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Textural and compositional controls on mudrock breakthrough pressure and permeability

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ABSTRACT

Breakthrough pressure and permeability control the structural sealing capacity of mudrocks in hydrocarbon reservoirs and CO_2 sequestration sites. Breakthrough pressure studies show a wide array of results due to the natural heterogeneity of mudrocks. This includes the effects of variability in grain size distribution, fine sediment fraction, and mineral composition. Resedimentation techniques have been widely applied to the study of mudrock permeability but little work focused on breakthrough pressure. Here we perform a parametric study on breakthrough pressure and permeability in resedimented mudrocks using samples composed of ternary mixtures of kaolinite, quartz, and lime. The texture, or grain size distributions, and composition, or mineralogical concentration, of our mudrock fabric is systematically varied. We show that lime is effective in cementing resedimented samples, producing mechanically competent mudrocks and allowing for robust mercury injection capillary porosimetry. We observed that breakthrough pressure increases and permeability decreases with fine-grained void ratio, rather than bulk void ratio, according to power-law relationships. The fine-grained void ratio is defined as the ratio of the volume of voids to volume of fine grains where the size of the fine grains is controlled by the structure of the coarse-grained percolating network. These relationships can be used to estimate the sealing capacity of shaly sandstones and sandy mudrocks and highlight the variability of transport properties in coarse-fine sediment mixtures.

Plain Language Summary: Carbon dioxide (CO_2), a greenhouse gas, can be injected into and sequestered within deep brine filled aquifers and hydrocarbon reservoirs to reduce atmospheric emissions. After injection, CO_2 rises buoyantly until it collects beneath a rock formation that is much less permeable than the reservoir. These formations which trap CO_2 are often mudrocks. Predicting how well a mudrock can hold back the CO_2 from rising to the surface is vital to the success of any underground sequestration effort, but this is a daunting task given the wide variability in natural mudrocks. To address this problem, we created artificial mudrocks in the laboratory and measured the amount of pressure it takes for CO_2 to first begin to flow, or breakthrough, the rocks. We determined that the breakthrough pressure of mudrocks can be predicted by the relative amounts of pores present within the fine and coarse grained size fractions in each mudrock.

1. Introduction

The effort to mitigate climate change due to greenhouse gas emissions has been considered as the Apollo program of our time (King et al., 2015). One promising strategy is the permanent sequestration of anthropogenic carbon dioxide (CO_2) in deep saline reservoirs (Benson and Cook, 2005). Target reservoirs are often composed of massive sandstone formations (reservoirs) capped by low permeability shales (caprocks) (Downey, 1984; Espinoza and Santamarina, 2017) under which the CO_2 collects and remains trapped. Caprock integrity is also important when evaluating hydrocarbon reserves. If the reservoir pressure exceeds the sealing capacity of the caprock then, over geologic timescales, the reservoir will be depleted until the pressures are equilibrated with the capillary forces. Thus, the successful evaluation of hydrocarbon reservoirs and the security of CO_2 sequestration in deep geologic formations as a climate change mitigation strategy hinges on how well the caprocks function as seals.

The sealing capacity of a caprock directly depends on two related factors: how much pressure is needed for a non-wetting fluid (e.g., CO_2, oil, or gas) to break through the barrier and how much pressure builds up under the caprock. The pressure difference experienced at the caprock interface by a continuous buoyant non-wetting plume with height h is:

$$\Delta P = \left(\rho_b - \rho_g\right) g h \tag{1}$$

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where ρ_b is the brine mass density, ρ_g is the non-wetting phase mass density, and g is the gravitational acceleration, and h is the height or thickness of the non-wetting plume, usually referred to as the CO_2/oil/gas column (Peters, 2012; Schowalter, 1979). The breakthrough pressure ($P_c{}^*$), or displacement pressure, of a given porous medium can be theoretically calculated using the Young-Laplace equation (Laplace et al., 1829; Young, 1805):

$$P_c^* = \frac{2\sigma\cos(\theta)}{r_c^*} \tag{2}$$

where θ is the contact angle of formation brine in the presence of the non-wetting fluid, σ is the interfacial tension between the brine and the non-wetting fluid and $r_c{}^*$, the critical pore radius, is the minimum pore radius along an interconnected and continuous path of pores across the porous medium at breakthrough. The breakthrough pressure is the minimum pressure differential for a nonwetting fluid to begin to flow across a sample, or formation.

Mercury intrusion capillary porosimetry (MICP) is widely used in the oil industry to investigate entry pressure, breakthrough pressure, and estimate the permeability of porous media. This method utilizes the Laplace equation and an understanding of the contact angle of mercury to develop a pore throat distribution. Once this pore throat distribution is known, an assumed contact angle can be used to estimate the entry pressure and breakthrough pressure of any fluid. This technique is easy to run and readily reproduced making it ideal for conducting a large number of breakthrough pressure experiments. The disadvantages of this technique are due to the uncertainties in contact angle and the possibility for shrinking of clays during the drying processes. For these reasons, the best way to establish definitive breakthrough pressures of CO_2 in a particular shale is through CO_2 breakthrough pressure experiments. In this paper, we present our MICP estimates in terms of CO_2, however our goal is to understand the fundamental controls on breakthrough pressure and not the absolute value of any one particular sample/fluid combination.

