

ENHANCED CHEMICAL WEAKENING OF CHALK DUE TO INJECTION OF CO₂ ENRICHED WATER

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ABSTRACT

Chalk deformation and especially the so called water weakening effect induced by seawater injection, which caused a dramatic increase in subsidence rate in the early 90s at the Ekofisk field, has been extensively studied worldwide. Potential CO₂ injection into depleted chalk reservoirs will lead to acidification of the formation brine or pore-water itself; since the solubility of carbonates is strongly pH dependent, enhanced water weakening should thus be expected.

CO₂ related chemical weakening of chalk has been experimentally verified at the laboratory of University of Stavanger. By use of a standard triaxial cell, series of chalk cores were isotropically loaded beyond yield and thereafter left to creep at constant effective stresses between 12 and 14 MPa. All experiments were performed at ambient temperature, and the cores were exposed to alternating flooding with and without CO₂ enriched fluids, distilled water, seawater, and/or seawater with 4 times the concentration of SO₄²⁻, at flow rates of 0.05 ml/min, i.e., 2 pore volumes (PV) per day, and a backpressure of 10 bar.

The tested chalk cores exposed to CO₂ enriched brines showed an average increase in creep strain rates of 19% compared to CO₂ enriched distilled water. Increasing the partial pressure of CO₂ in the brine leads to a higher equilibrium concentration of Ca²⁺. An increased equilibrium concentration of Ca²⁺ triggers two processes: Firstly, dissolution of the calcitic (CaCO₃) matrix, and secondly brine supersaturation with respect to gypsum (CaSO₄×2H₂O). When gypsum is precipitated, more Ca²⁺ needs to go into solution for the water to be in equilibrium with the core. Thus, the dissolution and water weakening effect of the chalk is not only dependent on the pH, but also the sulphate concentration in the brine.

From our work, it seems that long time storage of CO₂ could be challenging in chalk. One may also conclude that CO₂ used as an EOR fluid in chalk can trigger significantly increased compaction. The enhanced weakening to be expected when chalk is exposed to CO₂ injection will thus be governed by the reservoir conditions – temperature, in situ stresses, CO₂ phase and the fluids in which CO₂ dissolves. In order to successfully implement a CO₂ flood or storage project in chalk, the aspects related to significant

compaction, as well as well design and surface facilities, topics not covered in this study, need to be considered accordingly. If these issues are properly managed the compaction can result in significant enhanced oil recovery from chemically induced compaction.

INTRODUCTION

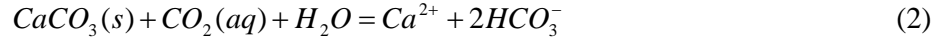
The reduction of industrial CO₂ emissions is considered to be one of the main challenges in order to stabilize the atmospheric CO₂ concentration. Large quantities of CO₂ can be stored subsurface, and the three main storing options are saline aquifers, existing gas and oil fields and unmineable coal seams (IPCC, 2005). The use of gas and oil fields has also economic benefits through enhanced oil recovery (EOR). Therefore, there is an increasing interest in the petroleum industry to use CO₂ in EOR projects and also to store CO₂ in depleted reservoirs after oil or gas production has stopped. CO₂ injection has been the most important EOR process in U.S. carbonate reservoirs since the early 1980's (Moritis, 2004).

Several issues have to be taken into account when a CO₂ flood is implemented in an offshore chalk reservoir, such as reservoir compaction and seabed subsidence. The Ekofisk field, a large chalk reservoir in the southern part of the North Sea, has experienced strong compaction during primary production and subsequent water flooding. Compaction observed at the Ekofisk field during the water flood, even though the reservoir was re-pressurized, has been termed the "water weakening effect of chalk". Total seabed subsidence at the Ekofisk field is almost 9 meters (Spencer et al., 2008). Different mechanisms have been proposed in order to explain this water weakening effect: physical (Andersen and Foged, 1992; Delage et al., 1996), physico-chemical (Risnes et al., 2005), and purely chemical effects (Hellmann et al., 2002; Heggheim et al., 2005; Korsnes et al., 2006).

