



Effect of ionic strength on barium transport in porous media

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ABSTRACT

Hydraulic fracturing (or fracking) is a well stimulation technique used to extract resources from a low permeability formation. Currently, the most common application of fracking is for the extraction of oil and gas from shale. During the operation, a large volume of brine, rich in hazardous chemicals, is produced. Spills of brine from wells or pits might negatively impact underground water resources and, in particular, one of the major concerns is the migration of radionuclides, such as radium (Ra^{2+}), into the shallow subsurface. However, the transport behaviour of Ra^{2+} through a reactive porous medium under conditions typical of a brine, i.e., high salinity, is not well understood, yet. Here, a study on the transport behaviour of barium (Ba^{2+} , congener of radium) through a porous medium containing a common mineral such as goethite ($\text{FeO}(\text{OH})$) is presented. Batch and column flood tests were carried out at conditions resembling the produced brine, i.e., large values of ionic strength (I), namely, 1 to 3 mol/kg. The measurements were described with the triple layer surface complexation model coupled with the Pitzer activity coefficient method and a reactive transport model, in the case of the transport tests. The experimental results show that the adsorption of Ba^{2+} onto $\text{FeO}(\text{OH})$ increases with pH but decreases with I and it becomes negligible at the brine conditions. Moreover, even if isotherms show adsorption at large I , at the same conditions during transport, Ba^{2+} travels without retardation through the $\text{FeO}(\text{OH})$ porous medium. The triple layer model agrees very well with all batch data but it does not describe well the transport tests in all cases. In particular, the model cannot match the pH measurements at large I values. This suggests that the chemical reactions at the solid-liquid interface do not capture the mechanism of Ba^{2+} adsorption onto $\text{FeO}(\text{OH})$ at large salinity. Finally, this study suggests that barium, and potentially its congeners, namely, radium, calcium, magnesium, and strontium, may travel at the average flow velocity through a soil where the dominant reactive mineral is goethite.

1. Introduction

Hydraulic fracturing (or fracking) is a well stimulation technique used to maximize the extraction of underground resources, such as oil, natural gas, geothermal energy, and water, from low permeability formations. In recent years, fracking technology has become widespread, particularly for the production of oil and gas from unconventional shale formations. For the extraction, wells may be drilled to a depth of 1,500–4,500 m and in lateral sections may extend 300–2000 m away from the well. The injected fluids are commonly made up of water, sand, and chemical additives (such as surfactants, chelating agents, and biocides) (FracFocus, 2017), which are pumped at high pressure into the geologic formations. Recovered fracturing fluids are referred to as flowback and produced water. Overall, drilling and fracking require approximately 2–4 million gallons of water per well (i.e., 8,000–15,000 m^3 per year) (Scanlon et al., 2017). However, only a fraction of 10–80% of the injected fluid returns to the surface again at

relatively high flow rates and relatively high in total dissolved solids (TDS) (Bi et al., 2016). After few weeks, the flow of water decreases significantly and the TDS increases sharply. This subsequent stream is known as produced water and will continue to flow to the surface over the entire lifetime of the well (Thiel and Lienhard, 2014; API, 2015). Flowback and produced water are both enriched with materials from the shale formation such as brines, hydrocarbons, and naturally occurring radioactive materials, with the latter having the greater concentration of formation materials. The produced water generated during the production of unconventional gas from onshore activities was estimated to be approximately 3268 million m^3 in 2012 (Veil, 2015).

As an example, the average composition of produced water from fracking in the Marcellus shale (Pennsylvania), which is by far the largest and the best documented shale gas resource in the United States, is shown in Fig. 1 with seawater for comparison. The figure reports in detail the concentrations of inorganic compounds, however, organic

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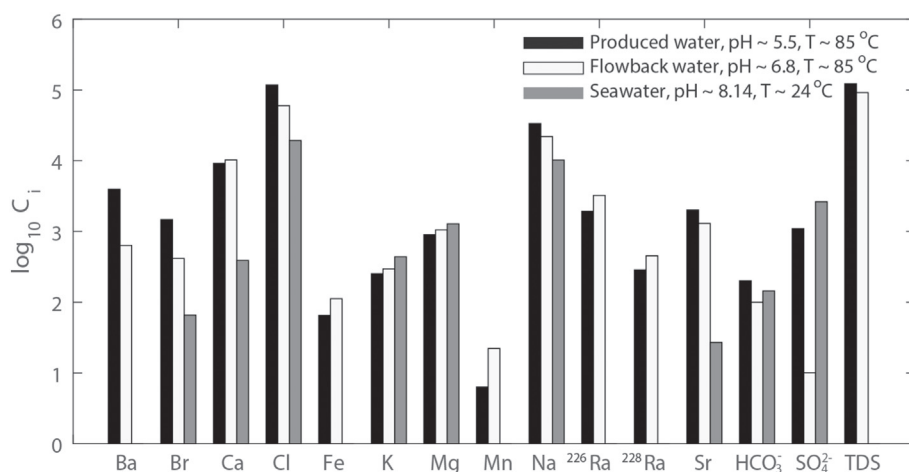


Fig. 1. Average composition of flowback and produced water from the Marcellus shale (Barbot et al., 2013; Warner et al., 2013; Acharya, 2011; Lester et al., 2015; Lauer et al., 2016; He et al., 2014) and of seawater (Nicolini et al., 2017; Yu et al., 2017) for comparison. The unit of concentration is in log scale of mg per liter (mg/L) except for radium for which is logarithm of picocuries per liter (pCi/L).

