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Drainage and imbibition CO_2/brine relative permeability curves at in situ conditions for sandstone formations in western Canada

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Abstract

One of the most important mechanisms for CO_2 storage in deep saline aquifers is CO_2 trapping at irreducible saturation, which depends on the relative permeability characteristics of CO_2/brine systems. CO_2 injectivity, pressure build-up and the evolution and long-term fate of the injected CO_2 also depend on the same relative permeability characteristics. Predicting the fate of the injected CO_2 using numerical models requires adequate relative permeability relationships for CO_2/brine systems (both drainage and imbibition cycles), yet very few experimental data exist in the literature. Considering that CCS will be deployed on a large scale in western Canada, three experimental programs of determining the relative permeability characteristics at in-situ conditions of prospective storage aquifers have been successively run between 2003 and 2011. The first two programs focused on testing rocks from various deep carbonate and sandstone aquifers in central Alberta in the vicinity of several very large CO_2 sources. The third testing program focused on measuring the relative permeability for CO_2/brine systems at various locations in the sandstone Basal Aquifer that overlies the Precambrian basement in western Canada. The results of the first testing program, comprising 14 measurements, have been published in 2008. The results for relative permeability testing on eight additional carbonate rocks have been published in 2010 and combined with the results for five tests from the first measurement program to obtain generalizations for carbonate rocks. Results of new CO_2/brine relative permeability measurements on 16 sandstone rocks from the second and third measurement programs are presented for the first time in this paper. These results are combined with results of testing on six sandstone rocks from the first measurement program, for a total of 22 tests, and are generalized similarly to the generalization for carbonate rocks published previously. The test data cover a very wide range of permeability values, from less than 0.1 mD to more than 500 mD. The generalizations in terms of relative permeability and residual saturation are defined based on rock characteristics that are routinely measured in core analyses, such as pore size and absolute permeability.

Keywords: *CO_2-brine systems, relative permeability, sandstone, western Canada*

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1. Introduction

Relative permeability of the CO_2/brine system at in-situ conditions is a critical characteristic for CO_2 storage in deep saline aquifers that, together with CO_2 buoyancy, viscosity of the two fluids, and capillary forces, controls the capacity and efficiency of CO_2 storage, and CO_2 injectivity and pressure build-up in the saline aquifer, and generally affects the long-term behavior of CO_2 in the storage unit [1] (medium heterogeneity is another factor that significantly affects all these storage parameters). Yet, despite its importance, no measurements of the relative permeability for CO_2/brine systems were performed until the early 2000's when CO_2 capture and storage became an important climate mitigation strategy. Sensing a gap in knowledge, Bennion and Bachu performed, using the unsteady-state method, and published in a series of SPE papers in the early-to-mid 2000's a series of 14 relative permeability tests at the respective in-situ conditions for CO_2/brine systems for sandstone, carbonate, shale and anhydrite rocks from the Alberta basin in central Alberta, Canada, that were subsequently summarized in 2008 [2]. Of these 14 tests, seven present only drainage characteristics, and the other seven present both drainage and imbibition characteristics. Between 2008 and 2010 Bennion and Bachu run a second relative-permeability testing program for both drainage and imbibition cycles, focused also on rocks in central Alberta where large stationary CO_2 sources emit ~130 Mt CO_2/year, with the purpose of covering all the potential storage aquifers in the stratigraphic succession that were not covered in the previous testing program. The results of new testing on eight carbonate rocks in this second program were compiled together with the results of testing on five carbonate rocks in the previous program and generalized in three broad categories linked to the absolute permeability k:1) low k (≤ 10 mD); 2) mid k ($10 < k \leq 100$ mD), and 3) high k (> 100 mD) [3]. The reasons for the generalizations were that:

- No correlation was found between the relative characteristics of CO_2/brine systems for these rocks and other commonly-measured rock properties, such as pore size, porosity and permeability;
- Relative permeability tests are expensive and time-consuming; and
- There is a need for providing industry with a means to estimate relative permeability and run numerical simulations of CO_2 storage at the site screening and selection stage, before engaging in a costly and lengthy site characterization program.

