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Short communication

Improving basalt wettability to de-risk CO₂ geo-storage in basaltic formations

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Abstract:

CO₂ geo-storage in basaltic formations has recently been identified as a viable option to rapidly dispose large quantities of CO₂, hence mitigating anthropogenic CO₂ emissions. However, it has been shown that basalt is weakly water-wet or intermediate-wet at typical storage conditions, which reduces capillary trapping capacities and increases lateral and vertical spreading of the CO₂ plume; and these effects increase project risk. We thus propose here to prime basalt surfaces with anionic surfactant (here we used sodium dodecyl benzene sulfonate), and demonstrate that such priming is highly efficient, and renders the basalt completely water-wet even at high pressures and minute sodium dodecyl benzene sulfonate concentrations. Such a wettability alteration can therefore significantly de-risk storage projects. This work aids in the improvement of CO₂ storage in basaltic formations and supports implementation of industrial-scale CO₂ geo-sequestration and climate change mitigation.

1. Introduction

Carbon geo-sequestration in basaltic formations has been identified as a possible route to mitigate climate change (Oelkers et al., 2008; Gudbrandsson et al., 2011; Wolff-Boenisch et al., 2011; Schaefer et al., 2013; Matter et al., 2016; McGrail et al., 2016). Basalt is igneous rock which is abundant and widespread in the subsurface (le Roex, 1998; Ernst, 2014). In principle, basalt is therefore an attractive CO₂ sink as it can potentially store very large quantities of CO₂, and that in a geographically widespread fashion (at a global level). Basalt has, however, been previously shown that basalt is not completely water-wet at typical storage conditions, but instead basalt is intermediate-wet or weakly water-wet (Iglauer et al., 2020). Reduced water-wettability leads to higher CO₂ relative permeabilities, which result in higher CO₂ mobilities and consequently higher lateral and vertical CO₂ spread (Al-Khdheawi et al., 2017, 2018). Furthermore, capillary trapping efficiencies are reduced in less water-wet rock (Al-Menhali and Krevor, 2016; Al-Menhali et al., 2016; Rahman et al., 2016). It is therefore desirable to develop methods with which a rock's water-wettability can be increased. Here we propose

to use anionic surfactant for this purpose, and we demonstrate that water-wettability can be drastically increased to even complete water-wetness via this method.

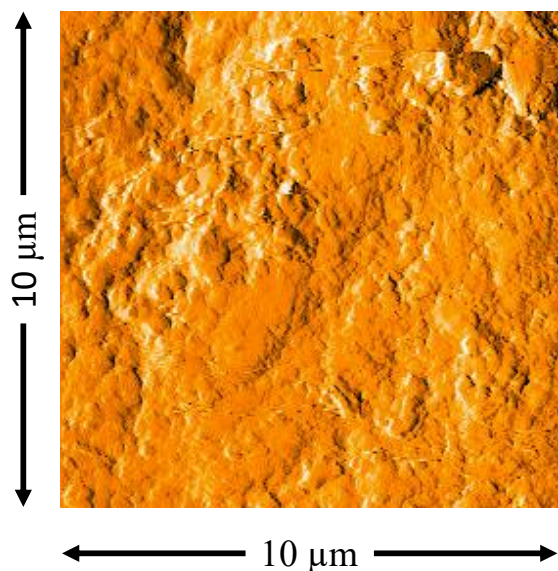
2. Experimental procedure

The basalt samples used here stem from the CarbFix injection site (in Iceland), and they have been thoroughly described previously (Gislason et al., 2010; Aradottir et al., 2012; Alfredsson et al., 2013; Snæbjörnsdóttir et al., 2017, 2018). The basalt was cut to cuboid basalt specimens with a high speed diamond blade (to ~ 0.5 cm × 1 cm × 1 cm) and each sample was exposed to air plasma for 1 min to remove organic surface contaminants (Love et al., 2005; Iglauer et al., 2014). The composition of the basalt tested was also measured via X-ray diffraction (XRD) (with a Bruker D8 Discover Plus/Atlas instrument), results are tabulated in Table 1.

