curve that capillary pressure
follows for decreasing wetting-phase saturation from the fully saturated state. The solid
blue curve is the primary imbibition curve that capillary pressure follows for increasing
wetting-phase saturation from the fully unsaturated state. (In the illustrated case, I assume
there is no residual saturation and therefore the primary and main drainage and imbibition
curves are identical.) The dashed lines show a scanning curve loop for a case where
capillary pressure is initially following the main imbibition curve, but then the direction
of saturation change reverses, or turns, at saturation $s_{t,1}$ and capillary pressure follows the
dashed red scanning drainage curve. A second reversal of direction occurs at $s_{t,2}$ and
capillary pressure follows the dashed blue scanning imbibition curve, which converges
with the main imbibition curve at $s_{t,1}$. Depending on the history of saturation in the
system, capillary pressure may be anywhere between the primary imbibition and drainage
curves for a particular saturation. In the case of the shale gas system, water is the wetting
phase and gas is the non-wetting phase.
Figure 3.3 – An illustration of capillary pressure hysteresis for a hypothetical system with zero residual phase saturations, showing capillary pressure versus wetting-phase saturation. The solid red curve is the primary or main drainage capillary pressure curve, the solid blue curve is the primary or main imbibition curve. The dashed lines show a scanning curve loop for a case where capillary pressure is initially following the main imbibition curve, then the direction of saturation change turns at saturation $s_{t,1}$ and capillary pressure follows the scanning drainage curve. Saturation again turns at $s_{t,2}$ and capillary pressure follows the scanning imbibition curve. The scanning imbibition curve converges with the main imbibition curve again at $s_{t,1}$.

The basis of numerical capillary pressure hysteresis models is to remember turning saturations ($s_{t,1}$ and $s_{t,2}$ in Figure 3.3) and to calculate the scanning curves based on these points. Some hysteresis models also require the capillary pressure at the turning saturations to be stored. Two of the most commonly used capillary pressure hysteresis models, and their limitations for modeling the shale gas system, are described in the following sections.
3.2.2.1 Kool and Parker Model

The Kool and Parker (1987) model calculates scanning curves by scaling the main drainage and imbibition curves based on the turning saturations and capillary pressures. The main drainage and imbibition curves are described using the van Genuchten (1980) capillary pressure function:

\[ p_c(s_w) = p_c^e \left( s_w^{1/m} - 1 \right)^{1/n} \]  

where \( p_c^e \) is the capillary entry pressure, and \( m \) and \( n \) are parameters that determine the curve shape. The drainage and imbibition curves are defined by separate sets of parameters.

During a simulation, the previous turning saturation and capillary pressure is stored for each cell and updated each time the direction of saturation change in a cell turns. In each timestep, the scanning drainage and imbibition curves are calculated for each cell by scaling the main curves based on the stored turning saturation and capillary pressure. The cell capillary pressure is calculated and assigned appropriately depending on whether the cell saturation is greater than or less than the turning saturation (i.e. in a state of imbibition or drainage).

Two characteristics of the Kool and Parker (1987) model created problems for simulating hydraulic fracturing fluid injection and production in shale gas formations. Firstly, the model scanning curves do not form closed loops because the scaling of the main curves to generate scanning curves does not consider any previous scanning curve paths. This means that “pumping” can occur if there are multiple saturation turns, which is illustrated...
in Figure 3.4. Such pumping is not observed in experimental data of capillary pressure scanning curves (Kool & Parker, 1987). This pumping behavior occurs when using the Kool and Parker (1987) capillary pressure model in the shale gas system simulations, shown in Figure 3.6. Secondly, the large change in the capillary pressure scanning curve gradient immediately after saturation turns, while consistent with experimental observations, presents a computational challenge for the Newton’s method solver in MRST that requires very short timesteps.

3.2.2.2 Killough Model

The Killough (1976) model calculates scanning curves using a function that interpolates between bounding main drainage and main imbibition curves. The interpolation function shape is governed by a fitting parameter. The function requires memory of the turning saturation at which the direction of saturation first turned from the main drainage or imbibition curve, as well as any second turning saturation, and the saturation in the previous timestep. The scanning imbibition curve equation of the Killough (1976) model is:

$$p_{ci}^{si}(s_w) = p_{ci}^{md}(s_w) - F^{d-i}(s_w)(p_{ci}^{md}(s_w) - p_{ci}^{mi}(s_w))$$

(3.2)

where $p_{ci}^{si}$ is the scanning imbibition capillary pressure at saturation $s_w$, $p_{ci}^{md}$ is the main drainage capillary pressure, and $p_{ci}^{mi}$ is the main imbibition capillary pressure. The main drainage and imbibition curves can be calculated by any capillary pressure function; I use the van Genuchten (1980) capillary pressure function (Eq. 3.1). $F^{d-i}$ is the interpolation function for scanning imbibition:
\[ F^{d-i}(s_w) = \frac{1}{s_w - s_w^{trn} + \sigma} - \frac{1}{\sigma} \]

where \( s_w^{trn} \) is the turning saturation from the main drainage curve, \( s_w^{max} \) is the maximum possible wetting-phase saturation considering the residual non-wetting saturation, and \( \sigma \) is the fitting parameter. The scanning drainage curve equation is:

\[ p^c_{sd}(s_w) = p^c_{mi}(s_w) + F^{i-d}(s_w)(p^c_{md}(s_w) - p^c_{mi}(s_w)) \]

where \( F^{i-d} \) is the weighting factor for scanning drainage:

\[ F^{i-d}(s_w) = \frac{1}{s_w^{trn} - s_w + \sigma} - \frac{1}{\sigma} \]

where \( s_w^{tr} \) is the residual wetting-phase saturation.

The model is initialized on either the main imbibition or main drainage curve. At a turn in saturation, capillary pressure follows a scanning curve. If saturation turns again while on the scanning curve, it follows a secondary scanning curve that forms a closed loop terminating at the point where the first scanning turned from the main curve. The scanning loop is illustrated in Figure 3.4. The secondary scanning curve is calculated by a similar, but more complex, method to the initial scanning curves shown above, which uses the two stored bounding saturation turns. If a third saturation turn occurs within the bounds of a scanning curve loop, capillary pressure jumps to the other scanning curve. An example of such a jump is illustrated in Figure 3.4. This feature ensures that capillary pressure scans within loops and avoids the pumping issue observed in the Kool and
Parker (1987) model. However, the Killough (1976) model has similar large changes in the capillary pressure scanning curve gradient immediately after saturation turns and also has abrupt changes in both capillary pressure gradient and value when jumps occur between scanning curves. This behavior is observed in the shale gas system simulations using the Killough (1976) capillary pressure model, shown in Figure 3.6, and requires even smaller timesteps for the Newton’s method solver in MRST than the Kool and Parker (1987) model.

![Capillary pressure scanning loops](image)

**Figure 3.4** – Capillary pressure scanning loops for the Kool and Parker (1987) and Killough (1976) hysteresis models. The Kool and Parker (1987) model does not have closed loops so capillary pressure pumping occurs. The Killough (1976) model has closed scanning curve loops, but abrupt jumps occur when changing between scanning imbibition and drainage curves.
3.2.3 Modified Capillary Pressure Hysteresis Model

Given the shortcomings of the Kool and Parker (1987) and Killough (1976) capillary pressure hysteresis models for simulating the shale gas system, I developed a modified hysteresis model with the goal of significantly improving computational performance while reasonably representing hysteretic capillary pressure behavior. There are two key desirable characteristics for the modified model: firstly, that it does not exhibit pumping upon multiple saturation turns, and secondly, that there are no abrupt changes in the gradient or quantity of capillary pressure that are restrictive for timesteps.

I adopted the framework of the Killough (1976) model with a different interpolation function, as suggested by Florian Doster (personal communication, 2017). The modified interpolation function has a single scanning curve for drainage or imbibition, which prevents pumping and jumps between curves. A single, smooth, scanning curve is followed regardless of how many saturation turns occur. The modified interpolation function also has a gradual deviation of capillary pressure gradient upon a saturation turn from the main drainage or imbibition curve, rather than the immediate and large change in gradient in the Kool and Parker (1987) and Killough (1976) models. The modified model adopts the scanning imbibition and drainage equations of the Killough (1976) model (Equation 3.2 and 3.4), and uses the following interpolation function:

\[ F = \frac{(s_h^\alpha)^n}{(s_h^\alpha)^n + (1 - s_h^\alpha)^n} \]  

(3.6)

where \( n \) is a fitting parameter and \( s_h^\alpha \) is a scaled wetting-phase saturation, with \( \alpha = i \) for imbibition or \( \alpha = d \) for drainage. The equations for \( s_h^i \) and \( s_h^d \) are:
\[
s_h^i = \begin{cases} \frac{s_w - s_w^{trn}}{\varepsilon(s_{w}^{max} - s_w^{trn})}, & \frac{s_w - s_w^{trn}}{\varepsilon(s_{w}^{max} - s_w^{trn})} \leq 1 \\ 1, & \frac{s_w - s_w^{trn}}{\varepsilon(s_{w}^{max} - s_w^{trn})} > 1 \end{cases} \tag{3.7}
\]

\[
s_h^d = \begin{cases} \frac{s_w^{trn} - s_w}{\varepsilon(s_{w}^{trn} - s_w^{r})}, & \frac{s_w^{trn} - s_w}{\varepsilon(s_{w}^{trn} - s_w^{r})} \leq 1 \\ 1, & \frac{s_w^{trn} - s_w}{\varepsilon(s_{w}^{trn} - s_w^{r})} > 1 \end{cases} \tag{3.8}
\]

where \( \varepsilon \) is a fitting parameter. The modified capillary pressure hysteresis model is demonstrated in Figure 3.5 for a hypothetical scanning drainage scenario. The \( F \) interpolation function and scanning curves show a gradual deviation from the main imbibition curve, which varies depending on the model parameters. The \( \varepsilon \) parameter controls the steepness of the scanning curve. The steepness could be selected based upon experimental data or the requirements of the particular scenario being modeled: for example, a steeper scanning curve is more appropriate for the scanning drainage example case in Figure 3.5, since it more closely approximates a typical scanning drainage curve. The \( n \) parameter controls the gradient at which the scanning curve initially deviates from the main curve. A greater value of \( n \) results in a curve that deviates more gradually from the main curve, which would improve computational performance for Newton’s method solvers and allow greater timesteps. However, a more gradual deviation is also further from the scanning behavior demonstrated in experiments and displayed by existing models, so the model performance benefits of the \( n \) parameter choice must be balanced with the reality of the model.
Figure 3.5 – A demonstration of the modified capillary pressure hysteresis model for a scenario where capillary pressure initially follows the main imbibition curve, followed by a saturation turn at $s_w = 0.7$ and then decreasing wetting-phase saturation in a scanning drainage event. The $F$ function for the modified model under different parameter cases (top) and the resulting capillary pressure scanning curves for those parameters (bottom) are shown. All parameter cases use $n = 2$, except for one case that is indicated where $n = 3$. A comparison with the Killough (1976) model is also shown.

3.3 Results and Discussion

My new modified capillary pressure hysteresis model and the existing Kool and Parker (1987) and Killough (1976) models were each used separately in identical simulation scenarios for the shale gas system, as described in Section 3.2.1. Simulated capillary pressure at a location 10 mm from the hydraulic fracture face for the three model cases are shown in Figure 3.6. The modified model displays smooth capillary pressure behavior
throughout the simulation, while the Kool and Parker (1987) model displays oscillations and pumping, and the Killough (1976) model shows abrupt jumps in capillary pressure. Consequently, the modified model has a substantial computational advantage compared with the two existing models in these simulations. A case with no capillary pressure hysteresis required 892 timesteps for the simulation. The modified capillary pressure hysteresis model required 1,366 timesteps, while the Kool and Parker (1987) model required around 21,000 timesteps and the Killough (1976) model required around 57,000 timesteps.

![Capillary pressure versus saturation in a simulation modeling hydraulic fracturing fluid injection, shut-in, and production, at a point in the shale matrix 10 mm from the hydraulic fracture face. Output for the three different hysteresis models are shown. Our new modified hysteresis model is shown by the blue line and exhibits a smooth curve throughout the simulation. The magnified inset graph shows that the Kool and Parker (1987) model displays oscillations and capillary pressure pumping, while the Killough (1976) model displays an abrupt jump.](image)

**Figure 3.6** – Capillary pressure versus saturation in a simulation modeling hydraulic fracturing fluid injection, shut-in, and production, at a point in the shale matrix 10 mm from the hydraulic fracture face. Output for the three different hysteresis models are shown. Our new modified hysteresis model is shown by the blue line and exhibits a smooth curve throughout the simulation. The magnified inset graph shows that the Kool and Parker (1987) model displays oscillations and capillary pressure pumping, while the Killough (1976) model displays an abrupt jump.
While delivering substantial computational advantage, the modified capillary pressure hysteresis model was also able to produce very similar results to the existing models with appropriate parameter selections. Figure 3.7 shows simulated water saturation profiles in the shale rock matrix at different times of the simulation and for the different capillary pressure models. Figure 3.7(A) shows the pattern of water saturation change throughout the simulation for the modified model: at the end of the hydraulic fracturing injection, a sharp front of water has flowed into the shale rock matrix from the hydraulic fracture at the left-hand side boundary. At the end of the shut-in period, this front of water has redistributed, with water saturation decreasing nearer to the fracture (drainage) and increasing further from the fracture (imbibition). Once production commences, some of the water flows back into the hydraulic fracture (drainage) and the water saturation throughout most of the shale matrix decreases by the end of the simulation, except for a small amount of continued imbibition at the edge of the water saturation front.
Figure 3.7 – Shale matrix water saturation versus horizontal distance from the center of the hydraulic fracture for simulations of hydraulic fracturing fluid injection, shut-in, and production, showing the following capillary pressure model cases: (A) Our modified hysteresis model at the end of injection, shut-in, and the end of the simulation. (B) A comparison of our modified model with the Kool-Parker and Killough models at the end of shut-in. (C) A comparison of our modified model with the Kool-Parker and Killough models at the end of the simulation period. (D) A comparison of our modified hysteresis model under different parameter scenarios, and a case with no hysteresis, at the end of the simulation period.

Figures 3.7(B) and (C) show a comparison between the saturation profiles simulated by the modified model and the existing models at the end of the shut-in period and end of the overall simulation, respectively. The modified capillary pressure hysteresis model
produces very similar results to the existing models. The impact of different parameter choices in the modified model is shown in Figure 3.7(D) for the same parameter cases shown in Figure 3.5. The cases with a higher $\varepsilon$ parameter value (with corresponding shallower scanning curves) produce a greater amount of redistribution of the injected water and do not match the saturation profiles of the existing models as closely as the best-fit lower $\varepsilon$ parameter choice.

Figure 3.7(D) also shows a comparison case with no hysteresis, where capillary pressure can only follow the main imbibition curve shown in Figures 3.5. Capillary pressure hysteresis results in less redistribution of water compared to the non-hysteretic case, and therefore leads to increased water saturation persisting close to the fracture face. This behavior is of particular interest from the perspective of maximizing gas production from shale gas formations, since increased water saturation at the fracture face results in lower gas relative permeability and therefore lower production rates. The occurrence is termed “water blocking” in the oil and gas industry, and is the target of well completion strategies and chemical additives that aim to minimize its effect. By causing more retention of injected water, capillary pressure hysteresis also implies that injected hydraulic fracturing fluids are more likely to remain in the shale formations and not be able to migrate upward toward groundwater aquifers.

Suitable models for capillary pressure hysteresis in shale gas formations are therefore important for modeling shales from both production optimization and environmental perspectives. My modified capillary pressure hysteresis model is a robust and computationally efficient method for including capillary pressure hysteresis in simulations of water and gas flow for systems with multiple saturation turns, such as
shale gas formations. Parameters can be selected for the model that produce behavior consistent with the currently accepted hysteresis models.

I note that, while the modified model produces results consistent with the existing hysteresis models, the inclusion of hysteresis in the simulations of water and gas production shown in Chapter 2 did not match the data as closely as a non-hysteretic model. There are a number of possible reasons for this result: hysteresis in-situ may not be as strong as shown by laboratory experiments on shales, or hysteresis may be important but other factors related to fracture geometry and the properties of the rocks and fracturing fluid may overwhelm the hysteresis effect in the observed whole-well water and gas production. Some of these factors known to be prevalent in shales include complex fracture geometry, mixed rock wettability, and changing water viscosity and surface tension with time as hydraulic fracturing fluid additives break down to reduce capillary pressure and enable greater recovery of the injected water.

Understanding and modeling of capillary pressure hysteresis in shale gas formations can be improved through further modeling and experimental work. Water and gas production from other wells and shale gas formations can be simulated, with availability of the necessary data, to determine whether the apparent limited effect of hysteresis is widespread or particular to the wells I modeled in Chapter 2. Further capillary pressure experiments could be conducted on samples from different shale formations to determine the importance of hysteresis in these rocks, and particular attention could be paid to investigating scanning curve behavior to determine if the proposed modified hysteresis model is an acceptable compromise between accuracy and improved computational performance.
Chapter 4

The Potential for Carbon Dioxide Storage in Shales

This chapter is a modified version of the article Edwards et al. (2015) published in Environmental Science and Technology. The article was modified to change the writing from the plural first person (we) to singular first person (I). Several sentences were added to the introduction detailing the connection to the work presented in Chapters 2 and 3. Supporting information for this chapter can be found in the supporting information of the published article, except for the model parameters that are also included in Appendix D of this dissertation.

4.1 Introduction

Deep geological formations have been proposed and investigated as potential reservoirs in which to inject CO$_2$ for long-term storage. Storage of CO$_2$ in this manner is considered one of the most feasible near-term options to reduce emissions to the atmosphere (IPCC, 2005). CO$_2$ has also been routinely injected into geological formations for the purpose of enhanced hydrocarbon recovery for several decades (Orr & Taber, 1984). Injection for both CO$_2$ storage and enhanced hydrocarbon recovery has involved relatively permeable formations such as depleted conventional oil and gas reservoirs and deep saline aquifers.
Despite extremely low permeability, other favorable characteristics of organic-rich gas shales have prompted investigation of injection of CO₂ into these formations for both purposes (Godec et al., 2013a; Liu et al., 2013; Tao & Clarens, 2013).