Between 2009 and 2012 a Stanford University team published results on relative permeability testing for CO_2/water systems using the steady-state method for the drainage cycle at conditions of 9 MPa and 12.4 MPa pressure and 50 ºC temperature on five samples from Berea, Mt. Simon and Tuscaloosa sandstones in the United States, and Otway and Paaratte sandstones in Australia [4-6]. Although Müller [7] has identified that gravitational segregation in a horizontal experimental set-up and capillary end effects may affect the test results, the Stanford University team has reached the conclusion, using CT scans of the CO_2-flooded core, that there are no gravity effects during testing even in the case of high absolute permeability (k ~ 1 D) [4], nor in the case of CO_2 exsolution [5], and that there are no experimental capillary end effects [4]. The Stanford team reached the conclusion that sub-core scale heterogeneity and rock structure and mineralogy, hence wettability, impact the relative permeability characteristics of CO_2/brine systems [4, 6] as identified by Müller [7]. The Stanford University team observed in general low relative permeability for CO_2 and higher for brine, and low end-point CO_2 saturations [4, 7], similarly to the results of Bennion and Bachu [2, 3].

During a recent study of CO_2 storage capacity and effects of CO_2 storage in the Basal Aquifer that overlies the Precambrian basement on the Alberta and Williston basin in Canada, relative permeability tests were performed on three core samples from the Basal Cambrian Sandstone in the Alberta basin and three core samples from the Deadwood Fm. (Basal Cambrian Sandstone equivalent) in the Williston basin. The results of these tests and of 16 previously unreported tests on sandstone rocks from central

Alberta are reported here, thus almost doubling the database of relative permeability testing reported previously by Bennion and Bachu [2, 3] and the Stanford University team [4-6].

Mathias et al. [8] have analyzed all the 25 sets of relative permeability data for CO_2/brine systems during drainage published by Bennion and Bachu [2, 3] and the Stanford University team [4-6], and the effects of relative permeability characteristics on CO_2 injectivity, and reached the conclusions that:

- There is no difference between the steady-state and unsteady-state measurement methods used in obtaining the two data sets;
- There is no link between relative permeability characteristics and rock porosity and/or permeability, similar to the findings of Bennion and Bachu [2, 3];
- There is no marked difference between sandstone and carbonate rocks;
- There is no link (during the drainage cycle) between the pressure increase as a result of injection, and lithology, permeability, porosity and/or interfacial tension (IFT) of CO_2 and brine; and
- Injectivity uncertainty due to relative permeability can be as high as close to 60% for open aquifers and for low-permeability ($k < 50$ mD) closed aquifers, and as low as 6% for high permeability ($k > 100$ mD) closed aquifers, where compressibility effects are more significant than relative permeability effects.

Some of these conclusions are reinforced by the new data set presented in this paper.

2. Results of Relative Permeability Testing on Sandstone Samples from Western Canada

Because of space limitations, the reader is sent to [2] for a description of the experimental set-up, techniques and methodology used in the experiments. Suffice to say that the unsteady-state experimental method was used. Each core sample, selected for general homogeneity and absence of micro-fractures based on visual inspection, and extracted carefully to maintain its mechanical integrity [7], was prepared for testing at the respective in situ conditions of temperature, pressure and brine salinity according to the methodology described in [2]. The pore size distribution, porosity and capillary pressures were measured using a mercury/air system (MICP), which was shown to be appropriate for CO_2/water systems [9]. Interfacial tension and permeability to brine were measured before running the relative permeability tests. Using a methodology similar to [10], the data were fitted through matching to the following Brooks-Corey equations [11]:

$$k_{rb} = k_{rb}^{max}\left(\frac{1 - S_{CO2} - S_b^{irr}}{1 - S_{CO2}^{c} - S_b^{irr}}\right)^{m} \qquad (1)$$

$$k_{rCO2} = k_{rCO2}^{max}\left(\frac{S_{CO2} - S_{CO2}^{c}}{1 - S_{CO2}^{c} - S_b^{irr}}\right)^{n} \qquad (2)$$

to estimate the power-law coefficients *m* and *n* (known also as Corey's coefficients) for both the drainage and imbibition cycles. In the above equations k is permeability, S is saturation, the subscripts *r*, *b* and CO_2 stand for relative, brine and CO_2, respectively, and the superscripts *max*, *irr* and *c* stand for maximum, irreducible (residual) and critical, respectively. For the drainage cycle, $k_{rb}^{max}=1$ and $S_{CO2}^{c}=0$, while for the imbibition cycle $S_{CO2}^{c}=S_{CO2}^{irr}$. The power-law coefficients *m* and *n* affect the shape of the relative permeability curve, which is linear for values equal to unity, and becomes increasingly concave with increasing values of *m* and *n*. In addition, the trapping efficiency was calculated as well. According to Land's trapping model [12], widely used in petroleum engineering, the residually-trapped non-wetting phase (CO_2 in this case) is a function of the maximum non-wetting phase saturation achieved at the end of the drainage cycle according to:

$$S_{CO2}^{irr} = \frac{S_{CO2}^{max}}{1 + CS_{CO2}^{max}} \tag{3}$$

hence the value of Land's coefficient C (dimensionless) can be easily calculated according to:

$$C = \frac{1}{S_{CO2}^{irr}} - \frac{1}{S_{CO2}^{max}} \tag{4}$$

The coefficient C provides a measure of trapping efficiency, whereby for C=0 all the CO_2 is trapped, while for $C \rightarrow \infty$ no CO_2 is trapped. Another measure of trapping efficiency was recently introduced [13] as the ratio of CO_2 saturation at the end of the imbibition cycle, S_{CO2}^{irr}, to the maximum CO_2 saturation at the end of the drainage cycle, S_{CO2}^{max}. This measure of trapping efficiency is related to Land's coefficient according to:

$$\frac{S_{CO2}^{irr}}{S_{CO2}^{max}} = \frac{1}{1 + CS_{CO2}^{max}} \tag{5}$$

but it is more intuitive in use and interpretation and it will be used here.

Table 1 presents the in-situ conditions and corresponding interfacial tension (IFT) for the new sandstone core samples from western Canada tested for CO_2/brine relative permeability characteristics.

Table 1: In-situ conditions for sandstone rock samples from western Canada used in the analysis of relative permeability and displacement characteristics of CO_2/brine systems reported in this paper.

Unit	*Depth (m)*	*Pressure (kPa)*	*Temperature (º C)*	*Salinity (mg/L)*	*IFT (mN/m)*
Viking Fm.* #3	941.87	8,600	35	28,300	32.1
Clearwater Fm.	398.80	2,000	20	14,488	56.3
Ellerslie Fm.* #2	941.94	10,900	40	97,200	32.5
Rock Creek Fm.	1793.34	15,400	75	42,300	32.9
Halfway Fm.	2049.58	21,800	74	191,300	34.6
Belloy Fm.	1823.34	13,500	56	92,200	33.1
Graminia Fm.	1064.51	9,600	39	112,014	35.1
Gilwood Fm.	1988.44	16,800	46	243,134	35.7
Basal Cambrian Ss* #2	2704.53	27,000	75	248,000	35.6
Basal Cambrian Ss #3	2129.12	18,000	58	49,680	29.5
Basal Cambrian Ss #4	2208.31	20330	60	103,450	30.2
Basal Cambrian Ss #5	2732.33	27,000	75	248.000	35.6
Deadwood Fm. #1	1648.04	15,244	47	90,684	36.1
Deadwood Fm. #2	2559.39	23,610	65	31,050	30.3
Deadwood Fm. #3	2102.40	22,300	61	167,936	31.9
Granite Wash	1,740.85	14,660	54	223,518	37.0

In this and following tables the samples are listed in descending stratigraphic order, regardless of depth, from the shallowest unit to the deepest unit overlying the Precambrian basement (the Granite Wash overlies the Precambrian basement in the northern part of the Alberta basin where the Basal Cambrian is absent due to pre-Devonian erosion, and the Deadwood Fm. is the equivalent of the Basal Cambrian in the Williston basin). Samples Viking #3, Ellerslie #2 and Basal Cambrian #2 (marked with an *) are numbered as such to differentiate them from the samples tested and reported previously [2]. These samples were tested at the same conditions of pressure, temperature and water salinity (Table 1) as in the case of corresponding sample from the same formation reported in [2], for the purpose of comparing the effects of differences in rock properties. The in situ conditions of pressure and temperature correspond to supercritical CO_2 phase for all samples except for the Clearwater sample, which was extracted from a relatively shallow core and for which the pressure and temperature conditions correspond to gaseous phase (Table 1). The IFT values for the Viking #3, Ellerslie #2, Basal Cambrian #2 and Clearwater samples were measured in the laboratory and reported in [2] for the first three samples. To save experiment time, the IFT values for all the other 12 tests were calculated based on the very good correlations developed between IFT and pressure, temperature and brine salinity for CO_2/brine systems [14]. The fact that all tests except for the Clearwater sample were performed for supercritical CO_2 is reflected in the corresponding IFT values, which vary in the 21.9 to 37 mN/m range for all samples, except for Clearwater for which the IFT is 56.3 mN/m. Table 2 below presents the pore characteristics and threshold capillary pressure for these core samples as measured in the laboratory using MICP.

