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Using mercury injection pressure analyses to estimate sealing capacity of the Tuscaloosa marine shale in Mississippi, USA: Implications for carbon dioxide sequestration

Celeste D. Lohr*, Paul C. Hackley

U.S. Geological Survey, Eastern Energy Resources Science Center, 12201 Sunrise Valley Dr., Reston, VA, 20192, USA

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ABSTRACT

This work used mercury injection capillary pressure (MICP) analyses of the Tuscaloosa Group in Mississippi, including the Tuscaloosa marine shale (TMS), to assess their efficacy and sealing capacity for geologic carbon dioxide (CO_2) sequestration. Tuscaloosa Group porosity and permeability from MICP were evaluated to calculate CO_2 column height retention. TMS and Lower Tuscaloosa shale samples have, respectively, Swanson permeability values less than 0.003 md and 0.00245 md; porosity from 3.86% to 9.86% and 1.34% to 7.96%; median pore throat sizes from 0.00342 to 0.0111 µm and 0.00311 to 0.017 µm; and pore radii from 0.0130 to 0.152 µm and 0.0132 to 0.149 µm. Mercury entry pressures for the TMS and Lower Tuscaloosa range from 4.9 to 57.1 MPa and 5.0 to 56.3 MPa, respectively. Calculated CO_2 column heights that the TMS sample set can retain in the reservoir range from 23 to 255 m when the TMS is near 100% water saturation. Potential top seal leakage is more likely to be influenced by the numerous well penetrations through the confining system of the TMS rather than capillary failure. Results of this study demonstrate desirable sealing capacity of the TMS for geologic CO_2 sequestration in reservoir sandstones of the Lower Tuscaloosa and could provide an analogue to other potential CO_2 sequestration top seals.

1. Introduction

Top seal evaluation is a critical step in determining the long-term integrity of a potential seal to keep carbon dioxide (CO_2) in place for CO_2 sequestration. It is also a required component of site characterization of CO_2 injection wells for geologic sequestration (Class VI wells) (U.S. Environmental Protection Agency, 2013). This evaluation is necessary to protect underground sources of drinking water, to ensure that the geologic formation has adequate storage capacity, and that the confining zone will prevent fluid movement out of the injection zone (U.S. Environmental Protection Agency, 2013). However, there are few published studies that characterize top seals of CO_2 sequestration projects in the United States in enough detail to evaluate seal potential. Lu et al. (2011) characterized the sealing capacity and diagenesis of the Tuscaloosa marine shale (TMS) at the Cranfield CO_2 injection site in Mississippi. Their work focused on one site in the TMS and, in order to better characterize the regional seal integrity of the TMS, a more detailed investigation that encompasses a wider geospatial area is necessary. Therefore, we utilized mercury injection pressure analyses (MICP) of TMS samples from across southern Mississippi to augment the existing seal evaluation narrative of this shale.

Seal potential includes 1) seal capacity (the capillary pressure, or column height, at which a trapped fluid begins to move through a seal); 2) seal geometry (structural position, thickness, and areal extent of the lithology); and 3) seal integrity (rock mechanical properties) (Daniel and Kaldi, 2009; Kaldi and Atkinson, 1997). It is generally accepted that caprock leakage of CO_2 could occur by diffusion, capillary breakthrough, and fracture flow (Busch et al., 2010; Chiquet et al., 2005; Edlmann et al., 2013). Studies have shown that diffusive losses through a seal are negligible (Busch et al., 2008, 2010; Hildenbrand and Krooss, 2003); however, top seal leakage by capillary breakthrough and fracture flow is a greater concern (Busch et al., 2010; Chiquet et al., 2007; Edlmann et al., 2013). Capillary breakthrough occurs when the difference between the pressure in the injected phase (such as CO_2) and the pressure in the brine that saturates the caprock exceeds the capillary entry pressure (Chiquet et al., 2005, 2007). Mudrocks are generally considered to be effective top seals because they have low permeabilities, high capillary entry pressures, and are usually laterally continuous (Grunau, 1987; Ingram and Urai, 1999). In some tight shales, the entry pressure is so high that capillary failure is nearly

* Corresponding author at: 12201 Sunrise Valley Dr., MS 956, Reston, VA, 20192, USA.
E-mail address: clohr@usgs.gov (C.D. Lohr).