Booming production of natural gas (the main constituent of which is CH₄) from organic-rich shales over the past decade has been enabled through the technologies of horizontal drilling and high-volume hydraulic fracturing. These technologies enable access to a larger volume of a target rock layer by a single well and significantly enhance flow of gas to the well (Martineau, 2007). Figure 4.1 shows a schematic of a typical hydraulically fractured horizontal well.
Natural gas exists in organic-rich shales as free gas in pores and as adsorbed gas on solid surfaces. Adsorption primarily occurs in the organic matter within the rock rather than the inorganic minerals (Zhang et al., 2012). Research suggests that a significant fraction (20 – 60%) of CH$_4$ stored in gas shales is adsorbed under natural conditions, and only a relatively small portion of this is produced during the lifetime of a shale gas well (Cipolla et al., 2010; NETL, 2011). Studies have shown that significantly more CO$_2$ adsorbs to the
organic-rich shales than CH₄ (Chareonsuppanimit et al., 2012; Godec et al., 2013a; Heller & Zoback, 2014; Weniger et al., 2010), and that CO₂ preferentially adsorbs over CH₄ when present in a mixture (Billemont et al., 2014; Dreisbach et al., 1999; Kurniawan et al., 2006; Ottiger et al., 2008). CO₂ injected into a shale gas reservoir would likely preferentially adsorb to the organic matter and displace CH₄ into the gas phase. This characteristic, along with the large number of shale gas wells drilled in the past decade and associated volume of accessible pore-space, has led to suggestions of possible injection of CO₂ into depleted shale gas reservoirs for geological storage and/or enhanced gas recovery. In the modeling in Chapters 2 and 3, gas adsorption was not included in our models since the focus was on hydraulic fracturing fluid. In the case of CO₂ injection in shales, however, gas adsorption is an important component that must be included in the model.

Recent studies have reported large capacity estimates for storage of CO₂ in shale gas reservoirs. Tao and Clarens (2013) estimated that 10.4 – 18.4 gigatonnes (Gt) (1 Gt = 10¹⁵ g) of CO₂ could be stored in the Marcellus Shale by 2030; Godec et al. (2013a) estimated 55 Gt could be sequestered in the Marcellus, with this number revised to 49 Gt in a subsequent study (Godec et al., 2014); and Nuttall et al. (2005) estimated that 28 Gt could be sequestered in the Devonian shales underlying Kentucky. Given that United States (US) annual anthropogenic CO₂ emissions are approximately 6.5 Gt per year (EPA, 2014), approximately 50% of which are from fixed-point sources and could potentially be captured, these capacity estimates for shales are significant. Although they identify a substantial total capacity for CO₂ storage, the aforementioned studies do not thoroughly investigate the dynamics (rates and timescales) of CO₂ injection into
individual shale wells. Regardless of total capacity, CO$_2$ must be cost-effectively injected into the wells at the required rates (the rate of CO$_2$ capture) for shale gas formations to be an attractive storage target.

This study aims to improve on the previous assessments of CO$_2$ storage potential by developing a well-scale model of gas flow in a shale reservoir that can easily be applied to investigate CO$_2$ injection dynamics in shale gas wells. The mathematical model is based on natural gas production data and the associated models consistent with those data. This model captures the most important physical processes for CO$_2$ injection (bulk gas flow, component transport, and adsorption of CO$_2$ and CH$_4$), while being relatively simple and readily applied to any shale gas play by choosing appropriate parameters and calibrating to natural gas production data. The model enables investigation of injection dynamics and allows for different injection scenarios, providing transient system responses and total (ultimate) capacity for individual wells.

4.2 Methods

4.2.1 Mathematical Model

The model presented herein of gas flow to a hydraulically fractured horizontal well is based on analyses of natural gas production data in the Barnett Shale (Patzek et al., 2013; Silin & Kneafsey, 2012). Patzek et al. (2013) analyzed data from 8,284 wells in the Barnett and observed that scaled gas production rates initially decline inversely proportional to the square root of time before transitioning to exponential-in-time decline at later times. This behavior is consistent with the solution to a one-dimensional diffusion equation in a finite domain, with a fixed-pressure boundary condition at one end of the
domain allowing for outflow and a no-flow boundary at the other end. Barnett Shale production data were shown to fit very well to this model (Patzek et al., 2013). The implication is that, despite complicated fracture geometries, highly heterogeneous rock structure at the small scale, and the presence of two phases (gas and brine), at the whole-well scale the flow of gas in the Barnett Shale behaves as simple single-phase, one-dimensional Darcy flow in an effectively homogeneous medium, between parallel, planar hydraulic fractures.

Figure 4.1 shows the implied geometry of the model and is broadly representative of the typical structure of shale gas wells: a horizontal well with perforations at regular intervals and approximately planar fractures perpendicular to the well. The well and hydraulic fractures both have essentially infinite permeability compared to the shale matrix and therefore pressure in the well and fractures is effectively equalized, resulting in the horizontal one-dimensional flow behavior between adjacent fractures. The following nonlinear diffusion equation describes the one-dimensional gas flow (Patzek et al., 2013):

\[
\frac{\partial}{\partial t} \left[ \phi S_g \rho^b (p) + \rho^a (p) \right] - \frac{\partial}{\partial x} \left( \frac{\kappa \rho^b (p) \partial p}{\mu^b (p) \partial x} \right) = 0
\]

(4.1)

where \( \phi \) is the rock porosity (volume of voids per total volume of rock), \( S_g \) is the gas saturation (volume of gas per volume of voids), \( \rho^b \) is the bulk free gas density (mass of gas per volume of gas), \( \rho^a \) is the adsorbed gas density (mass of adsorbed gas per total volume of rock), \( \kappa \) is the effective permeability (relative permeability multiplied by intrinsic permeability), \( \mu^b \) is the bulk gas viscosity, and \( p \) is the pore gas pressure. The gas density and viscosity, and the adsorbed density, are dependent on pressure (and temperature, which is assumed to be constant), leading to the nonlinear nature of the
equation. Gas saturation (and therefore relative and effective permeability) is assumed to be constant. The equation describes flow of a bulk gas in the shale matrix, accounting for both the bulk gas phase and adsorbed phase. The domain is between one hydraulic fracture and the plane of symmetry that occurs at half the distance to the adjacent fracture. This domain is repeated by symmetry along the length of a horizontal well (Figure 4.1). The applicability of the single-phase model implies that the brine phase is essentially immobile (after a short initial period where fracturing fluids flow back out of the well, which is not modeled). Rock compressibility is assumed to be negligible compared to the summed effect of gas compressibility and adsorption. Gas flow either out of the shale reservoir (production) or into the reservoir (injection) may be simulated with different model initial and boundary conditions.

In the case of CO₂ injection into a depleted shale gas well, natural gas will initially be present in the shale reservoir at relatively low pressure due to the prior production of the gas. CO₂ is then injected at high pressure into the well. The gas in the pores becomes a mixture of CO₂ and CH₄, and the properties of the bulk gas now depend not only on the pressure, but also on the composition of the bulk gas (the fraction of CO₂ in the gas). The movement of both the CO₂ and natural gas components within the bulk gas, and particularly their competition for adsorption sites, are now important in the system dynamics. The original single-phase, single-component model is therefore extended to a single-phase, two-component model for CO₂ injection modeling. Natural gas is represented by only its main component, CH₄, since CH₄ constitutes approximately 84% of natural gas on average. The model involves a system of two equations, one for each of the two components. I write these two equations as a pressure equation (Eq. 4.2), which
is the sum of the two component equations, and the CO\textsubscript{2} component transport equation (Eq. 4.3):

\[
\frac{\partial}{\partial t} \left[ \phi_S g b (\omega_c, p) + \rho_c^a (\omega_c, p) + \rho_m^a (\omega_c, p) \right] - \frac{\partial}{\partial x} \left[ \frac{\kappa \rho_b (\omega_c, p) \partial p}{\mu_b (\omega_c, p) \partial x} \right] = 0
\]

(4.2)

\[
\frac{\partial}{\partial t} \left[ \phi_S g b (\omega_c, p) \omega_c + \rho_c^a (\omega_c, p) \right] + \frac{\partial}{\partial x} \left[ \rho_b (\omega_c, p) u^b \omega_c \right] - \frac{\partial}{\partial x} \left[ \phi_S g b (\omega_c, p) D_c \frac{\partial \omega_c}{\partial x} \right] = 0
\]

(4.3)

In these equations, \( \omega_c \) is the mass fraction of CO\textsubscript{2} in the bulk gas phase, \( \rho_c^a \) is the adsorbed density of CO\textsubscript{2}, \( \rho_m^a \) is the adsorbed density of CH\textsubscript{4}, \( D_c \) is the CO\textsubscript{2} dispersion coefficient, and \( u^b \) is the Darcy flux of the bulk gas phase, defined as:

\[
u^b = \frac{\kappa}{\mu_b (\omega_c, p)} \frac{\partial p}{\partial x}
\]

(4.4)

The primary variables are the pressure of the gas and the mass fraction of CO\textsubscript{2}. Given the mass fraction of CO\textsubscript{2} in the bulk gas, the mass fraction of CH\textsubscript{4} is easily calculated since both components must completely fill the available pore space:

\[
\omega_c + \omega_m = 1
\]

(4.5)

The domain is finite \((0 < x < d)\), with a no-flow boundary at the plane of symmetry \((x = d)\) and fixed CO\textsubscript{2} mass fraction and either a fixed pressure or mass flux at the fracture face \((x = 0)\). The initial condition is a uniform pressure in the domain with no CO\textsubscript{2} present. Mathematical descriptions of the initial and boundary conditions are in the supporting information of Edwards et al. (2015).
4.2.2 Fluid Properties

To solve the model equations, the density and viscosity of the CO₂-CH₄ gas mixture must be calculated as a function of both pressure and composition of the gas. The bulk gas density is calculated using a Peng-Robinson equation of state (Gasem et al., 2001), with a mixing rule applied to determine parameters for the CO₂-CH₄ mixture based on the pure-component parameters and the composition of the mixture (Sandler, 2006). The viscosity of the bulk gas is similarly calculated by applying a mixing rule to pressure-dependent functions of viscosity for CO₂ and CH₄ (Herning & Zipperer, 1936).

4.2.3 Adsorption Model

Sorption behavior of CH₄ and CO₂ in porous media has been shown to fit well to the Langmuir adsorption model (Heller & Zoback, 2014; Zhang et al., 2012), which has been used as an adsorption model in studies estimating CO₂ storage capacity in shales (Godec et al., 2013a; Liu et al., 2013; Nuttall et al., 2005). The standard Langmuir model is:

\[ n = n^{max} \frac{Kp}{1 + Kp} \]  \hspace{1cm} (4.6)

where \( n \) is the amount of gas adsorbed (number of moles, or mass), \( n^{max} \) is the maximum amount that is adsorbed when all adsorption sites are occupied, and \( K \) is the Langmuir constant, which is the inverse of the pressure at which half the adsorption sites are occupied. The Langmuir function calculates the absolute amount of gas adsorbed. For systems where two gas components are present in a mixture and competing for adsorption sites, the two-component Langmuir model can be used to estimate adsorption of each component:
\[ n_1 = n_1^{max} \frac{K_1 p_1}{1 + K_1 p_1 + K_2 p_2} \]  \hspace{1cm} (4.7)

where \( n_1 \) is the amount of component 1 adsorbed in a binary system of component 1 and 2. Other parameters are analogous to those for Eq. 4.6, except \( p_1 \) and \( p_2 \) are the partial pressures of each of the component gases. The two-component Langmuir model has been shown to provide a good empirical estimate for CO\(_2\)-CH\(_4\) adsorption in porous activated carbon (Dreisbach et al., 1999), although it is not strictly theoretically applicable for most binary systems (Ruthven, 1984).

While the Langmuir model has been shown to model adsorption of CH\(_4\) and CO\(_2\) in porous media well, it is not applicable under all conditions for calculation of the total mass (free and adsorbed) of a gas in a porous medium. Since the adsorbed phase occupies a fraction of the porosity that would otherwise be filled with gas, the reduction of gas-filled porosity must be taken into account. In adsorption measurements, the results are often reported by assuming a constant porosity and introducing the concept of ‘excess adsorption’ (Heller & Zoback, 2014), which is the mass of a gas present in a porous medium in excess of the mass that would be present if there were no adsorbed phase and the entire porosity was filled with gas at a given density (Myers & Monson, 2002; Zhou et al., 2003; Zhou & Zhou, 2009). Mathematically, a simple expression for excess adsorption is the following:

\[ n^e = n^t - V_a \rho^b \]  \hspace{1cm} (4.8)
where $n^e$ is the excess amount adsorbed, $n^t$ is the total (absolute) amount adsorbed, $V_a$ is the volume of pore space occupied by the adsorbed phase, and $\rho^b$ is the density of the gas phase.

From Eq. 4.8 it follows that absolute adsorption and excess adsorption are equal when the product of adsorbed phase volume and density of the bulk gas phase is negligible compared to the absolute adsorbed amount. This is usually true at relatively low pressures (less than 5 MPa) under which experimental adsorption measurements are often performed. However, in shale gas formations, pressure (typically 20 – 40 MPa) and therefore gas density are relatively high. Adsorbed phase volume can also be relatively high in porous media with extremely small pores and correspondingly high surface area to pore volume ratio, such as shales. These effects mean that excess adsorption is significantly lower than absolute adsorption in the conditions prevalent in shale gas formations, which has been demonstrated in high-pressure adsorption experiments on organic-rich shales (Chareonsuppanimit et al., 2012; Weniger et al., 2010). This is particularly evident for CO$_2$, which has a sharp increase in density at 7 – 8 MPa as it transitions to the supercritical state. Figure 4.2 shows excess and absolute adsorption isotherms for pure CO$_2$ and CH$_4$ using adsorption data from shale gas formations. Methane deviates less from its absolute adsorption isotherm than CO$_2$ due to its relatively lower density at high pressure compared to CO$_2$. 
Figure 4.2 – Absolute (solid lines) and excess (dashed lines) adsorbed mass per cubic meter of rock as a function of gas pressure for CO$_2$ (red) and CH$_4$ (blue) using typical parameters from samples of shale gas formations (Nuttall et al., 2005; Weniger et al., 2010; Zhang et al., 2012).

Figure 4.2 demonstrates that application of a Langmuir absolute adsorption isotherm to calculate the total mass of CO$_2$ in a shale gas formation without adjusting the gas-filled porosity leads to a significant over-estimation of CO$_2$ adsorption and therefore total storage capacity (50 – 80% for our simulations). An excess adsorption function must be used to ensure the total mass in the domain is calculated consistently. Most experimental CO$_2$ adsorption measurements on samples from US shale gas formations have been performed at relatively low pressure (0 – 5 MPa) (Heller & Zoback, 2014; Nuttall et al., 2005; NYSERDA, 2011), below the CO$_2$ critical point, over which range the excess adsorption isotherms often appear to follow standard Langmuir isotherm curves. Langmuir function parameters fitted to these isotherms and extrapolated to shale-formation pressures are more reflective of absolute adsorption than excess adsorption. Assuming the reported Langmuir parameters describe absolute adsorption isotherms, the
two-component Langmuir isotherm equation can be modified to estimate excess adsorption as follows:

\[
\rho_1^e = \rho_1^{max} \frac{K_1 p_1}{1 + K_1 p_1 + K_2 p_2} \left( 1 - \frac{\omega_1 p^b(\omega_1, p)}{\omega_1^a(\omega_1, p) \rho_a} \right)
\]  

(4.9)

where \(\rho_1^e\) is the excess adsorbed mass of component 1 per unit volume of rock.

Adsorption is now expressed as a bulk density rather than an amount (as in Eq. 4.6 and Eq. 4.7). In Eq. 4.9, \(p^b\) is the density of the bulk gas and \(\rho_a\) is the adsorbed phase density, which is assumed to be constant. The adsorbed phase density can be estimated from experimental excess adsorption data (Weniger et al., 2010). \(\omega_1^a\) is the adsorbed mass fraction of component 1:

\[
\omega_1^a = \frac{\rho_1^{abs}(\omega_1, p)}{\rho_1^{abs}(\omega_1, p) + \rho_2^{abs}(\omega_1, p)}
\]  

(4.10)

where \(\rho_1^{abs}\) is the absolute adsorption of component 1 (Eq. 4.7) per volume of rock.

Eq. 4.9 is applied for each of the two components in the system. Details of the derivation of Eq. 4.9 beginning with Eq. 4.8 are given in the supporting information of Edwards et al. (2015). This method for modifying the Langmuir model has been shown to fit measurements of excess adsorption on organic-rich shales reasonably well (Gasparik et al., 2012; Weniger et al., 2010), and is applied as a model of two-component, competitive excess adsorption in my shale reservoir model.
4.2.4 Numerical Solution

The governing equations (Eq. 4.2 and Eq. 4.3) are solved numerically using a finite difference approximation. The approximation is cell-centered, with flux parameters upstream-weighted. A modified Picard iteration is applied to iterate the nonlinear coefficients and to enforce mass balance following the method of Celia et al. (1990). The pressure and transport equations are solved simultaneously and implicitly. This scheme was found to be necessary to prevent mass balance errors due to the nonlinear nature of the equations. The numerical solution is implemented in MATLAB. Further details and equations for the numerical solution scheme are included in the supporting information of Edwards et al. (2015).

4.2.5 Parameter Selection and History-Matching

In order to use the shale reservoir model to perform realistic simulations of CO$_2$ injection into shale gas wells and compare different shale plays, model parameters were history-matched to natural gas production data. Two major US shale gas plays were selected for analysis: the Barnett Shale in Texas, and the Marcellus Shale in Pennsylvania (PA). The Barnett Shale was selected because it was the first shale gas play to be commercially developed using horizontal wells and high-volume hydraulic fracturing, has the longest production history, and was the subject of the analyses that form the basis for this modeling approach (Patzek et al., 2013; Silin & Kneafsey, 2012). The Marcellus Shale was selected because it is the largest shale play, with the greatest potential CO$_2$ storage capacity, and a number of large stationary CO$_2$ sources (coal-fired power plants) overlie the formation. The Marcellus was split into two different regions for this analysis,
southwest and northeast PA, because the formation is much thicker in the northeast than southwest and consequently wells in the northeast are significantly more productive. The delineation of each region is included in the supporting information of Edwards et al. (2015).