Table 2: Pore characteristics of the rock samples listed in Table 1 used in the analysis of relative permeability and displacement characteristics of CO_2/brine systems.

Unit	*% Micro Porosity*	*% Meso Porosity*	*% Macro Porosity*	*Median Pore Size (μm)*	*Porosity (%)*	*Threshold Capillary Pressure (kPa)*
Viking Fm. #3	8.65	2.95	88.4	29.80	17.2	21.3
Clearwater Fm.	55.10	13.40	31.5	0.84	33.1	49.0
Ellerslie Fm. #2	5.58	2.42	92.0	25.80	29.0	28.2
Rock Creek Fm.	8.85	6.25	84.9	12.60	14.5	62.4
Halfway Fm.	15.80	11.80	72.4	8.81	17.7	69.4
Belloy Fm.	27.40	9.80	62.8	11.30	23.6	35.0
Graminia Fm.	4.60	2.90	92.5	20.60	31.6	35.1
Gilwood Fm.	54.3	17.4	28.3	0.89	11.5	193.0
Basal Cambrian Ss #2	97.50	2.50	0.0	0.21	11.6	963.0
Basal Cambrian Ss #3	3.30	5.20	91.5	15.23	11.9	48.7
Basal Cambrian Ss #4	6.40	7.30	86.3	13.67	11.9	41.2
Basal Cambrian Ss #5	89.60	10.40	0.0	0.25	12.5	618.0
Deadwood Fm. #1	7.80	12.90	79.3	11.30	17.6	28.0
Deadwood Fm. #2	27.30	5.90	66.8	13.20	16.2	28.0
Deadwood Fm. #3	7.20	6.50	86.3	15.96	19.3	3.7
Granite Wash	42.5	11.5	46.0	2.105	14.8	28.0

Tables 4 and 5 present the relative permeability characteristics for both drainage and imbibition cycles, as well as the trapping efficiency (eq. 4) for the 16 newly-tested samples. These results have been subsequently combined with the six measurements for sandstone rocks reported previously [2]: Viking #1, Ellerslie #1 and Basal Cambrian #1 for which only drainage data were measured, and Cardium #1 and #2, and Viking #2, for which both drainage and imbibition data were measured, to form a data set of 22 relative permeability measurements for CO_2/brine systems for sandstone rocks in western Canada.

Table 3: Relative permeability and displacement characteristics for the drainage cycle in CO_2/brine systems for the rock samples from western Canada whose characteristics are reported in Tables 1 and 2, respectively.

Rock Sample	***k_{brine} @ 100% Saturation (mD)***	***$k_{r\ CO2}$ @ Irreducible Brine Saturation***	***Irreducible S_b***	***Corey Parameter m for Brine***	***Corey Parameter n for CO_2***
Viking Fm #3	1558.65	0.0973	0.6010	1.33	4.34
Clearwater Fm.	0.0164	0.4939	0.3430	1.24	1.60
Ellerslie Fm. #2	3812.36	0.5735	0.3820	1.18	4.79
Rock Creek Fm	65.03	0.0434	0.4790	2.19	1.90
Halfway Fm.	54.23	0.2733	0.4660	3.12	3.48
Belloy Fm.	536.60	0.0762	0.6530	1.67	5.22
Graminia Fm.	133.90	0.1461	0.4420	1.42	4.98
Gilwood Fm.	0.749	0.5454	0.5655	1.75	3.73
Basal Cambrian Ss #2	0.0057	0.2105	0.5690	1.45	3.89
Basal Cambrian Ss #3	252.50	0.1562	0.4900	1.63	1.35
Basal Cambrian Ss #4	157.80	0.2100	0.6510	4.54	3.74
Basal Cambrian Ss #5	0.03	0.3255	0.2750	1.21	5.48
Deadwood Fm. #1	103.66	0.1062	0.4897	1.80	7.00
Deadwood Fm. #2	69.11	0.0941	0.5959	1.50	4.00
Deadwood Fm. #3	137.90	0.2597	0.6540	1.20	6.57
Granite Wash	70.13	0.4050	0.5789	1.15	1.81

Table 4: Relative permeability and displacement characteristics for the imbibition cycle in CO_2/brine systems for rock samples from western Canada whose characteristics are reported in Tables 1 and 2, respectively.