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inconceivable (Watts, 1987) and geologically unreasonable.

The TMS is a member of the Tuscaloosa Group, which is historically known as an oil and gas play in the U.S. Gulf Coast region, with conventional production from sand reservoirs of the Lower Tuscaloosa and continuous production from the overlying TMS (Allen et al., 2014; Ambrose et al., 2015; Berch and Nunn, 2014; John et al., 1997; Mancini et al., 1987). It has also received interest as a potential reservoir for geologic CO_2 storage (Drumm and Nunn, 2012) and a monitoring site for CO_2 sequestration and enhanced oil recovery (EOR) has been developed in the Cranfield oil field of Adams County, southwest Mississippi (Anderson et al., 2017; Lu et al., 2011, 2012, 2013; Rinehart et al., 2016). At Cranfield, CO_2 was injected into the Lower Tuscaloosa sandstones and the TMS acted as the seal (Hovorka et al., 2013; Lu et al., 2011). This setting has provided a unique opportunity to characterize, model, and monitor CO_2 injection to ultimately develop technologies to "support industries' ability to predict CO_2 storage capacity in geologic formations" and to "demonstrate that…injected CO_2 remains in the injection zones" (Hovorka et al., 2013; National Energy Technology Laboratory, 2011). The Cranfield site also provided the ability to evaluate and characterize a marine shale as a seal for a CO_2 injection zone. Several studies by Lu et al. (2011, 2015) provide petrophysical, mineralogical, and geochemical properties of the TMS in Cranfield field and conclude that this shale shows a high sealing capacity for CO_2 and should be a good confining system for CO_2 sequestration.

In this study, MICP analyses were performed on TMS samples in order to calculate CO_2 column height retention and to characterize porosity and permeability. Shale samples from the Lower Tuscaloosa were also analyzed *via* MICP to provide a comparison of the lower and middle Tuscaloosa Group units. This study does not integrate other factors that may affect sealing properties, such as ductility (Ingram and Urai, 1999), mineralogy (Busch et al., 2008; Lal, 1999; Wollenweber et al., 2010), or diagenesis (Jiao and Surdam, 1993; Katsube and Williamson, 1994; Lu et al., 2011). However, results presented herein address Kaldi and Atkinson's (1997) "seal capacity" classification of top seals and quantify the sealing capacity of the TMS and its implications for CO_2 sequestration. Results of well penetration density into the TMS are also used to examine potential effects on seal integrity.

2. Geologic setting

The TMS is part of the Upper Cretaceous Tuscaloosa Group, which represents a depositional cycle where the lower, middle (marine), and upper units correspond to the transgressive, inundative, and regressive components of the cycle, respectively (Spooner, 1964). The middle Tuscaloosa, or TMS, is a dark grey marine shale containing argillaceous components and minor calcareous zones (Spooner, 1964). In southern Mississippi, the TMS consists of laminated mudstone interbedded with silty, fossiliferous, calcareous siltstone and very fine-grained sandstone (Lu et al., 2011; Miranda and Walters, 1992); in northern regions of Mississippi, Alabama, and Tennessee, the predominant TMS lithology changes to sandstone (Griffith et al., 2011). In some areas of South Carlton field in Clarke and Baldwin counties, Alabama, a grey, silty, oyster packstone is present at the base of the TMS (Mancini et al., 1987).