Natural gas production data from all producing wells in each of the regions that began production within a certain time span (2007 – 2011 for the Barnett; 2010 – 2014 for the Marcellus) were obtained through Drilling Info, Inc (Drillinginfo, 2014). The data were then filtered to remove vertical wells, failed wells with very little production and wells with discontinuous production history; approximately 12% of wells were removed from the total dataset. The final sample included 8,951 wells in the Barnett Shale, 1,449 wells in southwest PA and 2,516 wells in northeast PA. Production data were averaged among wells commencing production within particular subset time periods in each region and model reservoir parameters were history-matched to the averaged data. Wells were grouped by age because shale horizontal well design has evolved (e.g. longer laterals, closer fracture spacing) leading to more productive wells over time. A range of parameter fits was made to account for the changing average well design and uncertainty in reservoir parameters. More detailed descriptions of the data selection and filtering and parameter history-matching procedures are included in the supporting information of Edwards et al. (2015).

The parameter history-matching was performed by running the model in single-component mode ($\omega_c = 0$) for natural gas production. Natural gas fluid properties were used and boundary and initial conditions set appropriately for a gas production scenario (i.e. low pressure at the fracture boundary and high initial shale reservoir pressure). The
model was fitted by adjusting two groups of fitting parameters: an ‘interference time’, \( \tau \), that dictates the ease of fluid flow through the shale (Eq. 4.11), and an ‘original mass of gas in place’ term, \( M \), that dictates the size of the reservoir and amount of gas accessible by the horizontal well (Eq. 4.12):

\[
\tau = \frac{d^2 \mu^b \left[ \phi S_g \frac{\partial \rho^b}{\partial p} + \frac{\partial \rho^a}{\partial p} \right]}{k \rho^b} \tag{4.11}
\]

\[
M = (n_f 2d) a_f (\phi S_g \rho^b + \rho^a) \tag{4.12}
\]

where \( d \) is half the distance between adjacent hydraulic fractures, \( n_f \) is the number of hydraulic fractures and \( a_f \) is the surface area of each hydraulic fracture plane. All fluid parameters are evaluated at initial reservoir pressure and temperature. The term \( n_f 2d \) represents the total length of the horizontal well lateral, which is known, so in practice \( n_f \) is set by the fracture spacing and horizontal lateral length.

Most parameters were either set to typical play parameters obtained from peer-reviewed and industry literature (reservoir pressure, temperature, porosity, gas saturation and adsorption), governed by physical properties of the fluids (density and viscosity), or set from known well data (well lateral length). The remaining parameters (permeability, fracture spacing and fracture surface area) were varied to fit the data, although these too were guided by typical values from the literature. Some selected parameters include porosity of 6% in all regions, effective permeability of 45 nanoDarcy in the Barnett and 20 – 25 nanoDarcy in the Marcellus, and average horizontal well length of 872 m in the Barnett, 1,556 m in southwest PA, and 1,675 m in northeast PA. More detailed information about data sources is included in the supporting information of Edwards et al.
and all selected parameters are included in Appendix D. The fitted shale reservoir models were able to closely match averaged production data from each of the three different regions analyzed. Figure 4.3 shows examples for production rate from the Barnett Shale, and Figure 4.4 displays cumulative production from southwest PA. These figures show data from age groups of wells with similar average well lengths, such that one parameter fit could be shown to match the data adequately (although the more recently drilled wells are visibly more productive due to increasing lateral lengths).

![Figure 4.3](image-url)

**Figure 4.3** – History-matched model fit of natural gas production rate (thousand standard cubic feet per day) versus time for an average Barnett Shale well (red line) compared with averaged production data from wells commencing production in 2008 (blue circle), 2009 (purple square) and 2010 (green diamond).
Figure 4.4 – History-matched model fit (blue line) of cumulative natural gas production (billion standard cubic feet) versus time compared with averaged production data for Marcellus Shale wells in southwest PA commencing production in Q1 2011 (orange circles), Q1 2012 (red squares) and Q2 2012 (green triangles).

Simulations of CO₂ injection were performed using the history-matched reservoir and well parameters for each of the three regions. Boundary conditions at the fracture face were an important consideration in the simulations. Two feasible injection scenarios were considered: injection of CO₂ at a constant maximum allowable pressure, and injection at a constant rate. The constant-pressure injection would maximize the mass sequestered by a single well for a given time, but injection rate declines strongly with time. Constant-rate injection would enable the matching of the injection rate to the steady output of CO₂ from an industrial emission source, with pressure at the injection well increasing with time, and eventually exceeding the maximum allowable pressure at the well. Both injection scenarios were simulated. It was assumed that the maximum allowable injection pressure would be selected to be as high as possible to maximize the mass that could be
sequestered in each well, while minimizing the risk of re-fracturing the rock. To meet these criteria, I chose the original natural gas reservoir pressure.

4.2.6 Model Limitations

While the CO₂ injection model is based on a simplified mathematical model consistent with production data from shale gas wells and parameters that have been selected by history-matching the model to production data, parametric and physical uncertainties remain due to the relative lack of understanding of shale formations. The largest uncertainties in the model are associated with adsorption: reliable adsorption data are scarce in the literature and, to my knowledge, no experimental data for CO₂ adsorption on samples of US shale gas formations at high (supercritical) pressure are available. The applicability of the adsorption model is therefore also uncertain. Although excess adsorption does not significantly increase total storage capacity and therefore any error would not fundamentally change the results for total capacity, we note that the absolute adsorbed mass is in fact a majority of the total stored CO₂. Plots showing the contributions of excess and absolute adsorbed CO₂ to total mass stored are included in the supporting information of Edwards et al. (2015).

The large amount of absolute CO₂ adsorption may possibly lead to substantial decrease in open pore volume and associated decrease in permeability, which would impact the dynamics of CO₂ injection. This phenomenon is known as ‘swelling’. Studies on the potential injection of CO₂ into coal bed methane reservoirs show that the organic matter in coal swells upon CO₂ adsorption and reservoir permeability consequently decreases significantly (Mazumder & Wolf, 2008; van Bergen et al., 2006). While shale gas
formations have much lower organic matter content than coals (2 – 10% compared with 75 – 90%) and associated lower adsorption, it has been shown that adsorption causes volumetric strain in some of the major mineral components of shale gas formations (Heller & Zoback, 2014). However, studies have yet to show whether swelling in the relatively small fraction of organic matter in shales yields a significant effect on overall rock permeability, so these effects were not included in my model. Also, while adsorption has the most uncertain parameters, selection of all parameters in the model would benefit from comprehensive data on the properties of shale gas formations: the parameter selections in the current model were guided by data from disparate sources.

4.3 Results and Discussion

Figure 4.5 shows the simulated constant-pressure injection rates of CO$_2$ into single, average horizontal shale gas wells in the Barnett Shale and the Marcellus Shale. Initially, injection rates decline rapidly, before displaying a more moderate decline rate at later times. These later-time injection rates are quite low compared to expected source rates for captured CO$_2$. For constant-rate injection simulations, pressure at the fracture face increases with time and eventually exceeds the maximum allowable pressure, with the time to exceedance related directly to the injection rate. An example plot of the injection pressure increase with time is included in the supporting information of Edwards et al. (2015). The simulated characteristic exceedance times for a range of injection rates are shown in Figure 4.6. Exceedance times decline considerably with increasing injection rate, and are less than 10 years for rates greater than 100 tonnes of CO$_2$ per day in all regions. Figure 4.7 shows cumulative CO$_2$ injected over time for constant-pressure injection in each of the regions. An associated plot showing the contributions of adsorbed
CO₂ to the total cumulative mass stored is in the supporting information of R. W. J. Edwards et al. (2015). Figures 4.5-4.7 show that there is a significant difference in injection rate and total mass able to be injected for average wells in each of the regions. Northeast PA has the highest capacity, mainly because the Marcellus Shale is significantly thicker in the northeast than the southwest. The Barnett Shale capacity is less than half that of southwest PA, which in turn is about half of the northeast PA capacity. The discrepancy between the Barnett and southwest PA is primarily due to horizontal laterals in the Barnett that are almost half the length of those in the Marcellus on average, rather than any major difference in formation thickness or other reservoir parameters.

**Figure 4.5** – Simulated constant-pressure CO₂ injection rate with time for average wells in the Marcellus Shale in northeast (green) and southwest (blue) PA at 35 MPa injection pressure, and in the Barnett Shale (red) at 25 MPa injection pressure. The inset graph shows rates at early time that are truncated in the main graph.
Figure 4.6 – Simulated time of constant-rate CO$_2$ injection before exceeding the maximum allowable pressure versus injection rate for average wells in the Marcellus Shale in northeast (green) and southwest (blue) PA for 35 MPa maximum pressure, and in the Barnett Shale (red) for 25 MPa maximum pressure.

Figure 4.7 – Simulated cumulative mass of CO$_2$ injected with time for constant-pressure injection in average wells in the Marcellus Shale in northeast (green) and southwest (blue) PA at 35 MPa injection pressure, and in the Barnett Shale (red) at 25 MPa injection pressure. The grey shaded areas represent the bound of upper and lower history-matched parameter scenarios based on changing well design with time and reservoir parameter uncertainty (See supporting information of Edwards et al. (2015) for detail of bound matching methods).
Although not visible in the regionally-averaged graphs, significant capacity variation is also observed within each play temporally (well laterals becoming longer with time), between companies (different well designs), and likely also spatially on smaller scales (due to geological differences). CO\textsubscript{2} injection sites could be targeted within each region at wells with higher capacity than the average (depending on proximity to emission sources and cost of transportation). However, for large-scale storage many wells would be required and therefore the regional averages are an important consideration. The average capacity of wells is likely to continue increasing over time, if average well lengths continue to increase as in the past. While the uncertainty ranges in Figure 4.7 make some allowance for this, the possibility remains that the averages could exceed these ranges at some future time. The Barnett Shale is a more mature play where drilling of new wells and overall production are declining, so less variation is expected over time.

4.3.1 Environmental Implications

The injection rates in average shale gas wells under constant-rate or constant-pressure scenarios (Figures 4.5 and 4.6) are relatively low compared with large industrial CO\textsubscript{2} sources. A typical 500 MW coal-fired power plant emits approximately 3 million metric tonnes (Mt) per year (MIT, 2007), or 8,200 tonnes per day, compared with rates of less than 100 tonnes per day for any significant duration in shale wells. The total capacities of shale wells are also relatively small: the capacity of an average well in northeast PA (the highest amongst the three regions analyzed) is approximately 0.6 Mt. Many shale wells would be required to sequester the emissions from a single coal-fired power plant. Considering the typical coal plant emitting 3 Mt per year and assuming a relevant timescale of storage to be 40 years (approximate plant lifetime), then 120 Mt are emitted,
requiring approximately 200 wells in northeast PA, 300 wells in southwest PA, or 700 wells in the Barnett Shale if all the emissions were captured and injected into shale wells. In comparison, conventional targets such as deep saline aquifers will have significantly larger capacity per well. For example, the Sleipner CO$_2$ injection in the North Sea has injected approximately 0.9 Mt per year (2,500 tonnes per day) into a single well in a very permeable saline aquifer for 19 years continuously, with over 15 Mt injected to date and only a small (0.2 MPa) increase in wellhead pressure observed (Chadwick et al., 2012). A CO$_2$ injection trial in the Mount Simon formation in Illinois injected 0.33 Mt per year (1,000 tonnes per day) into a single well for a period of three years (Finley, 2014).

Application of the average capacity estimates to the number of wells enables an estimation of the total CO$_2$ storage capacity of the analyzed regions. The number of wells in each region is dynamic, however, increasing quickly in the Marcellus Shale but more slowly in the Barnett Shale. Two estimates have therefore been made for each play: a current capacity using all existing horizontal wells, and a future projection using all existing and permitted (approved but not yet drilled) wells. The limitations for the future projection are that average well parameters will likely change in time and more wells will continue to be drilled following those already permitted, particularly in the Marcellus Shale. The capacity estimates are shown in Table 1. The capacity of all existing and permitted Marcellus Shale wells in PA is estimated to be 7.2 – 9.6 Gt. Given likely average well production lifetime of 20 – 40 years (Browning et al., 2013; Cipolla et al., 2010), this capacity would not be available for CO$_2$ sequestration for some time into the future. As a comparison, Godec et al. (2013a) estimated total theoretical CO$_2$ storage capacity for the Marcellus (in all states) to be 171 Gt (104 Gt in PA), and total technically
accessible capacity to be 49 Gt. For the technically accessible estimate to be consistent with my results, approximately 100,000 wells would be required (assuming well and reservoir parameters from PA are applicable to Marcellus wells in adjacent states). At current rates of drilling in the Marcellus, it will be approximately 50 more years until 100,000 wells have been drilled, with an additional several decades of production before all those wells would be available for CO₂ injection. Another study by Tao and Clarens (2013) estimated that 10.4 – 18.4 Gt of CO₂ could be injected in the Marcellus by 2030. Based on my results this would require approximately 22,000 – 39,000 wells to be depleted, available for CO₂ injection and filled by 2030. With approximately 8,000 wells drilled and producing in the Marcellus to date and considering the time scales for production and injection, this is an unlikely prospect.

Table 4.1 – Model input parameters for simulation of water imbibition into shale.

<table>
<thead>
<tr>
<th>Shale play region</th>
<th>Barnett Shale</th>
<th>Marcellus southwest PA</th>
<th>Marcellus northeast PA</th>
<th>Marcellus PA total</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Existing wells</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>capacity (Gt)</td>
<td>1.9 – 2.9</td>
<td>0.5 – 0.7</td>
<td>1.3 – 1.8</td>
<td>1.9 – 2.6</td>
</tr>
<tr>
<td>(number of wells*)</td>
<td>(15,729)</td>
<td>(1,731)</td>
<td>(2,801)</td>
<td>(4,845)</td>
</tr>
<tr>
<td><strong>Existing and</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>permitted wells</td>
<td>2.1 – 3.1</td>
<td>1.6 – 2.2</td>
<td>5.0 – 6.6</td>
<td>7.2 – 9.6</td>
</tr>
<tr>
<td>capacity (Gt)</td>
<td>(16,663)</td>
<td>(5,623)</td>
<td>(10,449)</td>
<td>(18,273)</td>
</tr>
<tr>
<td>(number of wells*)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*as of March, 2015.

In summary, a model of gas flow to a hydraulically fractured horizontal well incorporating the most important physical processes for CO₂ injection has been developed. Application of the model to the Barnett and Marcellus Shales shows that individual shale wells have small capacities and low injection rates compared with CO₂ emissions from large industrial sources. Large-scale storage would require many shale
wells. Although prior studies showed large overall capacities for CO₂ storage in shale formations, drilled and currently permitted wells provide access to only a fraction of this capacity and are unlikely to be available for CO₂ storage for some decades into the future. This study builds understanding of the engineering requirements and capacity availability for CO₂ storage in depleted shale formations and will help inform assessment of the relative cost effectiveness of storage in shale formations compared to conventional targets such as saline aquifers.
Chapter 5

Infrastructure to Enable Carbon Capture and Storage Deployment

This chapter is a modified version of a manuscript that is currently under review for publication in the journal *Proceedings of the National Academy of Sciences* (Edwards & Celia, 2018a). The manuscript was modified to change the writing from the plural first person (we) to singular first person (I). Supporting information for this chapter can be found in the supporting information of Edwards and Celia (2018a).

5.1 Introduction

Despite its importance, carbon capture and utilization, and storage (CCUS) deployment is lagging far behind estimates of what is required to meet the Paris target (Peters et al., 2017). Only about 31 million metric tonnes (Mt) per year of anthropogenic carbon dioxide are currently captured and injected into geological formations for permanent storage (GCCSI, 2017), while analyses estimate that 200-1,000 Mt per year are required by 2030, and 5,000-10,000 Mt per year by 2050 (IEA, 2017b; Mac Dowell et al., 2017; Rogelj et al., 2016; Shell, 2018). CCUS has been held back by inconsistent and insufficient policy support, a lack of economic drivers, and the inherent large scale and associated large cost of individual projects (IEA, 2016).
After years of relatively little policy support, in February 2018 the United States Congress passed substantial tax credits that incentivize new CCUS projects (Future Act, 2017). From 2018 to 2026, the Section 45Q tax credit value will increase linearly from $25.70 to $50 per metric tonne of carbon dioxide for secure geological storage, and from $15.30 to $35 per tonne used in carbon dioxide enhanced oil recovery (CO₂-EOR) that results in secure geological storage (or other uses that permanently store carbon dioxide). The tax credit value will increase at the rate of inflation after 2026. CO₂-EOR operations typically pay an oil-linked price near 40% of the per-barrel oil price for a tonne of carbon dioxide ($23 per tonne at the April 2018 oil price around $60 per barrel), which adds value for the case where captured carbon dioxide is used for EOR (Hill et al., 2013; Kuuskraa et al., 2011). Capture projects must begin construction by January 1, 2024 to receive the credits, and once in service will receive those credits for a 12-year period.

The tax credits will likely be insufficient to incentivize widespread carbon capture retrofits on electricity generation plants considering the current relatively high estimated capture costs around $50 and $75 per tonne of carbon dioxide for coal and gas plants, respectively (NETL, 2015b; Rubin et al., 2015). However, they will provide a strong incentive for lower-capture-cost opportunities, which are typically industrial sources with relatively concentrated carbon dioxide waste streams with capture costs in the range of $10-$55 per tonne (IEAGHG, 2016, 2017; Leeson et al., 2017; McKaskle, 2016; NETL, 2014b; Shell, 2017). Given our daunting climate targets and the need to rapidly scale up CCUS, these low-capture-cost sources represent an attractive pathway for near-term deployment. Deploying CCUS on these sources will not only reduce emissions, but also give an opportunity for additional learning, cost reductions, and the construction of
transport infrastructure that will help enable and accelerate future CCUS projects. With this as motivation, I investigate the following questions: Can the tax credits provide sufficient support to enable construction of large-scale carbon dioxide capture and transportation infrastructure? What additional policy support might be needed? What other challenges need to be addressed? In order to answer these questions, I consider the lowest-capture-cost carbon dioxide sources in the United States, the pipeline infrastructure needed to transport that carbon dioxide to where it can be utilized and stored, and whether the tax credits provide sufficient value to make the system economically viable.