CO₂ flooding in hydrocarbon reservoirs is generally expected to take place at depths below 800 m, where the in-situ pressures and temperatures will usually result in CO₂ being in a liquid or supercritical state (IPCC, 2005). In a water flooded reservoir, the CO₂ will come in contact with the brine, and dissolution of CO₂ into the water will take place. The properties of these mixtures can be modelled using Equations of State. When injecting CO₂ into a chalk reservoir, it is important to estimate the concentration of aqueous CO₂ in solution because of the importance of CO₂ (aq)-rock interactions in chalk. Knowledge of the solubility of CO₂ in brines with various ionic strength, as well as reservoir temperature and pressure is therefore of great importance.

The chemical weakening taking place when chalk is exposed to CO₂ injection is affected by the reservoir conditions since the solubility of CO₂ (g) in water is governed by the partial pressures, brine composition (ionic strength) and temperature. The solubility of CO₂ (g) in water increases with increasing partial pressure, but decreases with increasing ionic strength as well as temperature. The solubility of the chalk will therefore be a function of the partial pressure of CO₂, brine composition, and temperature.

In the reservoir, CO₂ will dissolve into the water and form carbonic acid (H₂CO₃) and subsequently bicarbonate (HCO₃⁻), acting as a weak acid and dissolving the chalk:



This chemical reaction of carbonate rock and CO₂ (aq) may therefore induce additional reservoir compaction and subsequent seabed subsidence. In worst case scenarios, this compaction may lead to CO₂ leakage in the impermeable cap rock, loss of production wells, and jacking up of platforms anchored on the seabed. Seawater, used as flooding fluid in North Sea reservoirs, contains cations (e.g., Ca²⁺ and Mg²⁺) that can react with bicarbonate to form stable carbonates:



Dissolution of chalk will increase the concentration of Ca²⁺ and HCO₃⁻ in the pore-water. In seawater flooded chalk reservoirs, this increase in Ca²⁺ can cause precipitation of CaSO₄ (s):



The solubility of CaSO₄ decreases with increasing temperature, i.e., precipitation increases with increasing temperature. At the Ekofisk field, where the reservoir temperature is 130 °C, CaSO₄ (s) is supersaturated in seawater (Azaroual et al., 2004). In the production wells, this is observed by a large retention of SO₄²⁻ ions due to precipitation of CaSO₄ (s) at seawater breakthrough (Petrovich and Hamouda, 1998; Mackay and Jordan, 2003). If dissolved Ca²⁺ is removed from the equilibrium solution by precipitation of CaSO₄ (s), then this reaction will promote even more dissolution of chalk, which may result in an even stronger water weakening effect.

Some experimental and modelling studies have been performed on the chemical weakening of chalk/limestone by CO₂ flooding. Schroeder et al. (2001) reported that injection of supercritical CO₂ had no significant effect on the mechanical strength of chalk, while Madland et al. (2006) observed that chalk became considerably weaker when flooded with carbonated water at stress levels beyond the yield point. Laursen (2007) performed unconfined compression tests on three different chalk types with varying porosity that had been pre-flooded with carbonated water in a core holder. The results from that study were not definitive, but the results tentatively indicated that flooding of carbonated water had an effect on mechanical properties such as the elasticity modulus and shear strength. A modelling study on the rate of pressure solution creep in limestone related to CO₂

sequestration showed that high partial pressures of dissolved CO₂ (up to 30 MPa) increased compaction rates by a factor of 50 to 75 (Renard et al., 2005).

The present work considers enhanced chemical water weakening of chalk due to flooding of CO₂ enriched water. It addresses the question whether chemical water weakening is affected by the ion composition of the CO₂ enriched water.

EXPERIMENTAL

Chalk Material

Outcrop chalk from the quarry of Roerdal, near Aalborg, Denmark was used in this study. Petrophysical and mineralogical studies have shown that Roerdal chalk is a suitable geomechanical analogue to the Valhall field chalk in the southern part of the North Sea (Hjuler, 2007). Roerdal chalk is of Late Cretaceous age, has a relatively high porosity (43-48%), permeabilities of 3-5 mD, and a silica content in the range of 2-7%, typically 4%.

