



Experimental insights into geochemical changes in hydraulically fractured Marcellus Shale[☆]



Virginia Marcon^{a, b, 1}, Craig Joseph^a, Kimberly E. Carter^{a, 2}, Sheila W. Hedges^c,
Christina L. Lopano^d, George D. Guthrie^e, J. Alexandra Hakala^{d, *}

^a Oak Ridge Institute for Science and Education Fellow, Office of Research and Development, National Energy Technology Laboratory, Pittsburgh, PA, USA

^b Department of Geology and Geophysics, University of Wyoming, Laramie, WY, USA

^c Office of Research and Development, National Energy Technology Laboratory, Pittsburgh, PA, USA

^d Research and Innovation Center, National Energy Technology Laboratory, Pittsburgh, PA, USA

^e Earth and Environmental Sciences Division, Los Alamos National Laboratory, Los Alamos, NM, USA

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ABSTRACT

Hydraulic fracturing applied to organic-rich shales has significantly increased the recoverable volume of methane available for U.S. energy consumption. Fluid-shale reactions in the reservoir may affect long-term reservoir productivity and waste management needs through changes to fracture mineral composition and produced fluid chemical composition. We performed laboratory experiments with Marcellus Shale and lab-generated hydraulic fracturing fluid at elevated pressures and temperatures to evaluate mineral reactions and the release of trace elements into solution. Results from the experiment containing fracturing chemicals show evidence for clay and carbonate dissolution, secondary clay and anhydrite precipitation, and early-stage (24–48 h) fluid enrichment of certain elements followed by depletion in later stages (i.e. Al, Cd, Co, Cr, Cu, Ni, Sc, Zn). Other elements such as As, Fe, Mn, Sr, and Y increased in concentration and remained elevated throughout the duration of the experiment with fracturing fluid. Geochemical modeling of experimental fluid data indicates primary clay dissolution, and secondary formation of smectites and barite, after reaction with fracturing fluid. Changes in aqueous organic composition were observed, indicating organic additives may be chemically transformed or sequestered by the formation after hydraulic fracturing. The NaCl concentrations in our fluids are similar to measured concentrations in Marcellus Shale produced waters, showing that these experiments are representative of reservoir fluid chemistries and can provide insight on geochemical reactions that occur in the field. These results can be applied towards evaluating the evolution of hydraulically-fractured reservoirs, and towards understanding geochemical processes that control the composition of produced water from unconventional shales.

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1. Introduction

Unconventional shale gas exploration and hydraulic fracturing of geologic formations for oil and gas has boomed over the past decade, creating 1.4 million new jobs (O'Neil et al., 2016), contributing billions to gross domestic production (GDP) (Institute for 21st Century Energy, 2013; O'Neil et al., 2016), yet generating numerous

environmental concerns (Bibby et al., 2013; Brantley et al., 2014; Myers, 2012; Vidic et al., 2013). The Marcellus Shale (Appalachian Basin, USA) produces the largest volume of tight shale gas in the United States (U.S. Energy Information Administration, 2014). During production of a single Marcellus gas well, one to seven million gallons of water (Jiang et al., 2014), along with specific chemical additives (Gallegos and Varela, 2015), are injected. Anywhere from 5 to 85% of the injected water volume is recovered (typically 30–50%) during flowback and gas production (Gregory et al., 2011; Nicot and Scanlon, 2012; Stringfellow et al., 2014) and often at salinities up to 300% greater than the initial fluid injected (Bibby et al., 2013; Gregory et al., 2011; Vidic et al., 2013). In some Marcellus wells, chloride concentrations were observed to

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* Corresponding author.

E-mail address: Alexandra.Hakala@netl.doe.gov (J.A. Hakala).

¹ Current address: Pennsylvania State University, State College, PA, USA.

² Current address: University of Tennessee Knoxville, Knoxville, TN, USA.

increase 6-fold (Blanch et al., 2009; Cluff et al., 2014). This salinity increase may occur in part from geochemical reactions between hydraulic fracturing fluids and the reservoir and from mixing between injected and existing reservoir fluids (Blanch et al., 2009; Capo et al., 2014; Cluff et al., 2014). As the demands for energy and unconventional resource exploration grow (U.S. Energy Information Administration, 2014), an understanding of the geochemical reactions that occur during the introduction of fracturing fluid is paramount to understanding how the chemical evolution of the reservoir can affect the composition of produced water and the long-term productivity of the fractured shale.

Knowledge of geochemical processes that affect shale formation integrity and the composition of produced waters during hydraulic fracturing is required to improve productivity of hydraulically-fractured shale reservoirs while minimizing environmental impacts. Geochemical changes within the fractured shale can affect both the chemical and physical composition of waters produced during the stimulation and gas production phases (Kekacs et al., 2015b) as well as physical properties of the shale formation through dissolution and precipitation reactions (Murali Mohan et al., 2013; Vidic et al., 2013). The changes in produced water chemistry provide direct observations on the chemical evolution of the reservoir, which can be used as input for optimizing reservoir productivity and guiding development of strategies for recycling, management, and disposal of produced waters (Barbot et al., 2013). Studying water-rock interactions at the laboratory scale provides the unique opportunity to monitor changes in both the aqueous chemistry as well as chemical and physical changes to the shale after fracturing fluid reaction.