Figure 5.1 shows the location and size of low-capture-cost sources in the United States. I include only source types for which there are already commercially implemented capture technologies and existing large-scale carbon capture projects and, therefore, for which carbon capture could be deployed within the next several years. The sources include natural gas processing, ethanol fermentation at biorefineries, and hydrogen and ammonia production, which account for a total of 87 Mt per year of emissions with low capture cost. More information on the emissions data and estimation is included in the supporting information of Edwards and Celia (2018). The map also shows the location of deep saline aquifers with potential for geological carbon storage and existing carbon dioxide pipelines that serve the CO₂-EOR industry.
Figure 5.1 – Low capture-cost carbon dioxide emissions in the United States, existing carbon dioxide pipelines, and potential saline storage formations. Co-located sources are summed so that the total emissions are observable. Total emissions are 87 Mt per year, including 43 Mt from ethanol fermentation at biorefineries, 22 Mt from hydrogen production, 5 Mt from ammonia production, and 17 Mt from natural gas processing. Data for the figure were obtained from multiple sources (EIA, 2018b; EPA, 2017; NEO, 2018; NETL, 2015a; RFA, 2018a, 2018b).

Carbon dioxide emissions from ethanol fermentation in the Midwest stand out in Figure 5.1 as the largest combined source (50% of the total and 43 Mt per year). They also stand out because they are not located near existing carbon dioxide pipelines, and they mostly do not overlie potential saline storage reservoirs. In order to capture a substantial proportion of carbon dioxide emissions from ethanol production, a regional pipeline network would be needed to aggregate emissions from many ethanol biorefineries and transport that carbon dioxide to storage locations. Such a network may be economically attractive since ethanol fermentation is a particularly low-cost capture opportunity: ethanol fermentation generates a gas outlet stream that is more than 99% carbon dioxide.
(once moisture is removed) and thus requires only compression and dehydration (Gollakota & McDonald, 2012; SWG, 2017c; NETL, 2014a). Correspondingly, carbon capture on ethanol biorefineries is already commercially deployed. There are approximately 210 ethanol biorefineries in the United States, of which about 40 already capture at least some carbon dioxide for sale to the EOR, food and beverage, and dry ice industries (EPA, 2018a; SWG, 2017c; NEO, 2018; RFA, 2018a, 2018b). At the largest scale, Archer Daniels Midland’s (ADM) Decatur ethanol biorefinery captures nearly 1 Mt per year and injects it into a saline aquifer in a government-funded demonstration project (Finley, 2014; Gollakota & McDonald, 2012, 2014; NETL, 2014a). Therefore, I specifically focus my study on the feasibility of capturing emissions from the Midwest region, with a focus on ethanol biorefineries, and developing a pipeline network to transport the carbon dioxide. Other regions tend to have either fewer low-capture-cost sources or existing carbon dioxide pipelines and, consequently, are not considered in my analysis.

The carbon dioxide pipelines in Figure 5.1 supply carbon dioxide from natural and anthropogenic sources to oil fields for CO₂-EOR (Wallace et al., 2015), an activity that involves the injection of carbon dioxide into depleted oil reservoirs to induce additional production (Godec et al., 2013b; Group, 2017c). Carbon dioxide injected for EOR is ultimately securely stored in the oil reservoirs (Hill et al., 2013; IEA, 2015). About 63 Mt per year is currently injected for CO₂-EOR in the United States, of which around 78% is sourced from natural underground reservoirs and 22% from anthropogenic sources (EPA, 2018a; Hill et al., 2013; Wallace et al., 2015). CO₂-EOR drives most existing CCUS: of the 31 Mt of anthropogenic carbon dioxide emissions currently captured and stored
globally each year, 90% is for CO$_2$-EOR, mainly in the United States (GCCSI, 2017). Just 1 Mt is injected for dedicated geological storage in the United States each year (the ADM project). About 4% of domestic oil production is through CO$_2$-EOR (Middleton et al., 2015). Around 65% of existing CO$_2$-EOR occurs in the Permian Basin of West Texas, 15% in the Rocky Mountains region, mainly in Wyoming, 13% along the Gulf Coast, and about 5% in the Mid-Continent, mainly in Oklahoma (EPA, 2018a; Godec et al., 2013b). This distribution correlates closely with the locations of carbon dioxide pipelines in Figure 5.1. The size of the CO$_2$-EOR industry is limited by lack of affordable carbon dioxide supply rather than a lack of potential (Godec, 2014; Gray & Goodyear, 2014; Hill et al., 2013; IEA, 2015): oil reservoirs in the United States could store enough carbon dioxide to meet projected carbon storage requirements under a two-degree pathway until at least mid-century (Godec et al., 2013b; Kuuskraa et al., 2011; Mac Dowell et al., 2017).

I therefore specifically target my study on transporting captured carbon dioxide to regions with demand for EOR. I focus on storage through CO$_2$-EOR rather than dedicated storage for a number of reasons:

1) The United States has an established CO$_2$-EOR industry with large potential for expansion (Godec et al., 2013; SWG, 2016). CO$_2$-EOR projects can likely be developed more quickly than dedicated storage projects, which face more stringent regulations and for which there is little experience (EPA, 2010; IEA, 2018).
2) The pipeline infrastructure would also be a long-term asset for dedicated carbon storage, crossing several prospective saline aquifer storage formations (see Figures 5.1 and 5.2).

3) The use of captured carbon dioxide for EOR is likely to be the most economically favorable option. Given the location of most ethanol biorefineries, a regional pipeline network would need to be developed in either case so transport cost would be similar for EOR or dedicated storage. Once the carbon dioxide has been transported, there would be additional cost for dedicated storage; by contrast, additional revenue is earned when the carbon dioxide is sold for CO₂-EOR. This differential is likely to exceed the $15 per tonne tax credit differential: dedicated storage costs are typically about $10 per tonne (Rubin et al., 2015; Shell, 2017), while sales revenue is typically around $20 per tonne (at the April 2018 oil price), a $30 per tonne differential. Carbon dioxide sales for CO₂-EOR also deliver cash revenue, which may improve the financial viability of the EOR option compared with dedicated storage projects that would only receive tax credits that must be monetized by a project owner. However, additional risk is introduced through exposure to volatile oil prices.

4) Major carbon dioxide capture and pipeline infrastructure projects based on CO₂-EOR are likely to be more broadly and strongly supported because they also benefit the oil and gas industry and oil-producing states. Indeed, it was a coalition including oil-state Republicans and climate-focused Democrats that enabled the passing of the increased tax credits (Future Act, 2017).
5) Tax revenue to federal and state governments due to additional oil production from CO2-EOR substantially covers tax revenue forgone by the tax credits. Governments receive about $10 in tax per barrel of oil produced, or $20-$30 per tonne of carbon dioxide used in EOR (Kuuskraa et al., 2013). Use of carbon dioxide for CO2-EOR may therefore give a fiscal argument for policies that support CCUS.

A concern with the use of captured carbon dioxide for CO2-EOR is that associated oil production could offset the emissions benefit of storage. However, a number of studies have found that CO2-EOR does result in a significant net emissions reduction on a lifecycle basis. Injection of carbon dioxide for CO2-EOR results in its permanent storage in the oil reservoir, with minimal carbon dioxide lost in the process (Hill et al., 2013; IEA, 2015). Each tonne of carbon dioxide injected yields 2-3 barrels of oil production (with variability between different oil fields) (Azzolina et al., 2016; Cooney et al., 2015; Dai et al., 2016; Godec et al., 2013b; IEA, 2015), which result in around 1.2 tonnes of carbon dioxide when used (IEA, 2015). These are not net additional emissions, however, since each barrel of oil produced by CO2-EOR mostly displaces other oil in the market. The International Energy Agency (IEA) estimates that 80% of each barrel displaces other oil production, and that, after accounting for all factors including the amount of oil produced, energy use in CO2-EOR operations, and the market impacts, each tonne of carbon dioxide injected for EOR results in 0.63-0.73 tonnes of carbon dioxide emissions abatement on average (IEA, 2015). Precise calculation of the net emissions benefit of each CO2-EOR project is complex and depends on the properties of the oil reservoir, the EOR process used, and market factors, including the type of oil displaced. Still, it is clear that a significant net emissions reduction results from CO2-EOR.
My analysis uses existing carbon dioxide capture and pipeline cost models and data to estimate costs. I evaluate the economic feasibility of the system considering the value of the tax credits and carbon dioxide sales for EOR. I consider four options for the basic pipeline network structure, shown in Figure 5.2, which represent two options for collecting carbon dioxide in the Midwest and two options for delivery of that carbon dioxide. The collector trunk pipeline options include a southern trunk pipeline that follows the shortest path between the largest ethanol biorefineries, or a northern collector trunk that passes through the center of ethanol biorefinery regional density in the Midwest. The delivery options include trunks delivering the carbon dioxide to Wyoming or to the Permian Basin, the two regions with the largest demand for CO$_2$-EOR. The pipeline pathways are restricted to existing major infrastructure corridors. After determining the best trunk pipeline combination, I evaluate three different financing scenarios for the pipeline network, representing different combinations of commercial and government financing. I ultimately estimate the total quantity of carbon dioxide that is economically viable to capture with the tax credits under different pipeline financing scenarios.
Figure 5.2 – Potential carbon dioxide trunk pipelines that were analyzed, including a northern collector trunk through the center of biorefinery regional density, a southern collector trunk linking the largest biorefineries, and delivery trunks to Wyoming or the Permian Basin. Data for the figure were obtained from multiple sources (EIA, 2018b; EPA, 2017; NEO, 2018; NETL, 2015a; RFA, 2018a, 2018b).

5.2 Methods

5.2.1 Emissions Data and Estimation

Carbon dioxide emissions for natural gas processing, hydrogen, and ammonia sources were obtained from EPA Greenhouse Gas Reporting Protocol data for 2016 (EPA, 2017). I estimated the emissions from the low-capture-cost syngas process stream of hydrogen and ammonia production from reported total production emissions. These emissions typically account for 50% of total production emissions (IEAGHG, 2017; Meerman et al., 2012; Soltani et al., 2014). I assumed that all of reported natural gas production emissions
are able to be captured at low cost. Ethanol biorefinery fermentation emissions are not reported to the EPA, so I used the stoichiometry of ethanol fermentation to estimate emissions based on biorefinery facility production capacity and reported ethanol production data available from the Renewable Fuels Association and Nebraska Energy Office (NEO, 2018; RFA, 2018a, 2018b; RTI, 2010). More detailed information on emissions estimation from each source type is provided in the supporting information of Edwards and Celia (2018a).

5.2.2 Pipeline and Capture Cost Estimation

Pipeline costs were estimated using the United States Department of Energy National Energy Technology Laboratory (NETL) CO$_2$ Transport Cost Model (NETL, 2018a, 2018b). I selected options within the model that produced the closest results compared to published costs from two major existing carbon dioxide pipelines and industry rule-of-thumb costs. The model generally over-estimated costs compared with the published project costs and gave costs approximately 15% above the industry rule-of-thumb averaged across the network. I implemented a Microsoft Excel macro to automate the repetitive calculation of costs for each pipeline segment in the network. I used a modified version of a macro originally developed by Dubois et al. (2017).

Hydrogen and ammonia capture costs were based on published cost data from the Shell Quest hydrogen production carbon capture project, supported by cost information from the Air Products Port Arthur project and hydrogen capture cost literature (IEAGHG, 2017; Meerman et al., 2012; NETL, 2017; Shell, 2017). Ethanol biorefinery capture costs were estimated using a model developed by the State CO$_2$-EOR Deployment Workgroup.
based on project data and input from people with direct project experience (SWG, 2017c). I modified the model to slightly increase operational costs. The model was compared with one publicly available project cost example: modeled capital cost was approximately 25% greater than the reported cost of the first stage of the ADM Decatur ethanol capture project (McKaskle, 2016).

5.2.3 Financial Analysis

I performed a discounted cash flow analysis of project costs and revenues for both the pipelines and capture facilities. For the pipelines, I used the discounted cash flow analysis in the NETL CO₂ Transport Cost Model (NETL, 2018a, 2018b). I modified the analysis to reflect the new corporate tax rate and interest tax deduction rules introduced by the Tax Cuts and Jobs Act of 2017. I added an explicit debt schedule to separate consideration of debt and equity, rather than using the default weighted average cost of capital (WACC) discount rate method. The key differing assumptions for the three financing scenarios are included in Table 1. The debt financing period was assumed to be 12 years for the full commercial case, in line with the duration of the tax credits. For the commercial-government and full government cases I assumed that government bears the post-tax credit policy risk and provided 20-year debt finance. The debt interest rate for the commercial-government case was assumed to be 1% above the 20-year United States Treasury bond interest rate (modeled on the Department of Energy Loan Program interest rate). For the full government case, I assumed the project is 100% financed by government debt at the 20-year bond interest rate. I used the maximum 20-year bond interest rate in the past 5 years. For each pipeline financing scenario, I apply the same financing parameters to all pipelines in the network.
Table 5.1 – Key financial parameter assumptions for each pipeline financing scenario.

<table>
<thead>
<tr>
<th>Financing scenario</th>
<th>Debt and equity percentage (%)</th>
<th>Debt interest rate (%)</th>
<th>Debt financing period (years)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Full commercial</td>
<td>50:50</td>
<td>6</td>
<td>12</td>
</tr>
<tr>
<td>Commercial-government</td>
<td>50:50</td>
<td>4.5</td>
<td>20</td>
</tr>
<tr>
<td>Full government</td>
<td>100:0</td>
<td>3.5</td>
<td>20</td>
</tr>
</tbody>
</table>

The capture facility financial analysis assumed 100% equity financing of the projects, since they are cash flow negative and could not make debt repayments. The projects rely on the tax credits for their positive value. I applied the legislated schedule for the value of the tax credits in each year (Future Act, 2017). I assumed the project owners could fully monetize the value of the tax credits. The target rate of return for the capture facilities was 15%, but a 10% target rate of return was used for marginal facilities after the first network iteration cycle. One exception in the financial analysis was consideration of ADM’s large Decatur ethanol biorefinery: a capture facility already exists, so this facility may be ineligible for the new tax credits (depending on whether it claimed the previous Section 45Q tax credits, which is not public information). I therefore assumed no new tax credits were received by this facility, which resulted in its financial unviability.

All pipeline and hydrogen capture projects were assumed to begin in 2020 with a 4-year capital expenditure period. Ethanol capture projects were assumed to begin in 2022 with a 2-year capital expenditure period. All projects become operational in 2024. I assumed 2% inflation for all costs and the carbon dioxide sales price. All costs are reported in United States 2018 dollars. Further information on the financial analysis, a full list of
assumptions, and the financial analysis model spreadsheets are all included in the supporting information of Edwards and Celia (2018a).

5.2.4 Network Analysis

I initially considered all of the low-capture-cost sources in the study area with emissions exceeding the 100,000 tonnes per year minimum for tax credit eligibility, shown in Figure 5.3. The first step in the network analysis was to analyze the trunk options, shown in Figure 5.2, as described in the Results section. After determining the best trunk route option, a pipeline network collecting carbon dioxide from all sources was designed; the full initial network is shown in Figure 5.3. All network design was performed with Esri ArcGIS. I limited potential pipeline routes to existing natural gas, ammonia, and carbon dioxide pipelines, as well as railways, interstate highways, and electricity transmission lines greater than 220 kV (large transmission lines). The GIS source data are listed in the supporting information of Edwards and Celia (2018a). I used the ArcGIS Network Analyst feature to find the shortest routes from each source to the Permian Basin destination, with manual constraints employed so that sources would aggregate efficiently and follow the trunk pipeline routes. The detailed ArcGIS methodology is included in the supporting information of Edwards and Celia (2018a).
Figure 5.3 – The full initial network and sources considered in the analysis. The total capture potential is 37 Mt per year. The map also shows the potential pipeline corridors considered, which included existing natural gas, ammonia, and carbon dioxide pipelines, as well as railways, interstate highways, and high voltage electricity transmission lines. The underlying map data were obtained from multiple sources (Natural Earth, 2018; EIA, 2018b; EPA, 2017; HIFLD, 2017; NEO, 2018; ORNL, 2009; RFA, 2018a, 2018b).

After determining the initial network design, I performed the financial analysis and network iteration process to find the stable, economically viable networks for each financing scenario. The process was as follows:

1. Determine the carbon dioxide flow rate for each segment of the pipeline network.
2. Calculate the pipeline size, costs, and required carbon dioxide transport tariffs using the modified NETL CO₂ Transport Cost Model (Dubois et al., 2017; NETL, 2018a, 2018b). Calculate the tariff that each source must pay based on the pipeline segments it uses.
3. Calculate the required carbon dioxide sales price for each source using the capture facility financial analysis model, with the pipeline tariff input from the previous step.

4. Eliminate all sources with a required carbon dioxide sales price above the $23 per tonne threshold. A threshold requiring 15% rate of return at $23 per tonne was used in the first iteration. Subsequent iterations allowed a 10% rate of return for marginal facilities, as explained in the Results section.

5. Update the pipeline network design (if required).

6. Repeat steps 1 to 5 until a stable system is found with all sources economically viable.

More detail on the network analysis methodology is included in the supporting information of Edwards and Celia (2018a). The network analysis was performed separately for each pipeline financing scenario. The required carbon dioxide sales price curves for each iteration of each financing scenario are shown in Figure 5.4.
5.3 Results and Discussion

The Permian Basin and Wyoming trunk options have very similar pipeline costs. The Permian Basin option requires a 5% higher total cost for a pipeline network to transport emissions from all low-capture-cost sources in the Midwest ethanol-producing region, due to the Permian Basin trunk being approximately 1,000 km compared with the 800 km Wyoming trunk. However, the Permian Basin has substantially greater existing and potential future demand for carbon dioxide. Approximately 40 Mt is currently injected in the Permian Basin each year compared with around 10 Mt in Wyoming (EPA, 2018a; Godec, 2014), and the Permian Basin is estimated to have at least 2,300 Mt of potential
future CO₂-EOR demand compared with 600 Mt for Wyoming (Kuuskraa et al., 2011). The Permian Basin trunk also passes through Kansas and Oklahoma, which are estimated to have at least a further 1,800 Mt of potential CO₂-EOR demand (Kuuskraa et al., 2011). Considering that the full potential Midwest low-capture-cost source network could supply 37 Mt per year, Wyoming has only 16 years of capacity (assuming CO₂-EOR projects could be scaled up quickly enough to absorb the new supply). I therefore selected the Permian Basin trunk as the better option since the additional cost is small but the potential to absorb the new carbon dioxide supply is much greater.