Overall, the TMS varies in thickness (Fig. 1) from approximately 23 m (75 ft) in eastern Louisiana and southwest Mississippi to 151 m (495 ft) in southeastern Louisiana and Amite County, Mississippi (Enomoto et al., 2017). The TMS is also characterized by a relatively high log resistivity zone (HRZ) of 5 ohm-m or greater in its basal portion, which ranges in thickness from zero to 99 m (325 ft) (John et al., 1997). This area of high resistivity, which peaks up to 11 ohm-m in the Pike County Sun #1 Spinks well studied by Lu et al. (2015), has become a primary zone of interest for hydrocarbon production and is the typical landing zone for horizontal well completions. The HRZ is lithologically similar to the overlying TMS according to X-ray diffraction mineralogy and programmed pyrolysis analyses; high resistivity is therefore interpreted to result from the presence of hydrocarbons (Enomoto et al., 2017). X-ray diffraction analyses reveal that illite, kaolinite, and quartz are the major minerals present in shales of the TMS and its basal HRZ in southern Mississippi (Enomoto et al., 2018; Lu et al., 2011). Programmed pyrolysis and organic petrology analyses of the TMS and TMS HRZ reveal that TOC (weight %) ranges from 0.14 to 5.22 and measured percent vitrinite reflectance ranges from 0.53 to 1.24 (Enomoto et al., 2018).

3. Materials and methods

3.1. Samples

We used a total of 31 shale samples for mercury intrusion capillary pressure (MICP) analyses. These included 14 TMS samples from three legacy cores acquired from the Mississippi Department of Environmental Quality (DEQ) Office of Geology in November 2016. These samples range in depth from 3,247.6 to 3,700.3 m (10,654.8 to 12,140.0 ft) in Amite, Pike, and Lincoln counties in Mississippi.

Two legacy TMS core samples from 3,100.9 to 3106.8 m (10,173.4 to 10193.05 ft) from the Bureau of Economic Geology (BEG) at University of Texas at Austin were also obtained from the Cranfield Unit 31F-2 well, the same well evaluated by Lu et al. (2011) for sealing capacity in the middle Tuscaloosa mudstone Cranfield CO_2 injection site in Mississippi. Additionally, two TMS core samples from a proprietary well in Wilkinson County, Mississippi were donated by Sanchez Oil and Gas Corporation.

Also included was one TMS core sample in Louisiana from the Louisiana Geological Survey (LGS) and four TMS and TMS HRZ cuttings samples in Mississippi from DEQ, all obtained in 2015 from ongoing USGS national oil and gas assessment research. The four Mississippi samples were composite samples, consisting of cuttings compiled from multiple depth intervals so that a sufficient amount of rock material was available for MICP analysis. Cuttings samples were high-graded for rock material that was darker in color to minimize inclusion of dried mud filtrate, mud additives, and out-of-section cavings contamination.

Additional MICP analyses were obtained for eight shale samples from Lower Tuscaloosa core. These samples were acquired from the Mississippi DEQ, LGS, and BEG during 2013–2015. The core samples range in depth from 3,275.1 to 4,339.5 m (10,745.0 to 14,237.2 ft) in Adams, Amite, and Pike counties, Mississippi and in East Feliciana and West Feliciana parishes, Louisiana. Locations of TMS, TMS HRZ, and Lower Tuscaloosa samples are illustrated in Fig. 1.

3.2. Mercury injection capillary pressure analysis

Mercury injection capillary pressure (MICP) analysis was utilized to characterize the pore systems and capillary properties of core and cuttings samples from the Tuscaloosa Group. All MICP analyses were performed by Core Laboratories. Upon receipt by the laboratory, each sample was photographed for internal reference before it was crushed to approximately 20–35 mesh size; the reasoning for this mesh size can be found in Comisky et al. (2011). Soxhlet extraction was performed to remove any salts, hydrocarbons, and water that might have been present in each sample. All samples were then vacuum oven dried at 82 °C (180 °F) and their weights tracked until stabilization, or < 0.01 g change over two days. Ambient properties, such as dry weight and grain volume, were recorded for all samples. Each sample was then placed into a penetrometer; the penetrometer is no more than half full to prevent material loss. A vacuum was applied for approximately 30–45 min before injection of mercury into the crushed sample. The injection of mercury into the sample begins at approximately 12 psi and increases stepwise for 85 pressure points after achieving an equilibrium time of two minutes at each step. Equilibration allows for sufficient time for the mercury to fill available pores at each stepwise pressure