Journal of Environmental Management 214 (2018) 370–378

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Research article

Effect of brine salinity and guar gum on the transport of barium through dolomite rocks: Implications for unconventional oil and gas wastewater disposal

Pouyan Ebrahimi, Javier Vilcáez*

Boone Pickens School of Geology, Oklahoma State University, Stillwater, OK 74078, USA

ARTICLE INFO

Article history:
Received 3 August 2017
Received in revised form
22 December 2017
Accepted 2 March 2018
Available online 13 March 2018

Keywords:
Unconventional oil and gas wastewater
Barium
Guar gum
Dolomite
Heavy metals transport

ABSTRACT

This research aimed to elucidate the effect of brine salinity and guar gum on the sorption and transport of Ba in dolomite rocks collected from the Arbuckle formation in Oklahoma, USA. Guar gum represents the most important organic additive used in viscosified fracturing fluids, and Ba constitutes the most common and abundant heavy metal found in unconventional oil and gas (UOG) wastewater. Batch experiments conducted using powdered dolomite rocks (500–600 μm particle size) revealed that at brine salinities of UOG wastewater, chloro-complexation reactions between Ba and Cl ions and pH changes that results from dolomite dissolution are the controlling factors of Ba sorption on dolomite. Competition of Ba with common cations (Ca and Mg) for hydration sites of dolomite, plays a secondary role. Core-flooding experiments conducted to analyze the transport of Ba through natural and synthetic dolomite core plugs are in agreement with the batch sorption experimental results. The transport of Ba through dolomite rocks, increases with increasing brine salinity (0–180,000 mg-NaCl/L). The presence guar gum (50–500 mg/L) does not affect the transport of Ba through dolomite rocks of high flow properties (25–29.6% porosity, 9.6–13.7 mD permeability). However, core-flooding experiments conducted using tight dolomite rocks (6.5–8.6% porosity, 0.06–0.3 mD permeability), revealed that guar gum can retard the transport of Ba by clogging high permeability/porosity regions of tight dolomite rocks. The mechanism of Ba sorption on dolomite can be represented by a sorption model that accounts for both surface complexation reactions on three distinct hydration sites (>$CaOH^o$, >$MgOH^o$, and >CO_3H^o), and the kinetic dissolution of dolomite. These results are important in understanding and predicting the fate of Ba present in UOG wastewater disposed into deep dolomite saline aquifers.

1. Introduction

Recent technological advances, combining horizontal drilling with hydraulic fracturing (HF) have led to a boom in the development of unconventional oil and gas (UOG) resources (including shale gas, coal bed methane, and tight oil) from low permeability formations (Arthur et al., 2009; Engle et al., 2014). HF involves the high-pressure injection of a fluid containing primarily water, proppant, and multiple organic additives such as solvents, viscosifiers, biocides, scale inhibitors, friction reduces, and surfactants (Ferrer and Thurman, 2015; Stringfellow et al., 2014). Typically, a large fraction of the injected HF fluid infiltrates to the geological formation, while 5–60% of the HF fluid flows back to the surface when pressure is released (Carter et al.). The returning UOG wastewater is characterized by high total dissolved solids (TDS), naturally occurring radioactive materials (NORMs) (Rowan et al., 2011), and variable concentrations of organic compounds (Strong et al., 2013). TDS includes heavy metals (e.g., Ba, As, Se and Sr) whose concentrations are in many cases hundreds of time above the USA drinking water standards (Engle and Rowan, 2014; Haluszczak et al., 2013). Organic compounds in UOG wastewaters could be sourced from the injected HF fluids (Akob et al., 2015; Cluff et al., 2014; Orem et al., 2014). Whereas, high concentrations of heavy metals in wastewater might be due to the primary dissolution of fracture-filling and/or pore-filling evaporate minerals, or due to the direct communication of basinal brine into a more permeable hydraulic fracture zone (Blauch et al., 2009; Renock et al., 2016).

* Corresponding author.
E-mail address: vilcaez@okstate.edu (J. Vilcáez).

https://doi.org/10.1016/j.jenvman.2018.03.008
0301-4797/

To prevent the contamination of underground sources of drinking water (USDW), UOG wastewater is frequently disposed into deep saline aquifers (Lutz et al., 2013). However, the common occurrence of permeable pathways (e.g., induced and natural fractures) overlying deep saline aquifers is raising serious concerns regarding the possibility of USDW contamination with UOG wastewater (EPA, 2012, 2016).

In general, the transport of heavy metals and organic compounds is to a large extent controlled by sorption (surface complexation) reactions (Appelo and Postma, 2005; Dokhani et al., 2016) of which very little is currently known for deep saline aquifers. Published work on the transport of heavy metals is constrained to shallow aquifers. Fundamental differences between shallow and deep saline aquifers arise from the different reactivity and geochemical composition of the mineral phase, different flow (porosity/permeability) properties and different water phase composition. An understanding of the variables controlling the sorption and transport of heavy metals in deep saline aquifers is fundamental to assess the feasibility of USDW contamination by heavy metals due to the possible upward migration of UOG wastewater from deep saline aquifers.

