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Transport of barium in fractured dolomite and sandstone saline aquifers

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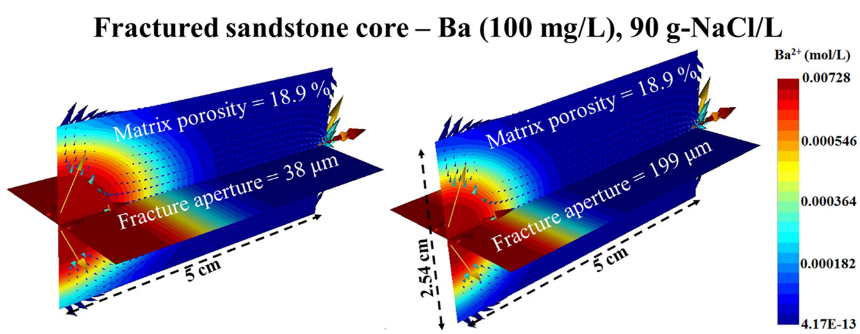
HIGHLIGHTS

- Similar Ba transport rates are possible in intact and fractured dolomites.
- Ba transport can be faster in intact than in fractured sandstones.
- Salinity increases Ba transport through the matrix pores bordering fractures.
- Matrix porosity and fracture aperture size play an important role.

GRAPHICAL ABSTRACT



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ABSTRACT

To understand the advective-dispersive transport of Ba in fractured dolomite and sandstone saline aquifers, we conducted core-flooding experiments and reactive transport simulations. We used intact and synthetic fractured dolomite and sandstone cores collected from formations where hydraulic fracturing (HF) wastewater is disposed in Oklahoma, USA. The core-flooding experiments were conducted using saline water containing typical concentrations of NaCl (90 g/L), Ca (5 g/L), Mg (1 g/L), and Ba (100 mg/L) in HF wastewaters. At typical concentrations of NaCl, Ca, and Mg in HF wastewater, our experimental results show similar Ba transport rates in both intact and fractured dolomites but faster Ba transport rates in intact than in fractured sandstones. We found a match between measured and simulated breakthrough curves of Ba in intact and fractured sandstones. This supports the hypothesis that the inhibitory effect of salinity on Ba sorption increases Ba transport through matrix pores bordering the fracture while reducing its transport through the fracture. This is reflected by a reduction of the overall rate of Ba transport through fractured dolomites and sandstones. We found that the effect of salinity in retarding Ba transport through fractured dolomites and sandstones increases with increased matrix porosity and/or fracture aperture size. We implemented the multiple interacting continua (MINC) method developed for modeling fluid flow in fractured porous media to successfully capture the effect of salinity, matrix porosity and fracture aperture size on Ba transport in fractured sandstones. The measured and simulated results have significant implications on efforts of field-scale simulations of Ba transport in dolomite and sandstone saline aquifers where HF wastewater is disposed.

1. Introduction

Hydraulic fracturing (HF) wastewater, which is disposed into geological formations, reused in other hydraulic fracturing operations, or disposed above ground after treatment (EPA., 2016) is characterized by high levels of total dissolved salts (mostly NaCl), heavy metals

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(i.e., Ba, Sr), radioactive materials (i.e., U, Ra) and a number of organic compounds (Veil et al., 2004). There is a continuous increase of volumes of HF wastewater disposed into geological formations (Kondash et al., 2017; Murray, 2013). This has created public debate about the possibility of contamination of underground sources of drinking water (USDW) with contaminants present in HF wastewater disposed into deep saline aquifers. The upward migration of HF wastewater water from deep saline aquifers through overlaying bedrocks of low permeability is generally considered to be a slow process. Under preexisting pressure gradients and bedrock permeabilities, the upward migration of HF wastewater would take >10^6 years (Flewelling and Sharma, 2014). However, the speed of upward migration of HF wastewater could dramatically increase if the disposed HF wastewater reaches conductive fault zones and wells with integrity (casing and/or cementing) failures (Jackson, 2014).

To our knowledge, very few studies have been conducted to understand the variables controlling the mobility and transport of contaminants (i.e., heavy metals and radioactive materials) present in HF wastewater injected into deep saline aquifers. Understanding these variables is of paramount importance to assess the real risk of contamination of USDW by the migration of HF wastewater through conductive fault zones and/or neighboring wells with integrity failures. Recent studies suggest that the presence of heavy metals and organic compounds (methanol and ethanol) exceeding the Environmental Protection Agency (EPA)'s drinking water maximum contaminant limits (MCLs) in private drinking water wells might be related to faulty drilling equipment and the well casings of nearby active gas wells (Fontenot et al., 2013; Llewellyn et al., 2015). Available information on the variables controlling the transport of contaminants in shallow aquifer systems cannot be used to predict the mobility and transport of contaminants in deep saline aquifers. Contrary to shallow aquifers which are characterized by close to ambient temperature, low salinity, and relatively low concentrations of organic matter conditions, deep saline aquifers injected with HF wastewater are characterized by thermophilic temperatures, high salinity, and relatively high availability of organic matter derived from the use of organic fracturing additives (i.e., guar gum, polyacrylamide, and glutaraldehyde) for hydraulic fracturing.