Limited publically-available data currently exist related to the geochemical reactions and physical changes that occur during hydraulic fracturing of Marcellus Shale. Previously-reported experimental studies conducted at reservoir pressures and temperatures focused on shales from Denmark and Germany and showed that the shale mineral composition exerts a significant influence on the system buffering capacity in the presence of hydraulic fracturing fluids (Wilke et al., 2015). Recent detailed geochemical studies performed under ambient pressures and temperatures report influences of acid-base and oxidation-reduction chemistry on fluids interacting with Eagle Ford and Bakken shales (Wang et al., 2015, 2016). A recent microscopy study reported dissolution of calcite and precipitation of gypsum on polished surfaces of Marcellus Shale in the presence of high-salinity fluids at reservoir pressures and temperatures (Dieterich et al., 2016).

In this study we focus on reactions between Marcellus Shale and hydraulic fracturing fluids under reservoir pressures and temperatures, and focus on geochemical changes in both the fluid and solid phases. The primary goals of this study are to 1) evaluate the effects of reactions between Marcellus Shale and fracturing fluids on produced water chemistry and reservoir mineralogy at elevated

temperature and pressure and 2) evaluate the release and evolution of major and minor cations, anions, and trace elements extracted from Marcellus Shale during interaction with hydraulic fracturing fluid. Additionally, we present observations on changes to the aqueous organic composition under experimental conditions that simulate downhole reservoir conditions.

2. Materials and methods

2.1. Experimental design and apparatus

Two parallel experiments were conducted to investigate the geochemical changes due to fracturing fluid-shale interactions: a baseline experiment containing synthetic brine and Marcellus Shale solids (BRINE-1) and a second experiment containing a 7:3 mixture of synthetic brine and fracturing fluid and Marcellus Shale solids (BRINE-2; Tables 1 and 2). The BRINE-1 experiment was designed to reflect formation conditions prior to injection, and absent of fracturing fluid chemical additives. The BRINE-2 experiment was designed to probe the impact of fracturing chemical additives on reservoir geochemical interactions. Both experiments were focused on evaluating fluid-rock interactions while removing influences of regional brine mixing that are observed in field-collected produced waters from Marcellus Shale (Barbot et al., 2013).

The fluid-rock interactions simulated with this experimental design occur in the fractures, away from the well bore, after the well completion process, and during the shut-in phase prior to production. Depending on the operator, the shut-in time frame can last from a few days to weeks (Vidic et al., 2013); the experiments in this study were designed to evaluate a two-week shut-in phase. The initial shut-in phase is the time frame during which the majority of reactions between fracturing fluids and shale are hypothesized to occur, based on prior investigations related to reactions between injected fluids and minerals in deep geologic reservoirs (Chopping and Kaszuba, 2012; Marcon and Kaszuba, 2015).

The experimental apparatus is designed such that fluid can be sampled while the experiment remains at experimental conditions (Fig. 1), and at termination of the experiment, the reacted solids can be recovered and analyzed. Hydrothermal experiments were performed using rocking autoclave reactors, flexible gold reaction cells (Seyfried et al., 1987), and titanium capillary exit tubes with experimental methods developed in previous studies (Kaszuba et al., 2003, 2005; Seyfried et al., 1987). Experimental vessels containing a 20:1 fluid-rock ratio were constructed one week prior to raising the system to the experimental conditions of 275 bars (4000psi) and 130 °C (Table 1). The experimental pressure was designed to represent field borehole conditions, and the temperature was elevated to enhance kinetics while remaining in the same mineral thermodynamic reaction realm modeled at 75 °C (actual

Table 1
Experimental parameters.

Experiment	BRINE-1	BRINE-2
Description	Marcellus + Formation Fluid	Marcellus + Formation Fluid + Fracturing Fluid
Fluid	100% Synthetic Brine	70% Synthetic Brine 30% Fracturing Fluid
Initial pH	7.59 ± 0.1	1.83 ± 0.1
Temperature (°C)	132.6 ± 0.6	129.1 ± 1.2
Pressure (MPa)	27.4 ± 1.8	28.0 ± 0.08
Rock Mass ^a	11.17	11.20
Initial Water:Rock Ratio	20.4: 1	18.6: 1
Total Reaction Time (hours)	378.92	356.53

Dissolved oxygen was removed from the formation brine by sparging with nitrogen.

Fracturing Fluid was not sparged of oxygen and assumed to be saturated (see section 2.5).

^a Rock mass input into reaction cell.

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