After selecting the Permian Basin trunk, I compared the northern and southern collector trunk options. I investigated the economic viability of capturing and transporting carbon dioxide from all low-capture-cost sources in the Midwest region for both options. I calculated the cost of each segment in the pipeline networks and the tariff the pipeline owners would need to charge for each tonne of carbon dioxide transported in order to achieve a 12% rate of return over the pipeline financial lifetime. The pipeline transport tariff that each source must pay was then calculated based on the pipeline network segments it uses. Capture facility cost for each source was calculated. Finally, I determined the carbon dioxide sales price that would be required in addition to the value of the tax credits for the capture facility owners to achieve a 15% rate of return over the capture project financial lifetime, considering the capture and transport costs for each source. (The sales price is for carbon dioxide delivered to the destination; the capture facility pays the transport cost.) The capture facilities have a higher target rate of return than the pipelines since they are less established industries, with revenue more closely tied to oil prices and, therefore, have higher risk.
After determining the required price for each source, I eliminated all sources whose required price was above a threshold set at the typical CO$_2$-EOR price of $23 per tonne of carbon dioxide (for the April 2018 oil price around $60 per barrel) (Kuuskraa et al., 2011). Only ethanol biorefinery sources remained due to their lower capture cost than the other source types. The removal of sources from the network reduces the number of sources sharing the pipelines and, therefore, the pipeline flow rates. Pipelines have strong economies of scale, so lower flow rates imply that the cost per unit of flow increases and pipeline transport tariffs increase for the remaining sources. The pipeline tariffs and required carbon dioxide sales prices were therefore re-calculated for each remaining source. Emissions abatement curves are often presented as static curves of quantity abated versus unit cost, but for our CCUS network system the cost of each unit of emissions abatement is dependent on all other units (sources) in the network: this is a dynamic system that cannot be represented by a static curve (Kesicki & Ekins, 2012; Levihn et al., 2014).

For the initial networks, the northern trunk option has 17.8 Mt of captured carbon dioxide below the price threshold compared to 16 Mt for the southern trunk. After elimination of the sources above the threshold and re-calculation of tariffs and prices (called “iteration” hereon), the northern trunk option has 9.5 Mt below the price threshold compared to 7.1 Mt for the southern trunk. I therefore selected the northern trunk as the better option. More detailed description of the analysis procedure is in the Methodology, and more information on the northern versus southern trunk comparison is in the supporting information of Edwards and Celia (2018a).
While the northern trunk option enables a cheaper overall system than the southern trunk, continued iteration of the network does not find a stable, viable system. The quantity of emissions below the price threshold is drastically reduced from the initial potential of 37 Mt to 9.5 Mt after the first iteration of the network. After a second iteration, just 3.4 Mt are below the price threshold (see Figure 5.4 in the Methodology). A third iteration would reduce the total close to zero. The capture of emissions from ethanol biorefineries in the Midwest and transport to the Permian Basin is not viable under my commercial financing scenario parameters. Smaller systems that capture emissions from some of the largest ethanol biorefineries and transport carbon dioxide to nearer CO₂-EOR opportunities in Kansas or Illinois may be viable (Dubois et al., 2017; SWG, 2017c), but the objective of this study is to explore the largest-scale capture and transport infrastructure that can be developed. I therefore consider two alternative pipeline financing scenarios. The first is a commercial project with half of the capital cost financed by longer-term government loans with lower interest rate than commercial debt (4.5% compared with 6%), termed the “commercial-government” scenario. The second is for the project to be fully financed by government debt (at 3.5% interest rate), termed the “full government” scenario. There are numerous possible financing arrangements, as discussed in reports by the State CO₂-EOR Deployment Workgroup (SWG, 2017a, 2017b), but the three scenarios represent both ends of the spectrum and an intermediate option. More detailed information on the financing scenarios is included in the Methodology.

The three different financing scenarios only apply to the pipeline network. Capture facilities have the same financial parameters in each scenario for several reasons. Firstly, pipelines are more capital intensive than capture facilities for ethanol biorefineries, so
greater system cost reductions are possible through cheaper finance for pipelines (see Figure 5.7). For the pipelines, around 80% of the total cost is capital and 20% operational costs; for the capture facilities, around 50% is capital and 50% operational costs. The capital cost of the pipeline network is also over double the summed capital cost of all ethanol biorefinery capture facilities in the network. Secondly, the capture facilities will receive financial support through the tax credits, so additional direct support may be less likely. Thirdly, there is a stronger precedent for direct government financing of shared infrastructure like pipeline networks.

The network analyses for the commercial-government and full government pipeline financing scenarios yielded stable systems with all connected capture facilities economically viable. Both stable networks are shown in Figure 5.5. The full government pipeline scenario network captures and transports a total of 28.7 Mt of carbon dioxide per year from 108 ethanol biorefinery sources, compared with 19.0 Mt from 63 ethanol sources for the commercial-government scenario. The capital cost of the system is estimated to be $6.7 billion for the pipeline network and $2.6 billion for the capture facilities in the stable full government scenario, compared with $4.3 billion for the pipelines and $1.6 billion for the capture facilities in the stable commercial-government scenario.

Either case would represent a significant increase in global CCUS and CO₂-EOR. For context, the full government scenario would approximately double the global total amount of anthropogenic CCUS (GCCSI, 2017). It would abate carbon dioxide emissions equivalent to roughly 11 gigawatts (GW) of wind electricity generation capacity in the United States (half the total installed wind capacity in Texas), or 14 GW of solar
photovoltaic capacity (two-thirds of the total installed solar capacity in California).
Carbon dioxide supply to the CO$_2$-EOR industry would increase by approximately 45%, which would drive around 200,000 barrels per day of oil production, equal to about 2% of current domestic production. The 1,900 km long main trunk pipeline would be more than double the length of the Cortez pipeline from Colorado to the Permian Basin, currently the world’s largest carbon dioxide pipeline, and would have equivalent diameter and flow capacity through the 1,000 km main transport trunk section (Wallace et al., 2015). The 7,000 km total pipeline network would nearly double the existing total length of carbon dioxide pipelines in the United States (Wallace et al., 2015). Such an expansion, if initially built with potential for increased capacity, would also help enable and accelerate future CCUS projects in regions near the pipeline.
Figure 5.5 – Carbon dioxide pipeline networks for the two pipeline financing scenarios with stable, viable systems. Top: commercial-government case, capturing and transporting a total of 19 Mt of carbon dioxide per year. Bottom: full government case, capturing and transporting 28.7 Mt per year. The diameter of each segment of the pipeline network is shown. Co-located sources are summed so that
the total emissions are observable. The underlying map data were obtained from multiple sources (NEO, 2018; RFA, 2018a, 2018b).

Figure 5.6 shows the carbon dioxide sales prices required for each capture facility to achieve the target 15% rate of return for the stable networks of both pipeline financing scenarios. (The price curves for all network iterations are shown in Figure 5.4.) Most of the sources in each scenario have required prices well below the threshold. There are also a number of marginal sources, whose required price for a 15% rate of return is above the threshold. In the latter iteration cycles, the target rate of return was 10% for these marginal sources. There are 11 marginal sources accounting for 1.6 Mt in the full government scenario, and 13 marginal sources accounting for 2.8 Mt in the commercial-government scenario. For these marginal sources, Figure 5.6 also shows their required price to achieve a 10% rate of return. All marginal sources in the full government network have a required price below the threshold for a 10% rate of return, while 4 marginal sources in the commercial-government scenario have a required price less than $1 above the threshold. The marginal sources are important for the viability of the systems because pipelines have a strong economy of scale: pipeline cost scales approximately linearly with pipe diameter, but flow capacity is determined by the pipe cross-sectional area and scales with diameter squared (SWG, 2017b; NETL, 2014c). Pipelines with greater flow are cheaper per unit of flow, decreasing costs for all sources sharing the pipelines.
Figure 5.6 – Required carbon dioxide sales price curves for both pipeline financing scenarios with stable systems. Each curve shows cumulative carbon dioxide captured and transported against the required sales price for each facility. The solid lines show the required prices for each source capture facility to achieve a 15% rate of return. The dashed lines show the required price for the marginal sources (those with required price for 15% rate of return above the $23 per tonne threshold) to achieve a 10% rate of return.

Levelized costs and revenues (the net present value per tonne of carbon dioxide captured and transported) for the different financing scenarios are shown in Figure 5.7. Decreasing pipeline financing costs has a strong effect on decreasing the total system cost, owing to the greater capital intensity and total capital cost of the pipelines, while a much smaller proportional reduction can be achieved by reducing ethanol biorefinery capture facility financing costs. The average levelized cost for each tonne in the full commercial case is greater than the levelized revenue, indicating its infeasibility, while the commercial-government and full government funding scenarios have lower average cost than revenue.
Figure 5.7 – Average levelized costs and revenues per tonne of carbon dioxide captured and transported for each network financing scenario, in 2018 dollars. Costs are shown by the bars and are the system-wide average. The revenue values are shown by dashed lines and are stacked in the graph: the tax offset value is $3.69, tax credit value is $26.97, and the carbon dioxide sales revenue is $23. The tax credit and offset are effective revenues since they are reduced tax liability rather than cash revenue. The tax offset is the value owners gain by using capture facility net operating losses and asset depreciation to offset other taxable income. I assume that the value of the tax credits and offsets can be fully monetized by project owners. Further detail is included in the supporting information of Edwards and Celia (2018a).

5.4 Policy Challenges and Implications

I demonstrate an opportunity to significantly expand CCUS in the United States in the near-term, spurred by the new tax credits, by targeting the lowest-cost capture opportunities and by deploying only commercially proven technologies. The pipeline network would deliver carbon dioxide to the regions of greatest demand for CO₂-EOR.
and also connect multiple prospective long-term dedicated carbon storage resources. This would be a long-term and flexible infrastructure asset for carbon management in the United States. There are, however, a number of significant challenges to building such a CCUS network.

A key challenge to the feasibility of the CCUS networks presented here is their need for substantial government financing. While the cost estimates in this study are scoping-level, and detailed engineering design and costs are needed to more precisely determine the viability of specific financing cases, it is clear that low-cost government financing of pipeline infrastructure would significantly reduce the required pipeline tariffs and increase the amount of economically viable capture opportunities. Government financing of carbon dioxide pipeline networks has not been prominently considered in public discussion in the United States. However, it could be the best option for initial projects if we hope to scale up CCUS as needed to achieve stated climate targets. Governments have often financed similar shared infrastructure with a public good aspect and economies of scale that are natural monopolies, such as highways, water and sewer pipelines, and telecommunications and electricity networks (Chan et al., 2009; Gramlich, 1994). Governments could build carbon dioxide pipeline networks and privatize them when the CCUS industry is mature. A relevant example is the 2,000 km long Big Inch and Little Inch oil pipelines from Texas to New England, which the United States government financed and owned in the 1940s during the Second World War. The pipelines were later sold to the private sector and continue to be in service today (SWG, 2017b).

Under any financing scenario, the timeline for building the network is formidable since all capture facilities must begin construction before January 1, 2024 to be eligible for the
tax credits. Therefore, the pipeline network would need to be constructed around that
time to transport captured carbon dioxide, as well as CO₂-EOR projects to use the new
supply. Planning, designing, permitting, and constructing the 2,000 km main trunk and
5,000 km of feeder pipelines of the full government scenario network within this
timeframe will be challenging. However, a comparison with recent natural gas pipeline
development in the United States suggests it is possible: an average of 1,500 km of new
major interstate natural gas pipelines have been completed each year for the past decade,
with a maximum of 4,400 km completed in a single year (EIA, 2018c; FERC, 2018).
Individual major pipeline projects greater than 200 km length have taken 2.2 years on
average from permit application filing date to construction completion (a process that
begins only after the route design is completed and right-of-ways have been negotiated,
which is itself a lengthy process) (EIA, 2018c; FERC, 2018). The tax credit timelines are
legislatively defined and could be changed—similar wind and solar tax credits have been
extended—but this possibility cannot be planned for.

The CCUS network development would require close coordination between the ethanol
and oil industries and state and federal governments, regardless of timing. A lack of
coordination would leave a chicken-and-egg situation where potential capture projects are
uncertain of demand and the availability of a pipeline network, while pipeline builders
and CO₂-EOR projects are uncertain of how much and when carbon dioxide will be
available. Without a coordinated effort, we will likely see smaller-scale and more local
projects that capture carbon dioxide from some of the larger ethanol biorefineries and
transport it to nearer CO₂-EOR or dedicated storage opportunities. Such an outcome
would severely erode the viability of a large-scale network and reduce the total emissions
that can be captured, since the economy of scale from combining all sources and sharing pipeline infrastructure is essential for reducing unit transport costs, as demonstrated in my network analysis. The ethanol industry could coordinate to cross-subsidize marginal sources to ensure the greatest total number of sources and emissions are captured (see Figure 5.6), thereby lowering transport tariffs for all sources.

The importance of scale demonstrated in this study will be generally important for all CCUS developments that require transportation over significant distances. The most cost effective long-term solutions will include regionally coordinated and shared infrastructure. Pipeline infrastructure built with a long-term view will enable subsequent CCUS projects to be developed more quickly and cost effectively. Public policy will be essential in driving infrastructure to be developed in this way. Even where carbon dioxide pipelines are commercially financed and owned, governments can ensure the infrastructure is regionally coordinated and fast-tracked. For example, the Wyoming state government’s Wyoming Pipeline Authority is pre-emptively securing rights-of-way permits for future carbon dioxide pipeline corridors to ease and accelerate the process for pipeline developers (WPA, 2014; Wallace et al., 2015).

Governments could also fund additional initial capacity for pipelines to facilitate increased future CCUS, since commercial projects are not likely to significantly overbuild capacity beyond contracted carbon dioxide flow rates (Group, 2017b). For example, my full government scenario main trunk pipeline has diameter ranging from 30-inch down to 8-inch and a maximum capacity around 34 Mt per year. The trunk could be expanded to be uniformly 36-inch diameter with 50 Mt per year capacity for a 30% increase in total network capital cost ($2 billion), or to 42-inch diameter with 70 Mt per
year capacity for a 50% increase ($3.6 billion). Figure 5.8 shows the final full government-scenario pipeline network with other, higher capture-cost, carbon dioxide sources that were not considered in my analysis. There are a total of 220 Mt per year emitted by sources within 50 km of the pipeline network, including a cluster of non-electricity industrial emissions in northern Illinois and Indiana emitting 35 Mt per year. There would not be capacity to subsequently add these sources without significant new capital expenditure if the pipeline network were built only to optimize for existing ethanol sources.

**Figure 5.8** – The full government scenario pipeline network is shown here with other carbon dioxide sources not considered in my analysis. These sources are often much larger than ethanol biorefineries, as can be seen from the emissions scale. A number of large electricity and industrial emissions sources are located near the pipeline network, particularly in northern Illinois and Indiana, and in southern Iowa and Nebraska. The light purple area indicates 50 km proximity to the pipeline network. Non-ethanol sources within 50 km of the pipeline network emit a combined...
220 Mt per year compared with the network total of 28.7 Mt from ethanol biorefineries. Emission data for the figure were sourced from NETL (2015a).

The successful development of wind energy in Texas is an analogous case that shared many of the characteristics and challenges of the potential CCUS network. Texas has an exceptional wind resource in the west of the state, but its population and electricity demand are mostly in the east. When wind energy became economically attractive in the mid-2000s (supported by the wind production tax credit), there was insufficient electricity transmission capacity to transmit electricity generated in west Texas to the eastern cities and a chicken-and-egg problem between wind and transmission developers. The Texas Legislature passed a bill in 2005 ordering new electricity transmission be built to enable wind energy development (S.B. 20). The network was planned and authorized by the Public Utility Commission of Texas with consideration of future wind development requirements (Staine, 2014). The “Competitive Renewable Energy Zone” transmission network was completed in January 2014, which enabled the subsequent wind energy boom in Texas. The network consists of 5,800 km of new transmission lines with 11.5 GW of additional capacity, built at a cost of $6.9 billion (Lashner, 2014). These parameters are very similar to the full government scenario CCUS network, including equivalent carbon emission abatement.

A potential uncertainty for the viability of a Midwest CCUS network is the future of corn ethanol in the United States’ energy supply. Ethanol use in transportation fuels is largely driven by the federal Renewable Fuel Standard (RFS) program (EPA, 2018c). However, corn ethanol is often criticized for its food displacement, fertilizer use, relatively low energy return on energy input, and modest greenhouse gas benefit compared with fossil
petroleum (Hill et al., 2006; Searchinger et al., 2008; Tilman et al., 2009). These concerns have led to proposals to eliminate or reform the RFS (Holman, 2016). If the ethanol industry is significantly reduced in the next twenty years, it would not make sense to build a CCUS network based on Midwest ethanol. On the other hand, CCUS presents an opportunity for the ethanol industry. Capture and storage of fermentation carbon dioxide emissions can significantly reduce the overall greenhouse gas footprint of corn ethanol (Flugge et al., 2017; Hornafius & Hornafius, 2015; Laude et al., 2011), increasing its value as a low-carbon fuel. Ethanol with CCUS could realize this value through the CCUS tax credits and also through programs such as the California Low Carbon Fuel Standard (LCFS) (Yeh et al., 2016), a transportation fuels carbon emissions trading scheme with average carbon credit prices in the past two years around $100 per tonne abated (CARB, 2018). CCUS may be recognized under the LCFS in the near future (CARB, 2017). If all fermentation emissions at an ethanol biorefinery were captured and sold for EOR, the CCUS value would be $0.09 per gallon of ethanol from the $35 per tonne tax credit, and around $0.21 per gallon from an $100 per tonne LCFS carbon credit. This represents approximately 20% of the average market ethanol price of $1.50 per gallon over the past four years. CCUS could provide a significant additional revenue stream for the ethanol industry.