Rock Sample	***$k_{r\ brine}$ @ Irreducible Gas Saturation***	***Irreducible S_{CO2} (trapped gas)***	***Corey Parameter m for Brine***	***Corey Parameter n for CO_2***	***Trapping Efficiency***
Viking Fm.* #3	0.5191	0.223	1.27	2.53	0.559
Clearwater Fm.	0.7683	0.145	1.15	2.25	0.221
Ellerslie Fm.* #2	0.2437	0.421	1.01	2.67	0.681
Rock Creek Fm.	0.0257	0.477	1.35	3.09	0.916
Halfway Fm.	0.0278	0.459	1.01	1.94	0.860
Belloy Fm.	0.0741	0.283	2.55	3.90	0.816
Graminia Fm.	0.0913	0.383	2.11	1.67	0.686
Gilwood Fm.	0.0654	0.3592	2.03	1.15	0.827
Basal Cambrian #2	0.3333	0.2339	1.25	3.01	0.543
Basal Cambrian #3	0.1549	0.4030	1.38	1.29	0.790
Basal Cambrian #4	0.2549	0.2690	1.45	1.41	0.771
Basal Cambrian #5	0.1779	0.5190	1.71	2.11	0.716
Deadwood #1	0.3950	0.3820	3.00	2.50	0.749
Deadwood #2	0.3722	0.2883	4.00	1.78	0.713
Deadwood #3	0.2424	0.2380	2.12	1.20	0.688
Granite Wash	0.1688	0.2256	1.05	1.45	0.536

3. Discussion and Summary

Relationships were sought for these 16 new and 6 previously-reported tests [2] first for intrinsic rock properties such as pore size distributions, porosity, threshold capillary pressure and permeability. Not all relative-permeability displacement characteristics could be determined and/or correlated for the three tests reported previously for which measurements were made only for the drainage cycle [2], such as irreducible CO_2 saturation, trapping efficiency and Land coefficient.

No correlation was found between porosity or threshold capillary pressure and pore size distribution, although microporosity, for example, varies between 3.3% and 97.5%. No relationship was found between absolute permeability and porosity either, but a relatively good correlation was found between absolute permeability and median pore size, of the form:

$$k = 0.507575 \times (\mathit{median\ pore\ size})^{2.197} \qquad R^2=0.8611 \tag{6}$$

No clear, direct relationships or dependencies were found between any of the relative permeability characteristics of the CO_2/brine systems (irreducible saturations, relative permeability at irreducible saturations, Corey coefficients, trapping efficiency, Land coefficient) and any of the commonly-measured rock petrophysical properties (pore size distributions, porosity and absolute permeability) or IFT, confirming the findings of [8] based on results reported in [2] and [4-6]. Generally the data display a broad scatter in a wide range. CO_2 irreducible saturation varies in the range 0.102 to 0.519, with an average of 0.314, comparable with results obtained by [4-6, 13, 15, 16]. However, the irreducible brine saturation (Table 3) broadly increases with increasing absolute permeability, likely due to channeling and bypassing of zones of lower permeability in the core. Trapping efficiency (Table 4) also displays a broad trend of increasing in value with increasing absolute permeability (Fig. 1a). Except for two very high values of 5.37 and 8.56, the Land coefficient varies between 0.177 and 2.213, with eleven values less than 1 and six values greater than 1, and with an average for these 17 cases of C = 1.054. Excluding the two cases with very high values for the Land coefficient, there is a broad trend of increasing CO_2 irreducible saturation with increasing maximum CO_2 saturation (Fig. 1b), as noted also by [6]. However, no relationship was found between maximum CO_2 saturation or the Land coefficient and absolute permeability, as found by [6] on four tested sandstone samples.