Among many physical and chemical factors that can affect the sorption and thus the transport of heavy metals in deep saline aquifers, organic additives such as viscosifiers remaining in UOG wastewater, and brine salinity (TDS > 10,000 mg/L) are hypothesized to play an important role in the transport of heavy metals through porous media of saline aquifers injected with UOG wastewater. This hypothesis is based on 1) studies showing that viscosifiers such as guar gum, which are added to allow for better proppant suspension and transport into developed fractures (Elsner and Hoelzer, 2016), can change the sorption capacity of porous media due to its sorption on minerals such as quartz (Ma and Pawlik, 2005, 2006), and 2) studies showing that brine salinity increases the mobility of heavy metals (Cu, Cd, Zn, Pb and Zn) through soil materials (Acosta et al., 2011; Zhao et al., 2013). The combined effect of viscosifiers and brine salinity on the transport of heavy metals at conditions relevant to UOG waste water disposal in deep saline aquifers has not been studied before.

Specifically, this research aims to elucidate the effect guar gum and brine salinity on the transport of Ba through dolomite rocks collected from the Arbuckle formation where wastewater from oil production is disposed in Oklahoma, USA. Guar gum represents the most common organic additive used in viscosified fracturing fluids. Contrary to expectations, a complete biodegradation of guar gum contained in UOG wastewater is difficult to achieve due to the high salinity of UOG wastewater that greatly decreases the activity of microbes (Lester et al., 2013). For the sake of simplicity, among the many heavy metals found in UOG wastewater, the focused of this research is on the transport of Ba, which constitutes the most common and abundant heavy metal found in UOG wastewater (EPA, 2016; Rozell and Reaven, 2012).

Because of the discrete nature of fractures which can facilitate the upward migration of UOG wastewater, it is apparent that the transport of heavy metals through porous media connecting natural fractures plays an important role. Hence, we first aim to understand the factors controlling the transport of heavy metals through porous dolomite rocks. With this aim, we stablished batch experiments to understand the effect of brine salinity and guar gum on the sorption of Ba on dolomite, and core-flooding experiments to analyze the effect of brine salinity and guar gum on the transport of Ba through natural and synthetic dolomite core plugs of homogeneous flow (porosity/permeability) properties.

2. Materials

2.1. Dolomite rock samples

Dolomite rocks used in this research were collected from outcrops of the Arbuckle formation where wastewater from oil and gas production is disposed in Oklahoma (Gadhamshetty et al., 2015). Fig. 1 shows representative X-rays diffraction (XRD), scanning electron microscopy (SEM), and micro-CT scan analysis conducted on collected rocks. Analyses revealed that Arbuckle dolomite rocks are composed of 97% dolomite and the rest is quartz and calcite (Fig. 1A), grain size is 0.125–0.5 mm (red polygons in Fig. 1B), diameter of pores (yellow polygons in Fig. 1B) is < 0.5 mm, and that pores are poorly connected to each other (Fig. 1C). Collected rocks were used to prepare three types of rocks: powdered rocks of uniform size, natural core plugs, and synthetic core plugs of uniform flow properties. Powdered rocks were used to conduct batch experiments, whereas synthetic and natural core plugs were used to conduct core-flooding experiments.

2.1.1. Powdered rocks

Powdered dolomite were sieved to obtain 150-212 and 500–600 μm particles sizes. Dust from the sieved particles was removed by using an ultrasonic bath with deionized water.

2.1.2. Uniform synthetic core plugs

Natural dolomite rocks are tight heterogeneous porous media. In order to be able to conduct reproducible core-flooding experiments, homogeneous synthetic core plugs of high flow (porosity and permeability) properties were prepared using a uniaxial compaction apparatus (Carver Laboratory Presses, Model 4387). The procedure consisted in mixing 90 g of powdered dolomite rocks of known particle size and 9 g of deionized water (10% of dolomite weight). The resulting aggregate is poured into a stainless steel mold and compressed at 34473.79 kPa for one hour using the uniaxial compaction apparatus (Fig. 2).

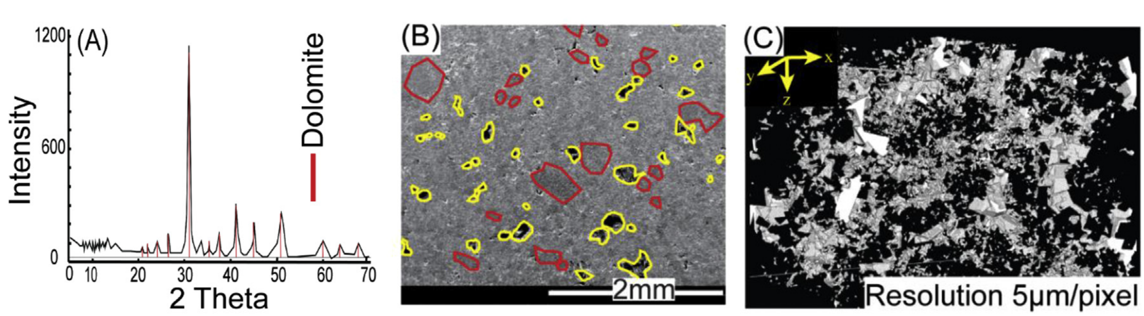

Fig. 1. Analysis of dolomite rocks collected from outcrops of the Arbuckle formation. (A) X-ray diffraction (XRD) analysis, (B) Scanning electron microscope (SEM) analysis, and (C) Micro-CT scan analysis.