Separately, various aqueous NaCl (reagent grade, from Sigma-Aldrich)-sodium dodecyl benzene sulfonate (SDBS) (technical grade, from Sigma-Aldrich) solutions were prepared (NaCl concentration was constant at 0.3 M, and a range of

Table 1. XRD analysis results for the bulk mineralogy of basalt.

Phase	Formula	Weight%
Labradorite	$((\text{Ca}, \text{Na})(\text{Al}, \text{Si})_4\text{O}_8)$	58.6
Augite	$(\text{Ca}, \text{Na})(\text{Mg}, \text{Fe}, \text{Al}, \text{Ti})(\text{Si}, \text{Al})_2\text{O}_6$	36.5
Montmorillonite	$(\text{Na}, \text{Ca})_0 \cdot 33(\text{Al}, \text{Mg})_2(\text{Si}_4\text{O}_{10})(\text{OH})_2 \cdot n\text{H}_2\text{O}$	4.6
Quartz	SiO_2	0.3

**Fig. 1.** Surface roughness profile of a typical basalt sample (deflection signal).

SDBS concentrations were used, i.e., 0, 0.01 wt%, 0.1 wt% SDBS), and the basalt samples were aged in these formulations for 7 days at 293 K and 0.1 MPa.

Subsequently the advancing (θ_a) and receding (θ_r) water contact angles were measured using the tilted plate method (Lander et al., 1993) for several pressures (5, 10, 15 MPa) at 323 K. 210 nm root-mean-square surface roughness of the basalt surface was measured via atomic force microscopy (using a DSE 95-200 instrument; surface roughness thus had an insignificant influence on the CO_2 -brine-basalt contact angles) (Al-Yaseri et al., 2016b; Alnili et al., 2018), compare also Fig. 1. All contact angles were measured thrice and the average standard deviation of the contact angle measurements was determined as $\pm 3^\circ$ based on these replicate measurements.

In addition, zeta potentials of the basalts were measured on crushed basalt samples with a ZetaNano sizer (Model ZEN3600). Again these measurements were performed thrice and the standard deviation of these measurements was ± 3 mV. The zeta potentials were again measured in 0.3 M aqueous NaCl solution at 323 K and 0.1 MPa.

3. Results and Discussion

Brine contact angles (θ) increased strongly with increasing pressure, Fig. 2. This was caused by the high CO_2 compressibility, which leads to a drastically elevated molecular CO_2 density at high pressures (Span and Wagner, 1996). A

higher CO_2 molecular density leads to stronger CO_2 -basalt interactions and a higher affinity of CO_2 towards basalt (while water is almost incompressible). This relative shift in affinity led to the dewetting of the rock (Iglauer et al., 2012; Al-Yaseri et al., 2016b; Pan et al., 2019). Note that such an increase in with pressure has also been observed for other rocks, e.g., quartz, calcite, coal, sandstone or shale (Broseta et al., 2012; Iglauer et al., 2015; Alnili et al., 2018; Arif et al., 2019). Furthermore, it has been shown previously that inorganic minerals (e.g., quartz or calcite) are water-wet (either strongly or weakly, depending on pressure, see above; e.g., Chiquet et al., 2007; Al-Yaseri et al., 2016a; Arif et al. 2017; Stevar et al., 2019) with the exception of montmorillonite (Fauziah et al., 2018, 2021). The basalt tested here (which consisted mostly of labradorite and augite with some quartz, Table 1) demonstrated a CO_2 -wettability higher than that of other minerals (quartz, calcite, kaolinite, illite), again with the exception of montmorillonite, which had an even higher CO_2 -wettability (Fauziah et al., 2018).