The ability of ethanol producers to monetize the tax credits is another potential challenge. The CCUS tax credits are not cash revenue; in order to monetize the value of the tax credits, project owners must have sufficient tax liability to offset. However, the wide variety of ownership structures of ethanol biorefineries, from large corporations to farmer cooperatives, means that many owners may not have sufficient tax liability. For those
with insufficient tax liability to offset directly, monetization of the tax credits would require either transfer of the credits to the company that stores the carbon dioxide through CO$_2$-EOR, which is allowed under Section 45Q, or tax equity investors and other complex financial arrangements that will likely be beyond the experience of these owners. Wind and solar projects receiving similar tax credits may provide a template for financial structures (Harper et al., 2007; Schwabe et al., 2017). The ethanol industry could make a coordinated effort to educate its members about the tax credits and options for fully realizing their value. My analysis assumed that all ethanol biorefineries would be able to fully monetize the value of the tax credits.

This study also has implications for the current and future structure of carbon emissions reduction policies. Carbon capture facility owners can claim the tax credits for a period of 12 years (Future Act, 2016). If there is no price on carbon dioxide or other supporting emissions reduction policy after their credit eligibility period, the capture facilities may cease to operate if transport and operational costs exceed the carbon dioxide CO$_2$-EOR sales price. The capture facilities will therefore need to be financed over a maximum of 12 years and repay their capital within that time. This also implies that the pipeline network financing will not be able to assume demand beyond 12 years. Pipelines are very long-lived assets, however, and initial tariffs could be lower if they can repay capital over longer time periods. In my financing scenarios, I assumed that government bears the post-tax credit policy risk and provides 20-year debt finance for the commercial-government and full government financing scenarios. Long-term confidence in emissions reduction policy would be beneficial for reducing system costs today and maximizing deployment.
Even assuming there are strong carbon emissions policies in place by the mid 2030s when the last tax credits are earned, they may not support carbon capture at ethanol biorefineries without targeted consideration in policy design. Ethanol fermentation emissions are biogenic and therefore are not currently reported to the EPA Greenhouse Gas Reporting Program and may not be taxed under a carbon tax or liable for permits in an emissions trading scheme (EPA, 2017). Such programs would need to be designed so that CCUS of biogenic emissions generates credits that can be sold to liable entities in order to incentivize CCUS on ethanol biorefineries.
Chapter 6

Concluding Remarks and Future Work

This dissertation addressed important questions regarding energy and the subsurface, focused on shale gas and geological carbon storage, using a range of modeling and analysis methodologies. The work includes practical application of an existing modeling framework, the development of new numerical models and their application to practical questions, and a network economic optimization and policy analysis. The subsurface flow modeling was guided by the collation of a comprehensive set of geological, physical, and engineering data on the shale gas system and by comparison with high quality production data.

In Chapter 2, I investigated the fate of hydraulic fracturing fluid injected into shale gas formations by building and implementing a numerical model of two-phase water and gas flow with the MRST subsurface flow modeling framework. I found that most of the injected fracturing fluid imbibes into the shale rock matrix and is retained there for the long-term, implying that the risk of upward migration of hydraulic fracturing fluid from shale gas formations is likely to be very low in the long-term. A priority for further research to build upon the work presented in this dissertation is to apply the same model to a number of other locations where sufficient high-quality data can be obtained. These locations should include different wells within the same formation and wells in different
shale formations. Application of the model to different locations will help determine how general my results are, guide further refinement of the model, and build more confidence in its implications.

In Chapter 3, I developed a modified numerical model of capillary pressure hysteresis that is significantly more robust and computationally efficient compared with existing models under conditions with multiple saturation turns. I showed that my modified model can produce very similar results to existing capillary pressure models. The modified model is particularly useful for application to the shale gas system where the complexity of capillary pressure behavior means that the existing models are impractical to use. Further experimental work on capillary pressure behavior in shales, particularly focused on determining hysteretic scanning curve behavior under in-situ conditions, would be useful to establish the applicability of the modified model and guide potential further alterations.

In Chapter 4, I developed a numerical model of single-phase, two-component carbon dioxide and methane flow in shale formations, including adsorption effects, for investigating carbon dioxide injection into depleted shale formations for geological carbon storage. Application of the model to major shale gas formations showed that the total capacity of individual shale wells is small and that injection rates would be very low. Therefore, shales are unlikely to be an economically feasible resource for large-scale carbon storage. The most uncertain element of the model is the applicability of the adsorption function and its parameters. Further experiments on methane and carbon dioxide adsorption in shale gas formations, especially under in-situ conditions, should be a priority for future work. Experiments should also a focus on whether there is a
significant relationship between adsorbed carbon dioxide and permeability (swelling) in shales. The model presented in this dissertation does not include geomechanics: if the model is to be applied to scenarios with higher injection pressures that exceed the initial shale formation pressure, geomechanics may be important to include in the model.

Finally, in Chapter 5, I performed a carbon dioxide pipeline network economic optimization analysis to investigate potential deployment of carbon capture, utilization, and storage as a result of new tax credits enacted in February 2018. I found that a large-scale network capturing carbon dioxide from ethanol biorefineries in the Midwest and delivering it for enhanced oil recovery in the Permian Basin, Texas, could be feasible with low-cost government financing of the pipeline network. There are many potential future possibilities for building upon this work; the most valuable directions will be guided by political, policy, and technological developments in carbon capture, utilization, and storage. Potentially useful further research includes exploring different financial and policy scenarios and assessing opportunities in different locations. One useful example may be assessment of a scenario with government-financed trunk pipelines but commercially-financed feeder pipelines. If the Midwest network presented in this dissertation gains stakeholder interest, an important future task will be to analyze how the pipeline network ownership and finance could be structured; for example, one option could be a new public utility jointly owned by stakeholder state governments and the federal government. Yet another option could be to consider potential pathways for the build-out and scale-up of carbon capture and storage beyond initial projects, to determine how initial pipelines and networks can be planned such that they contribute to an efficient future regional and national system.
References


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Eastern Regional Meeting, Charleston, WV, USA.


Appendix A
Tables of Collated Gas Shale Data
Collated data for the shale gas system that were used to inform my model development and parameter selection are presented in Tables A.1 to A.4. These data were published in a data report in *Water Resources Research* (Edwards & Celia, 2018b). The presented data are mainly from the Marcellus, Barnett and Horn River formations for which data are most readily available in publicly accessible sources (the Marcellus and Barnett because of their prominence in shale gas production, and the Horn River because of the public availability of data in British Columbia, Canada). The source data are heterogeneous in terms of the experimental and measurement techniques used, the way data were reported, units of measure, and the quality of the publications in which they appear. I have standardized the data, most commonly by converting from oilfield units to a common SI unit. The capillary pressure data were most complex to standardize, so additional information is given in Figure A.1 and in the supporting information of Edwards and Celia (2018b). I recommend that anyone interested in using a particular reported parameter should examine the original sources to fully understand the context of the data.

Despite the heterogeneity of the sources, the collated data show significant consistency in properties both within and between the different shale gas formations. For example, porosity, permeability, and organic content are similar. I do not present any data directly for shale oil formations (such as the Bakken) since my focus has been on shale gas formations, but there is likely much similarity in hydraulic fracturing and rock properties.

Due to the variety of sources, there are differing data formats shown within the tables. The first number reported is a typical value, mean, or median unless otherwise specified. In some cases, a typical range is shown. In parentheses, the number of wells from which
data were available, the time period for which the data were collected, or a typical range of variability is shown.

**Table A.1 – Shale Horizontal Well Data**

<table>
<thead>
<tr>
<th>Data parameter</th>
<th>Value</th>
<th>Formation and source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Horizontal well length, L_h</td>
<td>1,390 m (2010)</td>
<td>Marcellus&lt;sup&gt;a,b&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>1,460 m (2011)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1,560 m (2012)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1,710 m (2013)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1,850 m (2014)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>730 m (2007)</td>
<td>Barnett&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>815 m (2008)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>890 m (2009)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>945 m (2010)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1,090 m (2011)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1,990 m (max. 3,560 m) (184 wells)</td>
<td>Horn River&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>1,540 m (2008 – 2014)</td>
<td>Haynesville&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

| Perforation spacing, L_s        | 21 – 30 m                          | Marcellus<sup>d</sup> |
|                                 | 25 m or 40 m                       | Horn River<sup>c,e</sup> |
|                                 | 23 – 30 m                          | Haynesville<sup>f</sup> |

<table>
<thead>
<tr>
<th>Bottom-hole production pressure (long-term)</th>
<th>Value</th>
<th>Formation and source</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>~4 MPa</td>
<td>Marcellus&lt;sup&gt;g&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>3.5 MPa</td>
<td>Barnett&lt;sup&gt;h&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>3.5 – 7 MPa</td>
<td>Horn River&lt;sup&gt;e,i&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

<sup>a</sup>(Drillinginfo, 2014; Edwards et al., 2015). <sup>b</sup>(Q. Zhou et al., 2016). <sup>c</sup>(BCOGC, 2016). <sup>d</sup>(Byrnes, 2011; Clarkson & Williams-Kovacs, 2013; Shelley et al., 2014). <sup>e</sup>(Abbasi, 2013). <sup>f</sup>(J. W. Thompson et al., 2011). <sup>g</sup>(J. M. Thompson et al., 2011). <sup>h</sup>(Patzek et al., 2013). <sup>i</sup>(Zhang & Ehlig-Economides, 2014).

**Table A.2 – Hydraulic Fracturing Data**

<table>
<thead>
<tr>
<th>Data parameter</th>
<th>Value</th>
<th>Formation and source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total injected fracturing fluid volume</td>
<td>20,000 m³ (16,000 – 26,000 m³)</td>
<td>Marcellus&lt;sup&gt;a,g,h,u&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>19,000 m³ (11,000 – 23,000 m³)</td>
<td>Barnett&lt;sup&gt;b,e,f,i&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>77,000 m³ (mean), 66,000 m³ (median) (35 wells) (2013-2014)</td>
<td>Horn River&lt;sup&gt;c,d&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>64,000 m³ (2010-2012)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>19,000 m³ (6,000 – 25,000 m³)</td>
<td>Haynesville&lt;sup&gt;e,f&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>23,000 m³</td>
<td>Eagle Ford&lt;sup&gt;e&lt;/sup&gt;</td>
</tr>
<tr>
<td>Data parameter</td>
<td>Value</td>
<td>Formation and source</td>
</tr>
<tr>
<td>---------------------------------------------------</td>
<td>--------------------------------------------</td>
<td>----------------------</td>
</tr>
<tr>
<td>Injected fluid volume normalized by horizontal well length</td>
<td>14 m$^3$/m (235 wells)</td>
<td>Marcellus$^g,h$</td>
</tr>
<tr>
<td></td>
<td>19 m$^3$/m (2006)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>15 m$^3$/m (2008-2012)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>27 m$^3$/m (35 wells) (2012-2014)</td>
<td>Horn River$^e$</td>
</tr>
<tr>
<td>Injected volume flowback recovery</td>
<td>1 – 50%</td>
<td>Marcellus$^g,j$</td>
</tr>
<tr>
<td></td>
<td>65% (1 year)</td>
<td>Barnett$^l$</td>
</tr>
<tr>
<td></td>
<td>90% (2 years)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>100% (3 years)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>13% (8 wells)</td>
<td>Horn River$^k$</td>
</tr>
<tr>
<td></td>
<td>5%</td>
<td>Haynesville$^j$</td>
</tr>
<tr>
<td>Surface injection pressure</td>
<td>45 – 62 MPa</td>
<td>Marcellus$^m$</td>
</tr>
<tr>
<td></td>
<td>54 MPa (max. 22 wells)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>49 MPa (avg. 22 wells)</td>
<td>Horn River$^e$</td>
</tr>
<tr>
<td>Bottom-hole injection pressure</td>
<td>55 – 83 MPa (30 – 55 MPa surface injection pressure)</td>
<td>Woodford$^n$</td>
</tr>
<tr>
<td></td>
<td>48 – 85 MPa</td>
<td>Unspecified$^o$</td>
</tr>
<tr>
<td>Number of stages</td>
<td>12 (7 – 24) (184 wells)</td>
<td>Marcellus$^g$</td>
</tr>
<tr>
<td></td>
<td>18</td>
<td>Horn River$^{d,p}$</td>
</tr>
<tr>
<td>Fluid injection duration per stage</td>
<td>2 – 3 hours</td>
<td>Marcellus$^{l,m,q}$</td>
</tr>
<tr>
<td></td>
<td>3 – 4 hours</td>
<td>Horn River$^{e,r}$</td>
</tr>
<tr>
<td></td>
<td>2.5 – 3 hours</td>
<td>Woodford$^n$</td>
</tr>
<tr>
<td>Average injection flow rate (for the duration of each stage)</td>
<td>12 m$^3$/min</td>
<td>Marcellus$^{g,h,q,s}$</td>
</tr>
<tr>
<td></td>
<td>8 – 16 m$^3$/min</td>
<td>Barnett$^{b,t}$</td>
</tr>
<tr>
<td></td>
<td>16 m$^3$/min (35 wells)</td>
<td>Horn River$^e$</td>
</tr>
<tr>
<td></td>
<td>15 m$^3$/min</td>
<td>Woodford$^n$</td>
</tr>
<tr>
<td>Injected proppant mass (per well)</td>
<td>2,100 tonnes (400 – 3,600 tonnes) (187 wells)</td>
<td>Marcellus$^{g,h,u}$</td>
</tr>
<tr>
<td></td>
<td>3,000 tonnes (48 wells)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>4,000 tonnes</td>
<td>Horn River$^e$</td>
</tr>
<tr>
<td>Fracture height inferred from microseismic measurements, $H_m$</td>
<td>~160 m (median), ~500 m (max.)</td>
<td>Marcellus$^{g,v,w}$</td>
</tr>
<tr>
<td></td>
<td>~160 m (median)</td>
<td>Barnett$^w$</td>
</tr>
<tr>
<td></td>
<td>250 m (12 wells)</td>
<td>Horn River$^e$</td>
</tr>
<tr>
<td></td>
<td>~130 m (median)</td>
<td>Woodford$^w$</td>
</tr>
<tr>
<td></td>
<td>~100 m (median)</td>
<td>Eagle Ford$^w$</td>
</tr>
<tr>
<td>Fracture horizontal length inferred from microseismic measurements</td>
<td>~300 – 400 m</td>
<td>Marcellus$^g$</td>
</tr>
<tr>
<td></td>
<td>~600 – 900 m (12 wells)</td>
<td>Horn River$^e$</td>
</tr>
<tr>
<td>Data parameter</td>
<td>Value</td>
<td>Formation and source</td>
</tr>
<tr>
<td>--------------------------------------------------------------------------------</td>
<td>----------------------------------------------------------------------</td>
<td>----------------------</td>
</tr>
<tr>
<td>Hydraulic fracturing fluid-gas interfacial tension (measurements are mostly on HF fluid-air)</td>
<td>30 – 70 milliNewton/m (decreases with increasing pressure, temperature, or surfactant concentration)</td>
<td>Not applicable&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>Hydraulic fracturing fluid viscosity (lab temp., atm. pressure)</td>
<td>1 – 4 centipoise (cP)</td>
<td>Not applicable&lt;sup&gt;y,z&lt;/sup&gt;</td>
</tr>
<tr>
<td>Shut-in time</td>
<td>19 days (0 – 144 days) (189 wells)</td>
<td>Marcellus&lt;sup&gt;g,z&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>60 days (18 wells)</td>
<td>Horn River&lt;sup&gt;aa&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

<sup>a</sup>(Gallegos et al., 2015; Jackson et al., 2015; Mantell, 2011).  
<sup>b</sup>(Martineau, 2007).  
<sup>c</sup>(BCOGC, 2016).  
<sup>d</sup>(BCOGC, 2014).  
<sup>e</sup>(Mantell, 2011).  
<sup>f</sup>(Jackson et al., 2015).  
<sup>g</sup>(Q. Zhou et al., 2016).  
<sup>h</sup>(Shelley et al., 2014).  
<sup>i</sup>(Nicot et al., 2014).  
<sup>j</sup>(Boschee, 2014; He, 2011; Rahm et al., 2013; Vidic et al., 2013).  
<sup>k</sup>(Xu et al., 2015).  
<sup>l</sup>(King, 2012).  
<sup>m</sup>(BCOGC, 2016).  
<sup>n</sup>(Mantell, 2011).  
<sup>o</sup>(Jackson et al., 2015).  
<sup>p</sup>(Abbasi, 2013).  
<sup>q</sup>(Mayerhofer et al., 2011).  
<sup>r</sup>(Ahmed & Ehlig-Economides, 2013).  
<sup>s</sup>(Ciezobka, 2013; Ciezobka et al., 2016; Rodvelt et al., 2015).  
<sup>t</sup>(Neuhaus & Miskimins, 2012).  
<sup>u</sup>(Vidic et al., 2013).  
<sup>v</sup>(Martineau, 2007).  
<sup>w</sup>(Jackson et al., 2015).  
<sup>x</sup>(Bachu & Bennion, 2009; Busch & Amann-Hildenbrand, 2013; Howard et al., 2010; Kashefi et al., 2016; Rostami et al., 2016; L. Zhou et al., 2016).  
<sup>y</sup>(Shelley et al., 2014).  
<sup>z</sup>(Clarkson & Williams-Kovacs, 2013).  
<sup>aa</sup>(Ghanbari & Dehghanpour, 2016).