molecules are also present in the flowback and produced water. Primarily due to the composition of the fracturing fluids. Their concentration is within the micro molal range and their effect on the inorganic ion speciation and interaction is negligible (Thiel and Lienhard, 2014; Kharaka and Hanor, 2013). The salinity of produced water, given as total dissolved solids (TDS), can be as high as 10 times the TDS of seawater resulting in an ionic strength (I) between 1 and 4 mol/kg. The produced water was found to have the highest concentration of calcium (Ca^{2+}), barium (Ba^{2+}), magnesium (Mg^{2+}), sodium (Na^+), strontium (Sr^{2+}), and radium (Ra^{2+} , namely, ^{226}Ra and ^{228}Ra). The radionuclide such as radium makes the water a potential hazard in the case of a spill. Flowback and produced water leaks have been documented at locations in the U.S., such as at sites in Colorado, New Mexico, North Dakota, and Pennsylvania (Patterson et al., 2017). Between 2005 and 2014, reported spills ranged from the hundreds to the thousands of cubic meters of flowback and produced water. These spills usually occurred during storage in tanks and pits and when moving the fluids via flowlines. Upon a spill, the fluid may migrate towards the shallow aquifers and potentially contaminate it. To identify the level of risk, the prediction of the transport of the major hazardous compounds in the flowback and produced water is required. At this aim, of particular interest is the understanding of the transport of radium, because of its relatively long half-life and high radiotoxicity, especially ^{226}Ra (Zhang et al., 2015; Rowan et al., 2011; Kirby et al., 1964).

Radium is an alkaline earth metal. The sixth element in the Group II of the periodic table and it is a radioactive isotope part of the decay chains of U isotopes (Rowan et al., 2011; Kirby et al., 1964). Its physical and chemical properties (congener) resemble Ba^{2+} , which is generally used as a proxy in laboratory experiments instead of Ra^{2+} because of its less hazardous properties. Major processes affecting solute transport in subsurface permeable rocks are adsorption and desorption onto and from the reactive surface of the minerals coating the grains of the porous medium. Iron oxides, such as goethite ($\text{FeO}(\text{OH})$), are among the most common minerals in the subsurface and they are present often as coating on rocks (Mishra and Tiwary, 1999). Iron oxides are also very reactive towards Ra^{2+} and have been proposed as based sorbents for coagulants in uranium mining water treatment (Chellam and Clifford, 2002). Earlier studies (Bassot et al., 2001; Sverjensky, 2005; Sajih et al., 2014) have been carried out to investigate the adsorption of Ra^{2+} or Ba^{2+} as a function of pH and for I up to approximately 0.1 mol/kg. The results from these studies show negligible effect of the ionic strength on the cations' transport (Hayes, 1987). However, experimental data are not available at conditions typical of flowback and produced water, i.e., for I between 1 and 3 mol/kg (Thiel and Lienhard, 2014). Moreover, at the best of the authors' knowledge, there are no studies where the transport of Ba^{2+} has been investigated systematically at various pH and ionic strength.

In this paper, a study on the transport behaviour of Ba^{2+} through a porous medium containing $\text{FeO}(\text{OH})$ is presented. Experiments in batch and with a column-flood system were carried out under various conditions of pH, I , and temperature. The experiments were described with the triple layer surface complexation model coupled with the Pitzer activity coefficient method and a reactive transport model, in the case of the transport tests. The paper is divided into four sections. In Section 2, the materials and the methods used and applied during the experiments are described; in Section 3, the model implemented to describe the adsorption/desorption of Ba^{2+} in batch and during transport through the reactive porous medium is explained; in Section 4, the results and their comparison with the model are shown and discussed and simulations of radium transport are reported; finally in Section 5, the conclusions are drawn.

2. Materials and methods

2.1. Synthesis of goethite

Goethite was prepared following Atkinson (1967). A mass of 50 g of $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (98.0 to 101.0 % purity, Fisher Scientific, U.S.A.) was added to 825 mL of deionized water. A volume of 200 mL of 2.5 N KOH (Fisher Scientific, U.S.A.) was added to the previous solution and aged at 60 °C for 24 hours. Afterwards, the $\text{FeO}(\text{OH})$ suspension was purified through dialysis using a cellulose tube. The tube with the $\text{FeO}(\text{OH})$ suspension was immersed in deionized water and the water was changed twice a day until the nitrate (NO_3^-) concentration became less than 10^{-4} mol/kg. Then, a sample of suspension of approximately of 5 mL in volume was dried at 105 °C for 24 hours for characterization as explained below; whereas, the rest of the $\text{FeO}(\text{OH})$ suspension was stored in polythene bottles to be used as a coating for glass beads.

2.2. Coating the glass beads with synthesized $\text{FeO}(\text{OH})$

Silica dioxide beads (Potters, U.S.A.) of average size 0.2 mm were coated with $\text{FeO}(\text{OH})$ following the procedure formulated by Ying and Axe (2005), with some modifications. A mass of 230 g of beads were washed using 460 mL 1 N HNO_3 for 24 hours and dried at 105 °C for additional 24 hours. The beads were mixed with the suspension of synthesized $\text{FeO}(\text{OH})$ for 4 days at room temperature. The system was continuously controlled with a pH meter (Model Orbisint CPS11D, Endress Hauser, U.S.A.) and maintained at pH of 7.5. The mixture was dried at 60 °C for 6 days, rinsed with deionized water, and then dried at 60 °C for 6 days. The beads were cooled down at room temperature and stored in polythene bottles (Thermo Scientific, U.S.A.) to be then used for adsorption and transport tests as described in the corresponding sections below.

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