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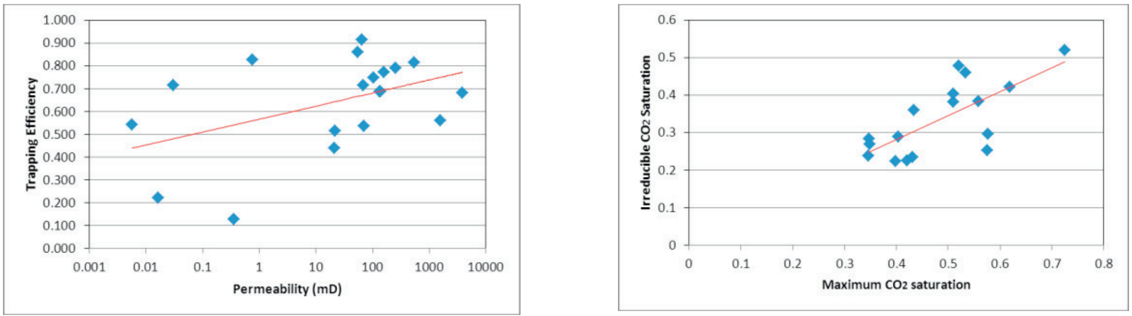

a)

b)

Fig. 1. Observed broad trends of relative permeability characteristics for the new and previously tested samples: (a) trapping efficiency as defined by eq. (5) and permeability (19 tests), and (b) irreducible versus maximum CO_2 saturations (17 tests).

It seems that in both cases of carbonate and sandstone rocks the relative permeability of the CO_2/brine system depends on rock fabric and structure, and likely wettability, more than any other measurable parameter. However, a broad pattern of dependency on permeability range was noticed for these sandstone rocks, as summarized in Tables 5 and 6, similarly to the findings reported in [3] for carbonate rocks.

Table 5: Relative permeability and displacement characteristics for the drainage cycle for the four categories of sandstone rocks, established based on the analysis of 22 core samples from western Canada.

Rock Group	**Permeability (mD)**	**Median Pore Size (µm)**	**$k_{r\,CO2}$ @ Irreducible Brine Saturation**	**Irreducible S_b**	**Corey Parameter m for brine**	**Corey Parameter n for CO_2**
Very low *k*	*k*<0.1	0.555	0.394	0.370	1.43	3.99
Low *k*	0.1<*k*10	1.123	0.380	0.495	2.01	2.71
Mid *k*	10<*k*<100	8.628	0.201	0.495	1.81	2.55
High *k*	100<*k*<500	15.346	0.176	0.545	2.12	4.73
Very high *k*	*k*>500	20.300	0.249	0.545	1.39	4.78

Table 6: Relative permeability and displacement characteristics for the imbibition cycle and trapping efficiency for the four categories of sandstone rocks, based on the analysis of 22 core samples from western Canada.

Rock Group	**Permeability (mD)**	**$k_{r\,brine}$ @ Irreducible Gas Saturation**	**$S_{CO2\text{-}irr}$ (trapped gas)**	**Corey Parameter m for Brine**	**Corey Parameter n for CO_2**	**Trapping Efficiency**
Very Low *k*	*k*<0.1	0.427	0.299	1.37	2.46	0.49
Low *k*	0.1<*k*10	0.485	0.231	1.62	1.18	0.48
Mid *k*	10<*k*<100	0.204	0.333	1.90	2.79	0.66
High *k*	100<*k*<500	0.228	0.335	2.01	1.61	0.74
Very high *k*	*k*>500	0.579	0.309	1.61	3.03	0.69

For the broad categories defined in Tables 5 and 6, it appears that the best CO_2 trapping efficiency in intragranular sandstones is achieved in the permeability range of tens to a few hundred mD. Slightly lower efficiency is achieved for very high permeability, likely due to channelling and bypassing effects, and even lower trapping efficiency is achieved for low permeability, where displacement efficiency is likely impeded by small pore throats and size. The trend of increasing irreducible brine saturation with increasing absolute permeability (Table 3) is even more evident at the rock group level. There is also a trend of decreasing trapping efficiency with increasing maximum CO_2 saturation, as noted in [13], although this trend is not apparent at the level of the individual data.

The results of these 22 measurements on sandstone rocks and of the previously reported results for 13 carbonate rocks [3] indicate that the relative permeability displacement characteristics of CO_2/brine systems at in-situ conditions are highly site specific, depending most likely of rock fabric, structure and wettability, and cannot be predicted based on any other commonly measured rock characteristics such as pore characteristics, porosity and absolute permeability, with subsequent implications for upscaling, numerical modeling and history matching of observed CO_2 plume behaviour. No significant difference between the tested 13 carbonate and 22 sandstone rocks has been observed, as noticed by [8] for the smaller data sample of [2] and [4-6].

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