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Structural trapping capacity of oil-wet caprock as a function of pressure, temperature and salinity

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ABSTRACT

Wettability is a major parameter which significantly influences structural trapping capacities in CO_2 geo-sequestration. In this context, the original wettability state of a caprock is of key importance, however, less attention has been given towards this aspect in the past. We thus evaluated the impact of caprock oil-wettability on storage potential; we used five mica substrates as representatives of caprock and modified their initial wettability to obtain different oil-wetness (0–118° water contact angle at ambient conditions), so that we were able to conduct a systematic study. Advancing and receding contact angles (θ_a and θ_r) were measured on all surfaces for wide ranges of pressure (0.1 MPa–20 MPa), temperature (308 K, 323 K and 343 K) and salinity (0 wt%–20 wt% NaCl). The results indicate that advancing and receding contact angles increase with pressure (when pressure increased from 0.1 MPa to 20 MPa at 343 K, θ_a increased from 0° to 67° for water-wet substrate and from 73° to 156° for oil-wet substrate), and salinity but decrease with temperature.

Finally we predict CO_2 column heights, which can be permanently stored beneath oil-wet caprocks. Clearly, the structural trapping capacity is significantly reduced in case of oil-wet caprock (when compared to water-wet caprock). We conclude that it is essential to evaluate CO_2-wettability of caprocks to determine safe limits of operation for containment security.

1. Introduction

In carbon geo-sequestration (CGS), CO_2 captured from a source is transferred to the wellsite and then injected into depleted hydrocarbon reservoirs or deep saline aquifers for long term underground storage (e.g. Blunt et al., 1993; Iglauer et al., 2013; Metz et al., 2005; Pentland et al., 2011). CGS is considered to be the most promising approach in terms of reducing anthropogenic CO_2 and thus ensures a cleaner environment (Lackner, 2003). CO_2 is also frequently injected into reservoirs for enhanced oil recovery to accelerate oil production by displacing oil from unswept zones and itself being stored in the process (Blunt et al., 1993; Iglauer et al., 2013). During CO_2 injection into reservoirs, a continuous buoyant CO_2 phase rises upwards due to lighter CO_2 density in comparison to formation water and tends to leak through the caprock. However, certain storage/trapping mechanisms render CO_2 immobile and these are: structural trapping (Hesse et al., 2008; Ketzer et al., 2012), residual trapping (Iglauer et al., 2011a,b; Juanes et al., 2006; Raza et al., 2015), dissolution trapping (Iglauer, 2011c; Lindeberg and Wessel-Berg, 1997), and mineral trapping (Gaus, 2010).

Structural trapping, which is the principal storage mechanism in the first decades of a project, refers to the barrier offered by overlaying caprock to prevent capillary leakage of CO_2 (Iglauer et al., 2015a). Caprock is characterized by an extremely low permeability layer and can be in the form of mudrocks, shales, clays and tight carbonates and is generally composed of anhydrite, halite, muscovite (mica), kaolinite, calcite and other minerals (Chiquet et al., 2007; Iglauer et al., 2015b; Skalli et al., 2006). As soon as injection begins, buoyant CO_2 is prevented from migration to shallower zones by the seal provided by caprock and thus structural trapping retains CO_2 within the storage formation.

In this context, wettability of caprocks for CO_2-brine systems is an important physicochemical parameter that dominantly influences the structural trapping capacity (Iglauer et al., 2015b). Few studies exist in the literature in which contact angles were measured on mica surfaces (as representative of caprock) to characterize wettability of caprocks at relevant thermophysical conditions (Arif et al., 2016; Broseta et al., 2012; Chiquet et al., 2007; Farokhpoor et al., 2013; Mills et al., 2011; Wan et al., 2014; Wang et al., 2013). However, we find that no significant attention has been given to oil-wet caprocks in order to evaluate their storage