Ba constitutes the most common and abundant heavy metal present in HF wastewater (Neff et al., 2011), and dolomite and sandstone aquifers constitute two of the most common saline aquifers where produced water is disposed in Oklahoma, USA (Murray, 2014, 2015). In previous studies we have elucidated the effect of physicochemical factors, including brine salinity, guar gum, competition of cations (Ca and Mg), and temperature on the mobility and transport of Ba in intact dolomites and sandstones. We found that the effect of temperature is secondary to the effect of salinity on Ba transport through porous dolomites and sandstones, and that guar gum (50–500 mg/L) only affects Ba transport through tight rocks (0.06–0.3 mD permeability) (Ebrahimi and Vilcáez, 2018a, 2018b). Therefore, this study does not include the effect of temperature and guar gum on Ba transport through fractured dolomites and sandstones.

The aim of this study is to elucidate the impact of fractures on the transport of Ba in dolomites and sandstones. This is important because of the common occurrence of fractures in dolomite and sandstone saline aquifers which can facilitate the transport of Ba from injection to conductive fault zones and/or neighboring wells with integrity failures. Also, the elucidation of Ba transport trough fractured rocks, might shed some light on induced seismic events. HF wastewater migration through fractured rocks could change the physical properties of fractures such as roughness (Keranen et al., 2014), triggering seismic events in highly stressed zones.

Reactive and non-reactive solutes transport in intact (Vilcáez et al., 2013; Vilcáez et al., 2008) and fractured (Neretnieks, 1993; Royer et al., 2002; Sharma et al., 2016) porous media has been widely studied numerically and experimentally at both the lab and field-scales. In most studies, solutes have been assumed to be transported only by dispersion through the matrix pores, and by advection-dispersion through the aperture of fractures. However, relatively high pressure gradients resulting from the injection of HF wastewater might result in the transport of Ba by advection-dispersion through the matrix pores bordering fractures (Kumar, 2008; Rasmuson and Neretnieks, 1981). This can play a significant role in the mechanical and chemical retardation of Ba in fractured dolomite and sandstone aquifers.

Mechanical retardation results from the spreading the solutes along the fractures (Venture, 2004). Chemical retardation, on the other hand, results from the transfer of a solute from the liquid phase to the solid phase due to multiple chemical reactions between the transporting fluid and walls of fractures and matrix pores. Among many possible water-rock chemical reactions, sorption reactions are the main reactions that retard the transport of heavy metals in subsurface.

To understand the mechanical and chemical retardation of Ba transport in fractured dolomites and sandstones, we conducted further core-flooding experiments using intact and synthetic fractured dolomite and sandstone cores. The core-flooding experiments were conducted using saline water composed of NaCl, Ca, Mg, Ba, and deionized water. To make a mechanistic interpretation of the experimental results and provide the means to conduct field-scale simulations of Ba transport in dolomite and sandstone aquifers, we conducted reactive transport simulations of Ba transport in intact and fractured dolomite and sandstone cores using the multiple interacting continua (MINC) method available in TOUGHREACT v1.2 reactive transport simulator (Xu et al., 2006).

2. Materials and methods

2.1. Fractured dolomite and sandstone rocks

Sandstone and dolomite lithologies are representative of sedimentary rocks where HF wastewater and petroleum produced water is disposed in Oklahoma, USA. The sandstone (Fig. 1A & B) was collected from a depth of <300 m where the Raton Formation is intersected in Las Animas County, Colorado. The dolomite (Fig. 1C & D) was collected from the Arbuckle Group, McDonald County, Missouri. Both the sandstone and dolomite have fine texture (Fig. 1E & F), but the effective porosity (18.1–19.9%) and permeability (1.1×10^{-15}–6.3×10^{-14} m^2) of sandstone is higher than the effective porosity (5.3–6.2%) and permeability (1.0×10^{-16}–3.0×10^{-16} m^2) of dolomite. The dolomite contained isolated vuggy pores (<3 mm), hence they did not affect the rock permeability. The photomicrographs in Fig. 1G & H shows the texture of the conducted on the collected sandstone and dolomite. The grain sizes of the collected sandstone and dolomite are respectively ~100–200 μm and ~50–150 μm. X-ray diffraction (XRD) analysis indicted that the sandstone is mainly composed of quartz (>98%) with <2% of clay minerals, and that dolomite is mainly composed of dolomite minerals (>99%) with <1% of carbonates and silt (Ebrahimi and Vilcáez, 2018b).

Cylindrical rock cores with a diameter of 2.54 cm and length of 5 cm were plugged from both the collected sandstone and dolomite (Fig. 1A & C). Synthetic fractured dolomites and sandstones were created by cutting the rock cores into two equal semi-cylinder segments along the longitudinal axis (Fig. 1E & F). To emulate real fractures, the smooth surface of the dolomite half-cylinder segments was uniformly roughened by a half round wood rasp file. After cutting the rock cores and abrading them, there was no distinct differences between the two rock synthetic fracture conjugate surfaces. The two conjugate half-cylinders were hold together by using a heat-shrink rubber sleeve (Fig. 1B & D). Using a digital microscope (20×–800× magnification), the fracture aperture size of the sandstone and dolomite were measured to be 280–400 μm and 150–300 μm, respectively.