Importantly, treatment with SDBS was highly efficient, and even small SDBS concentrations (0.01 wt%) reduced θ to zero, for all pressures, Fig. 2. For the pure basalt sample (SDBS concentration = 0), θ matched our previous measurements, and θ increased significantly with pressure (due to increased CO_2 -basalt intermolecular interactions at higher pressures, compare Iglauer et al., 2012; Chen et al., 2015; Al-Yaseri et al., 2016a; Iglauer, 2017; Abramov et al., 2019). Furthermore, a significant hysteresis between advancing (θ_a) and receding (θ_r) contact angles was observed-note that θ_r relates to a scenario where CO_2 is injected, while θ_a corresponds to subsequent water encroachment (either naturally or by artificial injection), Broseta et al. (2012), Arif et al. (2019). In addition, the zeta potential decreased with SDBS concentration (Table 2) as expected, e.g., compare Awan et al. (2020). The reason for this decrease is adsorption of the SDBS anion on the rock surface (Awan et al., 2020), which increases the negative electrical surface charge on the basalt. Such an increase in negative surface charge renders the rock surface more hydrophilic (Iglauer, 2017), thus CO_2 -wettability and θ dramatically decreased.

4. Conclusions and Implications

CO_2 geo-storage in basaltic formations is a viable option to significantly reduce anthropogenic CO_2 emissions (Matter et al., 2016). Here we demonstrate a method-ageing of basalt with anionic surfactant (SDBS)-with which basalt water-wettability can be drastically improved. A more water-wet

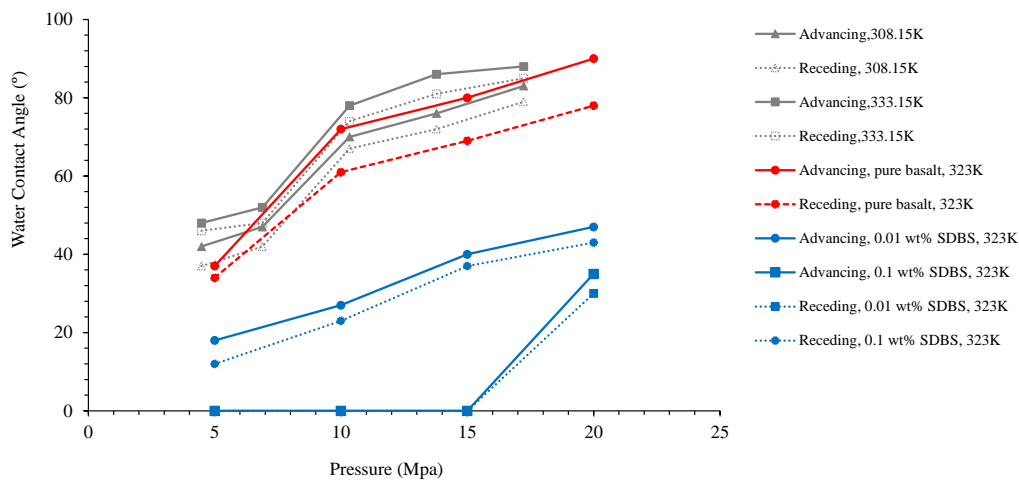


Fig. 2. Water contact angles measured on the pure and SDBS-aged basalt samples in CO₂ atmosphere.

Table 2. Zeta potentials measured for the basalt samples exposed to various SDBS formulations (NaCl concentration = 0.3 M).

Formulation	0 % SDBS (pure basalt)	0.01 wt% SDBS	0.1 wt% SDBS
Zeta potential [mV]	-19	-28	-36

rock surface is highly advantageous in terms of increased CO₂ storage capacity via capillary trapping, and reduced lateral and vertical spread of the injected CO₂ plume-resulting in reduced project risk (Al-Khdheawi et al., 2017, 2018). The method was very sensitive and even minute amounts of SDBS (0.01 wt%) were sufficient to render the basalt completely water-wet, even at high pressure (highest pressure tested was 15 MPa). This work aids in the improvement of CO₂ storage in basaltic formations, and therefore contributes to the industrial-scale implementation of CO₂ geo-sequestration.

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Conflict of interest

The authors declare no competing interest.

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