---

**Table A.3 – Hydraulic Fracture Property Estimates and Lab Measurements**

<table>
<thead>
<tr>
<th>Data parameter</th>
<th>Value</th>
<th>Formation and source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Propped hydraulic fracture conductivity (fracture permeability multiplied by aperture)</td>
<td>4 – 100 mD·ft</td>
<td>Marcellus&lt;sup&gt;a,b&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>0.5 – 500 mD·ft</td>
<td>Barnett&lt;sup&gt;c,d&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>0.1 – 10 mD·ft</td>
<td>Horn River&lt;sup&gt;e&lt;/sup&gt;</td>
</tr>
<tr>
<td>Closed un-propped fracture conductivity</td>
<td>0.0001 – 0.1 mD·ft</td>
<td>Horn River&lt;sup&gt;e&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>0.0001 – 1 mD·ft</td>
<td>Barnett&lt;sup&gt;e,f&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>0.05 – 5 mD·ft</td>
<td>Eagle Ford&lt;sup&gt;g&lt;/sup&gt;</td>
</tr>
<tr>
<td>Hydraulic fracture aperture (before closure)</td>
<td>2 – 8 mm</td>
<td>Non-specific&lt;sup&gt;h&lt;/sup&gt;</td>
</tr>
<tr>
<td>Estimated effective fracture volume</td>
<td>~43,000 m&lt;sup&gt;3&lt;/sup&gt; (28,000 – 60,000 m&lt;sup&gt;3&lt;/sup&gt;)</td>
<td>Horn River&lt;sup&gt;d&lt;/sup&gt;</td>
</tr>
<tr>
<td>Propped proportion of created fracture area</td>
<td>5 – 50%</td>
<td>Non-specific&lt;sup&gt;g,i&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

<sup>a</sup>(Clarkson & Williams-Kovacs, 2013).  
<sup>b</sup>(Izadi et al., 2014).  
<sup>c</sup>(Cipolla et al., 2010; Zhang et al., 2014).  
<sup>d</sup>(Cipolla & Wallace, 2014).  
<sup>e</sup>(Cipolla et al., 2014).  
<sup>f</sup>(Kam et al., 2015).  
<sup>g</sup>(Zhang et al., 2014).  
<sup>h</sup>(Wu et al., 2017).  
<sup>i</sup>(Kim & Moridis, 2015; King, 2012).  
<sup>j</sup>(Ezulike et al., 2016; Xu et al., 2015).  
<sup>k</sup>(Byrnes, 2011; Sharma & Manchanda, 2015).
**Table A.4 – Shale Formation Data**

<table>
<thead>
<tr>
<th>Data parameter</th>
<th>Value</th>
<th>Formation and source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Depth to top of target formation, $D_t$</td>
<td>1,600 – 2,600 m</td>
<td>Marcellus$^{a,b,c,d}$</td>
</tr>
<tr>
<td></td>
<td>1,400 – 2,600 m</td>
<td>Barnett$^{b,c,e}$</td>
</tr>
<tr>
<td></td>
<td>2,420 m (180 wells)</td>
<td>Horn River$^f$</td>
</tr>
<tr>
<td></td>
<td>3,300 – 3,500 m</td>
<td>Haynesville$^{c,g}$</td>
</tr>
<tr>
<td></td>
<td>2,000 – 3,900 m</td>
<td>Woodford$^b$</td>
</tr>
<tr>
<td></td>
<td>1,800 – 3,600 m</td>
<td>Eagle Ford$^b$</td>
</tr>
<tr>
<td>Formation thickness, $T_f$</td>
<td>15 – 100 m</td>
<td>Marcellus$^{a,h}$</td>
</tr>
<tr>
<td></td>
<td>75 – 200 m</td>
<td>Barnett$^{a,i}$</td>
</tr>
<tr>
<td></td>
<td>140 – 280 m</td>
<td>Horn River$^{d,j}$</td>
</tr>
<tr>
<td>Pay zone (productive) thickness</td>
<td>65 m (5 – 170 m) (170 wells)</td>
<td>Horn River$^f$</td>
</tr>
<tr>
<td></td>
<td>70 – 100 m</td>
<td>Haynesville$^k$</td>
</tr>
<tr>
<td>Bottom-hole (formation) temperature</td>
<td>60 – 80 °C</td>
<td>Marcellus$^{a,l}$</td>
</tr>
<tr>
<td></td>
<td>129 °C (70 – 183°C) (118 wells)</td>
<td>Horn River$^f$</td>
</tr>
<tr>
<td></td>
<td>150 °C</td>
<td>Haynesville$^e$</td>
</tr>
<tr>
<td>Formation pressure</td>
<td>28 – 43 MPa</td>
<td>Marcellus$^{l,m}$</td>
</tr>
<tr>
<td></td>
<td>20 – 27 MPa</td>
<td>Barnett$^{a,e,n}$</td>
</tr>
<tr>
<td></td>
<td>36 MPa (76 wells)</td>
<td>Horn River$^f$</td>
</tr>
<tr>
<td>Gas composition (C$_2$ = ethane, C$_3$ = propane)</td>
<td>95.5 % CH$_4$, 0.3 % CO$_2$, 0.2 % N$_2$, 3 % C$_2$, 1 % C$_3$ (1 well)</td>
<td>Marcellus (NE dry region)$^o$</td>
</tr>
<tr>
<td></td>
<td>81 % CH$_4$, 0.4 % CO$_2$, 0.3 % N$_2$, 14 % C$_2$, 3.5 % C$_3$ (3 wells)</td>
<td>Marcellus (SW wet region)$^o$</td>
</tr>
<tr>
<td></td>
<td>85 % CH$_4$, 1.6 % CO$_2$, 2.7 % N$_2$, 6.5 % C$_2$, 1.9 % C$_3$ (4 wells)</td>
<td>Barnett$^p$</td>
</tr>
<tr>
<td></td>
<td>87 % CH$_4$, 10 % CO$_2$, 2.3 % N$_2$, 0.2 % C$_2$ (87 wells)</td>
<td>Horn River$^{f,q}$</td>
</tr>
<tr>
<td></td>
<td>95.5 % CH$_4$, 4.8 % CO$_2$, 0.1 % N$_2$, 0.1 % C$_2$</td>
<td>Haynesville$^o$</td>
</tr>
<tr>
<td>Porosity</td>
<td>5 – 7 %</td>
<td>Marcellus$^{a,r,ff}$</td>
</tr>
<tr>
<td></td>
<td>4.5 – 6.5 %</td>
<td>Barnett$^{a,j,na}$</td>
</tr>
<tr>
<td></td>
<td>3 – 8 %</td>
<td>Horn River$^{f,j,cc}$</td>
</tr>
<tr>
<td></td>
<td>8 – 12 %</td>
<td>Haynesville$^{ka}$</td>
</tr>
<tr>
<td></td>
<td>6 % (2 – 10 %) (200 samples)</td>
<td>Woodford$^r$</td>
</tr>
<tr>
<td>Horizontal intrinsic permeability</td>
<td>20 – 1,000 nD</td>
<td>Marcellus$^{c,d,w,aa}$</td>
</tr>
<tr>
<td></td>
<td>20 – 1,800 nD</td>
<td>Barnett$^{a,aa}$</td>
</tr>
<tr>
<td></td>
<td>100 – 900 nD</td>
<td>Horn River$^{j,y,cc}$</td>
</tr>
<tr>
<td>Data parameter</td>
<td>Value</td>
<td>Formation and source</td>
</tr>
<tr>
<td>----------------------------------------------</td>
<td>--------------------------------------------</td>
<td>----------------------</td>
</tr>
<tr>
<td>Horizontal intrinsic permeability</td>
<td>100 – 500 nD</td>
<td>Haynesville&lt;sup&gt;ze&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>6 – 40 nD (1 sample)</td>
<td>Eagle Ford&lt;sup&gt;aa&lt;/sup&gt;</td>
</tr>
<tr>
<td>Vertical intrinsic permeability</td>
<td>1 – 10% of horizontal</td>
<td>Marcellus&lt;sup&gt;cc&lt;/sup&gt;</td>
</tr>
<tr>
<td>Initial in-situ water saturation</td>
<td>15 – 30 %</td>
<td>Marcellus&lt;sup&gt;ac,mm,pp&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>25 – 40 %</td>
<td>Barnett&lt;sup&gt;an&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>25 %</td>
<td>Horn River&lt;sup&gt;fa,cc&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>20 – 30 %</td>
<td>Haynesville&lt;sup&gt;ik&lt;/sup&gt;</td>
</tr>
<tr>
<td>Residual non-wetting gas phase saturation</td>
<td>26 – 35% (2 samples)</td>
<td>Alberta shales&lt;sup&gt;dd&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>20 – 40% (13 samples)</td>
<td>Unspecified tight rock&lt;sup&gt;ee&lt;/sup&gt;</td>
</tr>
<tr>
<td>Total organic content (TOC)</td>
<td>2 – 10% by weight</td>
<td>Marcellus&lt;sup&gt;a,ff,gg,hh&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>2 – 7% wt.</td>
<td>Barnett&lt;sup&gt;e,n,gg,hh,ii&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>3.6 % wt. (1 – 5%)</td>
<td>Horn River&lt;sup&gt;cc,gj&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>0 – 8% wt.</td>
<td>Haynesville&lt;sup&gt;ka,gg,hh&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>5.3% wt. (0 – 13%) (193 samples)</td>
<td>Woodford&lt;sup&gt;tv&lt;/sup&gt;</td>
</tr>
<tr>
<td>Maximum CH&lt;sub&gt;4&lt;/sub&gt; adsorption</td>
<td>1.5 – 10.7 kg CH&lt;sub&gt;4&lt;/sub&gt;/m&lt;sup&gt;3&lt;/sup&gt; rock</td>
<td>Marcellus&lt;sup&gt;kk,ll,mm&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>3.9 – 6.4 kg CH&lt;sub&gt;4&lt;/sub&gt;/m&lt;sup&gt;3&lt;/sup&gt; rock</td>
<td>Barnett&lt;sup&gt;ll&lt;/sup&gt;</td>
</tr>
<tr>
<td>Langmuir adsorption constant</td>
<td>0.11 – 0.27 MPa&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>Marcellus&lt;sup&gt;kk,ll&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>0.17 – 0.34 MPa&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>Barnett&lt;sup&gt;ll&lt;/sup&gt;</td>
</tr>
<tr>
<td>Water-rock contact angle (in water-air fluid pair)</td>
<td>20 – 70°</td>
<td>Marcellus&lt;sup&gt;am,pp&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>14 – 65°</td>
<td>Horn River&lt;sup&gt;oo&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>40 – 50°</td>
<td>Haynesville&lt;sup&gt;pp&lt;/sup&gt;</td>
</tr>
<tr>
<td>Capillary pressure</td>
<td>Shown in Fig. 2</td>
<td>Barnett&lt;sup&gt;c,qq&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>Shown in Fig. 2</td>
<td>Barnett&lt;sup&gt;c,qq&lt;/sup&gt;</td>
</tr>
<tr>
<td>Water relative permeability Corey exponent</td>
<td>4 – 4.3 (2 samples)</td>
<td>Alberta shales&lt;sup&gt;dd&lt;/sup&gt;</td>
</tr>
<tr>
<td>Gas relative permeability Corey exponent</td>
<td>2.2 – 3.5 (2 samples)</td>
<td>Alberta shales&lt;sup&gt;dd&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

<sup>a</sup>(NETL, 2011). <sup>b</sup>(Fisher & Warpinski, 2012). <sup>c</sup>(Byrnes, 2011). <sup>d</sup>(He, 2011). <sup>e</sup>(Jarvie et al., 2007). <sup>f</sup>(BCOGC, 2016). <sup>g</sup>(J. W. Thompson et al., 2011). <sup>h</sup>(Shelley et al., 2014). <sup>i</sup>(Petzet, 2008). <sup>j</sup>(BCOGC, 2014; Ghanbari & Dehghanpour, 2016). <sup>k</sup>(Hammes et al., 2011). <sup>l</sup>(Clarkson & Williams-Kovacs, 2013). <sup>m</sup>(Engelder, 2012). <sup>n</sup>(Bowker, 2007). <sup,o</sup>(Bullin & Krouskop, 2009). <sup>p</sup>(Hill et al., 2007). <sup>q</sup>(BCOGC, 2014). <sup>r</sup>(Gu et al., 2015). <sup>s</sup>(Vega et al., 2013). <sup>t</sup>(Dong et al., 2015; King et al., 2015; Nieto et al., 2009; Z. Zhou et al., 2016c). <sup>u</sup>(Kuila et al., 2014). <sup>v</sup>(Gupta et al., 2013). <sup>w</sup>(Bostrom et al., 2014; Chakraborty & Karpyn, 2015; Ettehadtavakkol & Jamali, 2016). <sup>x</sup>(Vermylen, 2011). <sup>y</sup>(Chalmers et al., 2012b). <sup>z</sup>(Chakraborty & Karpyn, 2015). <sup>aa</sup>(Heller et al., 2014). <sup>bb</sup>(King, 2014). <sup>cc</sup>(Chen & Hannigan, 2016). <sup>dd</sup>(Bennion & Bachu, 2007, 2008). <sup>ee</sup>(Dacy,
Figure A.1 Capillary pressure data. The graph shows water-gas capillary pressure (megapascals) versus water saturation. Data are from lab measurements on rock samples from the Barnett Shale (Kale et al., 2010; Sigal, 2013) Horn River Formation (Chalmers et al., 2012b) and an unspecified shale gas formation (Byrnes, 2011). The thick lines are primary drainage curves, and the thin dashed blue curves are main imbibition curves for the two Barnett Shale sources. These drainage-imbibition curve pairs show strong capillary pressure hysteresis. The experimental data from all sources are from mercury-air intrusion experiments, but the experimental methods and data formats differ among the different publications. The Chalmers et al. (2012b) primary drainage curve is dashed because the data are effective saturations (normalized by the residual saturations), while the Byrnes (2011), Sigal (2013), and Kale et al. (2010) data are absolute saturations. Since residual saturation data were not reported, I could not convert all data to a common saturation format. Note that the primary drainage curves end at a common capillary

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pressure limit imposed by the experimental apparatus, so the curves do not reach residual water saturation. Despite the heterogeneity in the data sources, the conversion issues will not fundamentally alter the comparison between the data: they show similar capillary pressure magnitude among these different shale gas formations.
Appendix B

Model Parameters for Hydraulic Fracturing Fluid Injection Modeling
Table B.1 – Model parameters for the hydraulic fracturing fluid injection simulations in Chapter 2. Hydraulic fracture parameters are differentiated between propped and unpropped regions; where no differentiation is indicated, the parameter is applied to both regions. Indirect parameters (that were used to calculate direct parameters for the model, or are calculated based on direct model parameters) are shown in italics.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Source / Guidance</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Well and Hydraulic Fracture Geometry Parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Horizontal well length</td>
<td>2,200 m</td>
<td>Direct well data (Table A.1)</td>
</tr>
<tr>
<td>Hydraulic fracture spacing, (d)</td>
<td>25 m / 40 m</td>
<td>Direct well data (Table A.1)</td>
</tr>
<tr>
<td><em>Number of hydraulic fractures, (n_f)</em></td>
<td>88 / 55</td>
<td>Calculated from above data</td>
</tr>
<tr>
<td>Propped hydraulic fracture height, (H_p)</td>
<td>60 m</td>
<td>History-matching + data (Table A.2)</td>
</tr>
<tr>
<td>Propped hydraulic fracture half-length, (L_p)</td>
<td>100 m</td>
<td>History-matching + data (Table A.2)</td>
</tr>
<tr>
<td>Un-propped hydraulic fracture height, (H_u)</td>
<td>160 m</td>
<td>Horn River Fm. thickness data (Table A.4)</td>
</tr>
<tr>
<td>Un-propped hydraulic fracture half-length, (L_u)</td>
<td>450 m</td>
<td>Horn River Fm. microseismic data (Table A.2)</td>
</tr>
<tr>
<td>Propped hydraulic frac. effective aperture</td>
<td>22 mm</td>
<td>History-matched</td>
</tr>
<tr>
<td>Un-propped hydraulic fracture effective aperture</td>
<td>3.6 mm</td>
<td>History-matching + data</td>
</tr>
<tr>
<td>Propped hydraulic fracture porosity</td>
<td>60 %</td>
<td>Assume fractures partially filled by sand proppant</td>
</tr>
<tr>
<td>Un-propped hydraulic fracture porosity</td>
<td>60 %</td>
<td>Assume irregular fracture surface, partially closed fractures</td>
</tr>
<tr>
<td><em>Propped hydraulic fracture pore volume (fracture spacing in brackets)</em></td>
<td>14,000 m³ (25 m) 8,700 m³ (40 m)</td>
<td>Calculated from fracture parameters</td>
</tr>
<tr>
<td><em>Un-propped hyd. fracture pore volume (fracture spacing in brackets)</em></td>
<td>25,100 m³ (25 m) 15,700 m³ (40 m)</td>
<td>Calculated from fracture parameters</td>
</tr>
<tr>
<td><em>Propped hydraulic fracture surface area (fracture spacing in brackets)</em></td>
<td>2.1×10⁶ m² (25 m) 1.3×10⁶ m² (40 m)</td>
<td>Calculated from fracture parameters</td>
</tr>
<tr>
<td><em>Un-propped hyd. fracture surface area (fracture spacing in brackets)</em></td>
<td>23.2×10⁶ m² (25m) 14.5×10⁶ m² (40 m)</td>
<td>Calculated from fracture parameters</td>
</tr>
<tr>
<td><strong>Well Completion and Hydraulic Fracturing Process Parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hyd. fracturing fluid injection duration</td>
<td>4 hours</td>
<td>Horn River Fm. data (Table A.2)</td>
</tr>
<tr>
<td>Hydraulic fracturing bottom-hole injection pressure</td>
<td>70 MPa</td>
<td>Injection pressure data (Table A.2)</td>
</tr>
<tr>
<td>Parameter</td>
<td>Value</td>
<td>Source / Guidance</td>
</tr>
<tr>
<td>--------------------------------------------------</td>
<td>----------------------------</td>
<td>--------------------------------------------</td>
</tr>
<tr>
<td><strong>Shut-in period duration</strong></td>
<td>60 days</td>
<td>Direct well data (Table A.2)</td>
</tr>
<tr>
<td><strong>Production period duration</strong></td>
<td>4 years +</td>
<td>Exceeds production data duration</td>
</tr>
<tr>
<td><strong>Formation, Fracture and Fluid Parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Shale matrix porosity, $\phi$</td>
<td>6 %</td>
<td>Horn River Fm. data (Table A.4)</td>
</tr>
<tr>
<td>Initial shale matrix water saturation</td>
<td>25 %</td>
<td>Horn River Fm. data (Table A.4)</td>
</tr>
<tr>
<td>Initial hydraulic fracture water saturation</td>
<td>100 %</td>
<td>Implicitly assume fractures created and filled with water instantly</td>
</tr>
<tr>
<td>Shale matrix residual saturations (water / gas)</td>
<td>23 % / 40 %</td>
<td>Initial saturation (water) / other shale fm. data (gas, Table A.4)</td>
</tr>
<tr>
<td>Hydraulic fracture residual saturations water / gas</td>
<td>5 % / 5 %</td>
<td>Typical coarse, very permeable sand</td>
</tr>
<tr>
<td><strong>Gas composition</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gas composition</td>
<td>90 % CH$_4$, 10 % CO$_2$</td>
<td>Horn River Fm. data (Table A.4)</td>
</tr>
<tr>
<td><strong>Formation temperature</strong></td>
<td>130 °C</td>
<td>Horn River Fm. data (Table A.4)</td>
</tr>
<tr>
<td>Water density, $\rho_w(p)$</td>
<td>~1,100 – 1,130 kg per m$^3$</td>
<td>Defined by fm. water salinity (assume 150,000 mg/L), fm. temp., pressure. See graph in Edwards et al. (2017) supporting information</td>
</tr>
<tr>
<td>Water viscosity, $\mu_w$</td>
<td>0.7 centipoise (cP)</td>
<td>History-matching guided by water &amp; HF fluid viscosity at room temp. (Table A.2), water visc.-temp. function (NIST, 2016) and formation temp.</td>
</tr>
<tr>
<td>Gas density, $\rho_g(p)$</td>
<td>~20 – 330 kg per m$^3$</td>
<td>Defined by gas composition &amp; temp. See graph in Edwards et al. (2017) supporting information</td>
</tr>
<tr>
<td>Gas viscosity, $\mu_g(p)$</td>
<td>~0.015 – 0.04 cP</td>
<td>Defined by gas composition &amp; temp. See graph in Edwards et al. (2017) supporting information</td>
</tr>
<tr>
<td>Initial formation gas pressure, $p_f$</td>
<td>36 MPa</td>
<td>Horn River Fm. data (Table A.4)</td>
</tr>
<tr>
<td>Production bottom-hole pressure, $p_f(t)$</td>
<td>30 MPa – 6 MPa</td>
<td>Horn River Fm. data (Table A.1). See graph in Edwards et al. (2017) supporting information</td>
</tr>
<tr>
<td>Intrinsic shale matrix horizontal permeability</td>
<td>50 nanoDarcy (nD)</td>
<td>Effective permeability history-matched from one-phase simulation, divided by maximum relative permeability (0.3)</td>
</tr>
<tr>
<td>Intrinsic shale matrix vertical permeability</td>
<td>2.5 nD</td>
<td>Other shale formation data (Table A.4)</td>
</tr>
<tr>
<td>Shale matrix relative permeability (water / gas)</td>
<td>Corey n = 4 / 2.2</td>
<td>Other shale formation data (Table A.4)</td>
</tr>
<tr>
<td>Hydraulic fracture relative permeability (water / gas)</td>
<td>Corey n = 1 / 1</td>
<td>Convention for fractures</td>
</tr>
<tr>
<td>Parameter</td>
<td>Value</td>
<td>Source / Guidance</td>
</tr>
<tr>
<td>--------------------------------------------------------------------------</td>
<td>------------------</td>
<td>----------------------------------------------------------------------------------</td>
</tr>
<tr>
<td>Propped hydraulic fracture permeability (injection period)</td>
<td>100,000 Darcy</td>
<td>Parallel plate cubic law permeability assuming ~1mm open aperture</td>
</tr>
<tr>
<td>Un-propped hyd. fracture permeability (injection period)</td>
<td>10,000 Darcy</td>
<td>Parallel plate cubic law permeability assuming ~0.35mm open aperture</td>
</tr>
<tr>
<td>Propped hydraulic fracture permeability (shut-in and production period)</td>
<td>500 milliDarcy</td>
<td>History-matching + Horn River Fm. and other data (Table A.3)</td>
</tr>
<tr>
<td>Un-propped hyd. fracture permeability (shut-in and production period)</td>
<td>0.015 mD</td>
<td>History-matching + Horn River Fm. and other data (Table A.3)</td>
</tr>
</tbody>
</table>