CO_2 breakthrough pressure measurements on a variety of natural formations range from approximately 0.1 MPa to 5 MPa (Harrington et al., 2009; Hildenbrand et al., 2002, 2004; Wollenweber et al., 2010). Evaporites exhibit some of the highest breakthrough pressures with values as great as 21 MPa for CO_2 (Li et al., 2005). CO_2 breakthrough pressure experiments on resedimented samples, have shown that $r_c{}^*$ (Eq. 2) is generally larger than the mean pore throat radius in homogeneous porous media (Espinoza and Santamarina, 2010). A summary of recent breakthrough pressure experiments highlights the role of particle size distribution and specific surface on $P_c{}^*$ (Espinoza and Santamarina, 2017; Makhnenko et al., 2017).

Breakthrough pressure plays an important role in the Sleipner CO_2 sequestration project in the North Sea, for example, where nearly a million tons of CO_2 have been injected each year since 1996 (Arts et al., 2004; Chadwick et al., 2010). Monitoring of the CO_2 plume with 4D seismic surveys reveal that the plume has ascended rapidly through eight thin shaly barriers at unexpectedly low breakthrough pressures before being trapped beneath a large competent caprock (Cavanagh and Haszeldine, 2014). This unexpected plume behavior suggests that some properties of the CO_2 plume barriers have not been accurately characterized. The causes for such phenomenon partly originate on the spatial variability and heterogeneous nature of mudrocks. Mudrocks are usually recognized by its amount of clay minerals in the rock matrix. Typically well logging measurements, such as gamma ray emission and spontaneous potential, measure bulk volume fractions of clay content in shaly sediments, but cannot tell the location of clays within the rock matrix and their effect on transport properties (Peters, 2012). Hence, it is not straightforward to assess the sealing capacity of shaly sandstones or sandy shales from gamma ray or spontaneous potential only. The study of synthetic resedimented mudrock samples helps understand better these sediment mixtures.

Resedimentation is the process by which slurries are incrementally compressed to high effective stress under oedometric conditions (one-dimensional strain) to mimic the sedimentation process (Germaine and Germaine, 2009). The process can be used to create synthetic mudrocks with prescribed mineralogy, grain size, and a target porosity or effective stress. The creation of resedimented mudrocks allows for the systematic study of a single factor such as the mineral composition, grain size, or effective stress, thus reducing the variability produced by the study of natural materials (Schneider et al., 2011). Resedimentation experiments have been used to explore permeability, porosity and compressibility as a function of mineralogical composition, however, generally employing a narrow grain size distribution and/or low effective stress as required for studying deep mudrocks (Bandini and Sathiskumar, 2009; Dewhurst et al., 1999, 1998; Espinoza and Santamarina, 2017; Mondol et al., 2008, 2007; Reece et al., 2013; Schneider et al., 2011; Shafiee, 2008). For example, Schneider et al. (2011) used resedimentation techniques on a natural illitic mudrock that was mixed with silt-sized silica to create five different mixtures ranging in composition from 36% to 57% clay by weight. Each sample was consolidated to a maximum vertical effective stress of 2.4 MPa and vertical permeability was calculated assuming a constant strain-rate during consolidation increments. Scanning electron microscopy (SEM) analysis of these samples showed that the silt sized silica grains formed “silt bridges” which allowed for increased permeability with decreasing clay fraction. They proposed a dual porosity model where the clay and silt fractions have separate permeabilities which contribute to the overall effective permeability.

Espinoza and Santamarina (2017) used resedimentation experiments to explore breakthrough pressure in sediments with CO_2. The authors consolidated homogeneous specimens of fine sand, calcium carbonate, kaolinite, and montmorillonite to a maximum effective stress of 3 MPa within a spring-loaded oedometer flow-through cell. They found that breakthrough pressure corresponded with pore throats larger than the mean for their specimens with breakthrough pressures ranging from 0.11 MPa in silt-sized calcium carbonate to a maximum of 3.0 MPa in pure montmorillonite clay. However, their work did not explore various sediment mixtures which is more representative of real mudrocks. Makhnenko et al. (2017) conducted CO_2 breakthrough pressure experiments on a resedimented opalinus clay and found good agreement between resedimented and natural samples.

Our work presented here builds on that of Espinoza and Santamarina (2017) and Schneider et al. (2011), as well as other previous studies of resedimented mudrocks by exploring the effect of mixtures of quartz, kaolinite, and lime on the transport properties of mudrocks. These are representative of typical constituents of mudrocks – fine siliciclastic grains (represented by quartz), clay (kaolinite) and a carbonate cement (lime). By mixing three minerals into our slurries we produce samples with varied mineralogy, porosity, and pore structure. This helps achieve our goal of developing predictive relationships between mineral composition and grain texture with mudrock permeability and breakthrough pressure simultaneously. Here, for the first time, we analyze the entry pressure, breakthrough pressure, and permeability estimated from MICP of controlled sediment mixtures consolidated to high effective stresses. We synthesize the results by regression and then compare them to previous work.

2. Experimental methods

2.1. Materials and consolidation technique

Three types of minerals were used: fine grained quartz, hydrated lime, and kaolinite (Table 1). The quartz was supplied by Allied Compounds and passed a 200 mesh (74 µm). The hydrated lime was supplied by Aqua Phoenix Scientific with 94% passing a 325 mesh (44 µm) and 99% passing a 200 mesh. A hydrometer was used to measure the particle size distribution for each mineral component according to ASTM D7928 (Fig. 1). A high powered commercial blender (Waring Model

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