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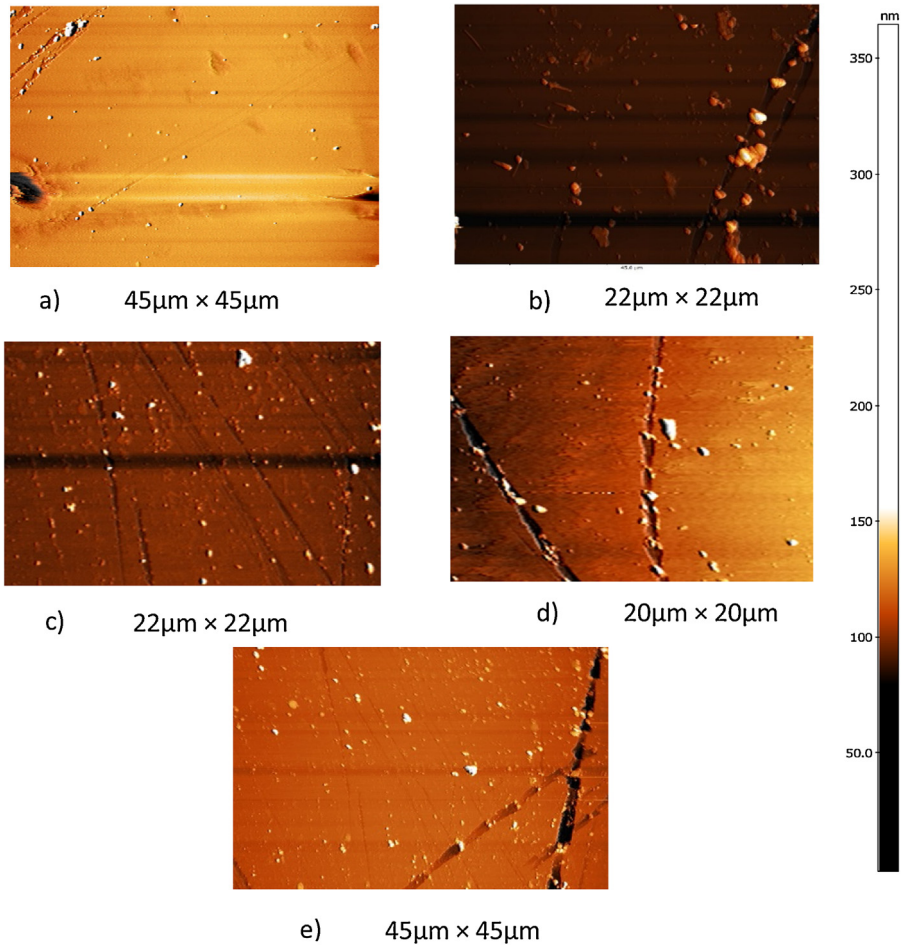

Fig. 1. Atomic force microscopy images (2D-view) for all mica substrates used in the study; a) M1 (roughness = 12 nm), b) M2 (roughness = 13 nm), c) M3 (roughness = 16 nm), d) M4 (roughness = 15 nm), e) M5 (roughness = 10 nm). Scale bar on the right represents surface heights associated with different spots on the image.

potential, particularly when considering the fact that caprocks can be naturally water-wet, intermediate-wet or oil-wet (Larter and Aplin, 2005; Ingram et al., 1997; Larter et al., 1996, 2002; Taylor et al., 1997). Moreover, the wetting properties of seals may change over time too: an initially water-wet seal may evolve into an oil-wet seal due to adsorption of a variety of compounds from crude oil, such as asphaltenes (Anderson, 1986). Although it is established that in a hypothetical CO_2-wet rock, upwards directed suction force is created and CO_2 can leak through the caprock causing significant reduction in structural trapping (Iglauer et al., 2015a), and residual trapping is reduced, too (Chaudhary et al., 2013; Iglauer et al., 2012a; Spiteri et al., 2008), yet two vital questions exist: 1) what wettability is offered by an oil-wet caprock surface to CO_2-water systems at storage conditions?, 2) what are safe limits of operating conditions for CO_2 storage in strata which are overlayed by oil-wet, intermediate-wet, and water-wet caprocks?

To answer these questions and to improve the understanding of CO_2 wettability of different sorts of caprocks, we altered the wettability of four mica surfaces and measured advancing and receding water contact angles on strongly oil-wet, weakly oil-wet, intermediate-wet, weakly water-wet and strongly water-wet substrates at different pressures (0.1 MPa, 5 MPa, 10 MPa, 15 MPa, 20 MPa) and temperatures (308 K, 323 K, and 343 K) relevant to geo-sequestration conditions. Finally, we discuss the implications of the measured data and highlight the consequences for CO_2 storage in formations sealed by oil-wet caprocks.

2. Materials and methods

2.1. AFM measurements

Five mica (muscovite) samples M1, M2, M3 M4 and M5 were used in this study and as a first step surface roughness of the samples were measured using an atomic force microscope (AFM instrument model DSE 95-200, Semilab). The samples were very homogeneous with surface roughness ranging from 10 to 16 nm (Fig. 1). We note that the sample sizes were ~1 cm × 0.5 cm × 0.2 mm, and images taken at different areas of the sample resulted in almost identical surface roughness values implying that roughness is representative at the scale of the contact angle measurements. Moreover, the similarity of the samples is such that we do not expect any significant variation in contact angle due to surface roughness effects (e.g. cp. Al-Yaseri et al., 2015a).

2.2. Materials

We used an alkyl silane namely 'Dodecyltriethoxysilane' (Product specification: 98 mol%, Sigma Aldrich, $C_{19}H_{40}O_3Si$, MW: 332.59 g/mol, density = 0.875 g/ml at 20 °C) because of its efficiency in terms of wettability alteration (Al-Anssari et al., 2016; Grate et al., 2012; London et al., 2013; Wei et al., 1993). Surface alkylation is considered a fundamental procedure adopted in laboratory studies on rocks/minerals for wettability alteration (Arkles et al., 2009; Fadeev, 2006; Menawat et al., 1984; Tiab and Donaldson, 2011; Wei