**Capillary Pressure Parameters**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Source / Guidance</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shale matrix main imbibition curve van Genuchten parameters</td>
<td>p_c = 1.45 MPa</td>
<td>Fitted to Horn River Fm. and other shale formation data. See Edwards et al. (2017) supporting information</td>
</tr>
<tr>
<td></td>
<td>n = 1.7, m = 0.41</td>
<td></td>
</tr>
<tr>
<td>Hydraulic fracture main imbibition curve van Genuchten parameters</td>
<td>p_c = 0.2 MPa</td>
<td>Typical sandstone capillary pressure parameters</td>
</tr>
<tr>
<td></td>
<td>n = 2, m = 0.5</td>
<td></td>
</tr>
<tr>
<td>Water-rock contact angle (used for conversion of shale cap. pressure data)</td>
<td>45°</td>
<td>Horn River Fm. and other formation data (Table A.4)</td>
</tr>
<tr>
<td>Water-brine interfacial tension (used for conversion of shale cap. pressure data)</td>
<td>30 milliNewton/m</td>
<td>Hydraulic fracturing fluid data (Table A.2)</td>
</tr>
</tbody>
</table>
Appendix C

Model Parameters for Capillary Pressure Hysteresis Modeling
Table C.1 – Model parameters for the capillary pressure hysteresis simulations in Chapter 3. Parameters were selected based on typical shale gas well, hydraulic fracturing, and formation data. Indirect parameters (that were used to calculate direct parameters for the model) are shown in italics.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Source / Guidance</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Well, Hydraulic Fracture, and System Geometry Parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hydraulic fracture spacing</td>
<td>30 m</td>
<td>Well data. See Appendix A</td>
</tr>
<tr>
<td>Hydraulic fracture aperture</td>
<td>5 mm</td>
<td>Data and simulations. See Appendix A</td>
</tr>
<tr>
<td>Model domain fracture aperture</td>
<td>2.5 mm</td>
<td>Half of full fracture aperture</td>
</tr>
<tr>
<td>Model domain shale matrix length</td>
<td>15 m</td>
<td>Half distance between fractures</td>
</tr>
<tr>
<td>Propped hydraulic fracture porosity</td>
<td>60 %</td>
<td>Assume fractures partially filled by sand proppant</td>
</tr>
<tr>
<td><strong>Well Completion and Hydraulic Fracturing Process Parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hyd. fracturing fluid injection duration</td>
<td>3 hours</td>
<td>Hydraulic fracturing data. See Appendix A</td>
</tr>
<tr>
<td>Hydraulic fracturing bottom-hole injection pressure</td>
<td>70 MPa</td>
<td>Hydraulic fracturing data. See Appendix A</td>
</tr>
<tr>
<td>Shut-in period duration</td>
<td>1 day</td>
<td>Time for saturation rate of change to become minimal</td>
</tr>
<tr>
<td>Production period duration</td>
<td>1 day</td>
<td>Time for saturation rate of change to become minimal</td>
</tr>
<tr>
<td>Production bottom-hole pressure, $p_f$</td>
<td>15 MPa</td>
<td>Well data. See Appendix A</td>
</tr>
<tr>
<td><strong>Formation, Fracture and Fluid Parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Shale matrix porosity, $\phi$</td>
<td>6 %</td>
<td>Shale data. See Appendix A</td>
</tr>
<tr>
<td>Initial shale matrix water saturation</td>
<td>25 %</td>
<td>Shale data. See Appendix A</td>
</tr>
<tr>
<td>Initial hydraulic fracture water saturation</td>
<td>100 %</td>
<td>Implicitly assume fractures created and filled with water instantly</td>
</tr>
<tr>
<td>Shale matrix residual saturations (water / gas)</td>
<td>23 % / 40 %</td>
<td>Shale data. See Appendix A</td>
</tr>
<tr>
<td>Hydraulic fracture residual saturations water / gas</td>
<td>5 % / 5 %</td>
<td>Typical coarse, very permeable sand</td>
</tr>
<tr>
<td><strong>Gas composition</strong></td>
<td>90 % $CH_4$, 10 % $CO_2$</td>
<td>Shale data. See Appendix A</td>
</tr>
<tr>
<td><strong>Formation temperature</strong></td>
<td>130 °C</td>
<td>Shale data. See Appendix A</td>
</tr>
<tr>
<td>Water density, $\rho_w(p)$</td>
<td>$\sim$1,100 – 1,130 kg per m³</td>
<td>See graph in Edwards et al. (2017) supporting information</td>
</tr>
<tr>
<td>Water viscosity, $\mu_w$</td>
<td>0.7 centipoise (cP)</td>
<td>See graph in Edwards et al. (2017) supporting information</td>
</tr>
<tr>
<td>Parameter</td>
<td>Value</td>
<td>Source / Guidance</td>
</tr>
<tr>
<td>-------------------------------------------------------------------------</td>
<td>------------------------------</td>
<td>------------------------------------------------------------------------------------</td>
</tr>
<tr>
<td>Gas density, $\rho_g(p)$</td>
<td>$\sim 20 - 330$ kg per m$^3$</td>
<td>Defined by gas composition &amp; temp. See graph in Edwards et al. (2017) supporting information</td>
</tr>
<tr>
<td>Gas viscosity, $\mu_g(p)$</td>
<td>$\sim 0.015 - 0.04$ cP</td>
<td>Defined by gas composition &amp; temp. See graph in Edwards et al. (2017) supporting information</td>
</tr>
<tr>
<td>Initial formation gas pressure, $p_i$</td>
<td>36 MPa</td>
<td>Shale data. See Appendix A</td>
</tr>
<tr>
<td>Intrinsic shale matrix horizontal permeability</td>
<td>300 nanoDarcy (nD)</td>
<td>Shale data. See Appendix A</td>
</tr>
<tr>
<td>Shale matrix relative permeability (water / gas)</td>
<td>Corey $n = 4 / 2.2$</td>
<td>Shale data. See Appendix A</td>
</tr>
<tr>
<td>Hydraulic fracture relative permeability (water / gas)</td>
<td>Corey $n = 1 / 1$</td>
<td>Convention for fractures</td>
</tr>
<tr>
<td>Hydraulic fracture permeability</td>
<td>500 milliDarcy (mD)</td>
<td>Shale data. See Appendix A and B</td>
</tr>
<tr>
<td><strong>Capillary Pressure Parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Shale matrix main imbibition van Genuchten parameters</td>
<td>$p_{e} = 1.45$ MPa</td>
<td>Shale data. See Figure A.1</td>
</tr>
<tr>
<td></td>
<td>$n = 1.8$, $m = 0.44$</td>
<td></td>
</tr>
<tr>
<td>Shale matrix main drainage van Genuchten parameters</td>
<td>$p_{e} = 15.7$ MPa</td>
<td>Shale data. See Figure A.1</td>
</tr>
<tr>
<td></td>
<td>$n = 3.05$, $m = 0.67$</td>
<td></td>
</tr>
<tr>
<td>Hydraulic fracture main imbibition van Genuchten parameters</td>
<td>$p_{e} = 0.2$ MPa</td>
<td>Typical sandstone capillary pressure parameters</td>
</tr>
<tr>
<td></td>
<td>$n = 2$, $m = 0.5$</td>
<td></td>
</tr>
<tr>
<td>Hydraulic fracture main imbibition van Genuchten parameters</td>
<td>$p_{e} = 0.2$ MPa</td>
<td>Typical sandstone capillary pressure parameters</td>
</tr>
<tr>
<td></td>
<td>$n = 2$, $m = 0.5$</td>
<td></td>
</tr>
<tr>
<td>Water-rock contact angle (used for conversion of shale cap. pressure data)</td>
<td>45°</td>
<td>Shale data. See Appendix A</td>
</tr>
<tr>
<td>Water-brine interfacial tension (used for conversion of shale cap. pressure data)</td>
<td>30 milliNewton/m</td>
<td>Hydraulic fracturing fluid data. See Appendix A</td>
</tr>
<tr>
<td>Killough model hysteresis fitting parameter (for simulations in Figs. 3.6 and 3.7)</td>
<td>0.02</td>
<td>Shale capillary pressure data. See Figure A.1</td>
</tr>
</tbody>
</table>
Appendix D
Model Parameters for Carbon Dioxide Injection Modeling
The reservoir and well parameters history-matched to natural gas production data and used for the CO₂ injection simulations are shown in Tables D.1-D.3. Additional parameters required for the CO₂ injection simulations that were not matched in the single-component history-matching are also included (gas dispersion coefficient and CO₂ sorption parameters). Two parameter groups govern the production or injection behavior of the model: the interference time and total gas in place. The interference times associated with each scenario are shown in Table D.3.

**Table D.1** – Reservoir parameters for both plays that are consistent in all parameter scenarios.

<table>
<thead>
<tr>
<th>Shale Play</th>
<th>Marcellus</th>
<th>Barnett</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas Saturation (%)</td>
<td>75</td>
<td>75</td>
</tr>
<tr>
<td>CH₄ Langmuir K (MPa⁻¹)</td>
<td>0.17</td>
<td>0.17</td>
</tr>
<tr>
<td>CO₂ Langmuir K (MPa⁻¹)</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>Reservoir Temperature (°C)</td>
<td>60</td>
<td>80</td>
</tr>
<tr>
<td>Gas Dispersion Coefficient (m²/s)</td>
<td>1×10⁻⁹</td>
<td>1×10⁻⁹</td>
</tr>
<tr>
<td>Fracture Face Pressure (production) / Initial Pressure (injection) (MPa)</td>
<td>3.5</td>
<td>3.5</td>
</tr>
<tr>
<td>Adsorbed Phase Density, ρₐ (kg/m³)</td>
<td>1000</td>
<td>1000</td>
</tr>
</tbody>
</table>

**Table D.2** – Reservoir parameters for each region and each history-matching parameter scenario. Shaded boxes show parameters that were varied in the history-match to fit the model to the observations, while unshaded boxes show parameters selected based on well data or literature.

<table>
<thead>
<tr>
<th>Shale Play &amp; Scenario</th>
<th>Porosity (%)</th>
<th>Reservoir Pressure (production) / Max. Pressure (injection) (MPa)</th>
<th>Effective Permeability (nanoDarcy)</th>
<th>Max. CH₄ Adsorption (kg/m³)</th>
<th>Max. CO₂ Adsorption (kg/m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Marcellus SW Avg.</td>
<td>6</td>
<td>35</td>
<td>25</td>
<td>6.5</td>
<td>55</td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>35</td>
<td>25</td>
<td>8.5</td>
<td>70</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>33</td>
<td>30</td>
<td>4.5</td>
<td>40</td>
</tr>
<tr>
<td>Marcellus NE Avg.</td>
<td>6</td>
<td>35</td>
<td>20</td>
<td>6.5</td>
<td>55</td>
</tr>
<tr>
<td>Shale Play &amp; Scenario</td>
<td>Porosity (%)</td>
<td>Reservoir Pressure (production) / Max. Pressure (injection) (MPa)</td>
<td>Effective Permeability (nanoDarcy)</td>
<td>Max. CH₄ Adsorption (kg/m³)</td>
<td>Max. CO₂ Adsorption (kg/m³)</td>
</tr>
<tr>
<td>-----------------------</td>
<td>--------------</td>
<td>-------------------------------------------------</td>
<td>---------------------------------</td>
<td>-----------------</td>
<td>-----------------</td>
</tr>
<tr>
<td>High</td>
<td>6</td>
<td>35</td>
<td>20</td>
<td>8.5</td>
<td>70</td>
</tr>
<tr>
<td>Low</td>
<td>5</td>
<td>33</td>
<td>25</td>
<td>4.5</td>
<td>40</td>
</tr>
<tr>
<td>Barnett Avg.</td>
<td>6</td>
<td>25</td>
<td>45</td>
<td>5</td>
<td>45</td>
</tr>
<tr>
<td>High</td>
<td>6</td>
<td>25</td>
<td>45</td>
<td>7</td>
<td>60</td>
</tr>
<tr>
<td>Low</td>
<td>5</td>
<td>24</td>
<td>45</td>
<td>3</td>
<td>30</td>
</tr>
</tbody>
</table>

**Table D.3** – Well parameters for each region and each history-matching parameter scenario. Shaded boxes show parameters that were varied in the history-match to fit the model to the observations, while unshaded boxes show parameters selected based on well data or literature. The interference time is calculated by Eq. 4.11 using the selected parameters for each scenario.

<table>
<thead>
<tr>
<th>Shale Play &amp; Scenario</th>
<th>Lateral Length (m)</th>
<th>Fracture Height, V (m)</th>
<th>Fracture Width, H (m)</th>
<th>Fracture Spacing, 2d (m)</th>
<th>Interference Time, τ (years)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Marcellus SW Avg.</td>
<td>1556</td>
<td>32</td>
<td>200</td>
<td>30</td>
<td>6.6</td>
</tr>
<tr>
<td>High</td>
<td>1644</td>
<td>32</td>
<td>200</td>
<td>24</td>
<td>4.2</td>
</tr>
<tr>
<td>Low</td>
<td>1556</td>
<td>32</td>
<td>200</td>
<td>30</td>
<td>5.0</td>
</tr>
<tr>
<td>Marcellus NE Avg.</td>
<td>1675</td>
<td>50</td>
<td>200</td>
<td>30</td>
<td>6.8</td>
</tr>
<tr>
<td>High</td>
<td>1759</td>
<td>50</td>
<td>200</td>
<td>28</td>
<td>5.9</td>
</tr>
<tr>
<td>Low</td>
<td>1675</td>
<td>50</td>
<td>200</td>
<td>30</td>
<td>5.0</td>
</tr>
<tr>
<td>Barnett Avg.</td>
<td>872</td>
<td>30</td>
<td>200</td>
<td>30</td>
<td>5.8</td>
</tr>
<tr>
<td>High</td>
<td>1000</td>
<td>30</td>
<td>200</td>
<td>30</td>
<td>5.8</td>
</tr>
<tr>
<td>Low</td>
<td>872</td>
<td>30</td>
<td>200</td>
<td>30</td>
<td>5.0</td>
</tr>
</tbody>
</table>