Equipment and Test Procedure

The triaxial cell used for the mechanical test is basically a hydraulically operated test cell as described by Korsnes et al. (2006). Two pressure vessels were used to flood the injection fluids during mechanical testing; one pressure vessel containing the carbonated fluid and the other pressure vessel containing the same fluid without CO₂.

CO₂ saturated water was prepared in a pressure vessel with 8 bar partial pressure. Three different fluids were enriched with CO₂: distilled water (DW), synthetic seawater (SSW), and synthetic seawater (SSW4) with a sulphate concentration 4 times higher than the sulphate concentration in SSW, Table 1. All samples were saturated with DW, SSW or SSW4 without CO₂ prior to mechanical testing.

Table 1. Compositions of saturating brines.

Ions	SSW [mole/l]	SSW4 [mole/l]
HCO ₃ ⁻	0.002	0.002
Cl ⁻	0.525	0.343
SO ₄ ²⁻	0.024	0.096
Mg ²⁺	0.045	0.045
Ca ²⁺	0.013	0.013
Na ⁺	0.450	0.419
K ⁺	0.010	0.010
TDS [g/l]	33.39	33.39

Hydrostatic tests with an additional creep phase at ambient temperature were performed. During flooding, the water was injected axially from the bottom of the sample, and the flooding rate was 0.05 ml/min (approximately 2 pore volumes per day) for each test. All tests were performed with continuous flooding during the entire test period.

Before the hydrostatic loading phase, the effective stress was set to 0.5 MPa with the pore pressure set to 0.8 MPa and the confining pressure set to 1.3 MPa. All samples were hydrostatically loaded beyond yield, 12 to 14 MPa, before the cores were left to creep. In the creep phase, alternating flooding periods with non-carbonated and carbonated water were performed in order to study the chalk samples' creep response.

RESULTS

Hydrostatic Loading Phase

The values of hydrostatic yield strength and bulk modulus were determined before the subsequent creep phase. As expected, at ambient temperature no significant differences in yield points and bulk moduli were observed when using the three different brines, Table 2. Recent published studies have, however, shown that the specific ion composition of various brines injected at elevated testing temperature will have a significant effect on the mechanical strength of chalk. Korsnes et al. (2006) showed that the hydrostatic yield point for high porosity (48-50%) outcrop chalk cores from Stevns Klint, Denmark, was increased by a factor of 1.3 when flooded with DW compared to cores flooded with SSW and SSW2 (sulphate concentration 2 times higher than the sulphate concentration in SSW) when tested at 130 °C. The suggested weakening mechanism was explained as an ion exchange process between Mg^{2+} ions from solution and Ca^{2+} ions in the thin water films close to intergranular contacts in the presence of SO_4^{2-} . Another weakening process is the precipitation of $CaSO_4$ (s) from seawater supersaturated at 130°C, inducing chalk dissolution due to the loss of Ca^{2+} ions in the equilibrium solution. Considering the actual brines used in the present study; at ambient temperature these processes should not affect the obtained values of yield strength and bulk modulus.

Table 2. Results from hydrostatic loading phase at ambient temperature.

Sample no.	Porosity [%]	Yield point [MPa]	Bulk-modulus [MPa]	Injected fluid
AH 10	45.5	8.7	0.62	SSW4
AH 12	43.4	8.7	0.66	SSW4
AH 13	44.6	8.5	0.58	SSW
AH 14	44.4	8.3	0.65	SSW
AH 15	44.4	8.8	0.55	DW

Creep by Alternating Flooding Phases

Creep with alternating flooding of non-carbonated and carbonated brines was studied. The non-carbonated brines were injected in the first part of the creep phase, prior to the first flooding phase with carbonated brine. After this flooding phase, a new flooding phase with non-carbonated brines was begun, and thereafter the tests were ended with a second flooding phase with carbonated brines, as seen in Figure 1.

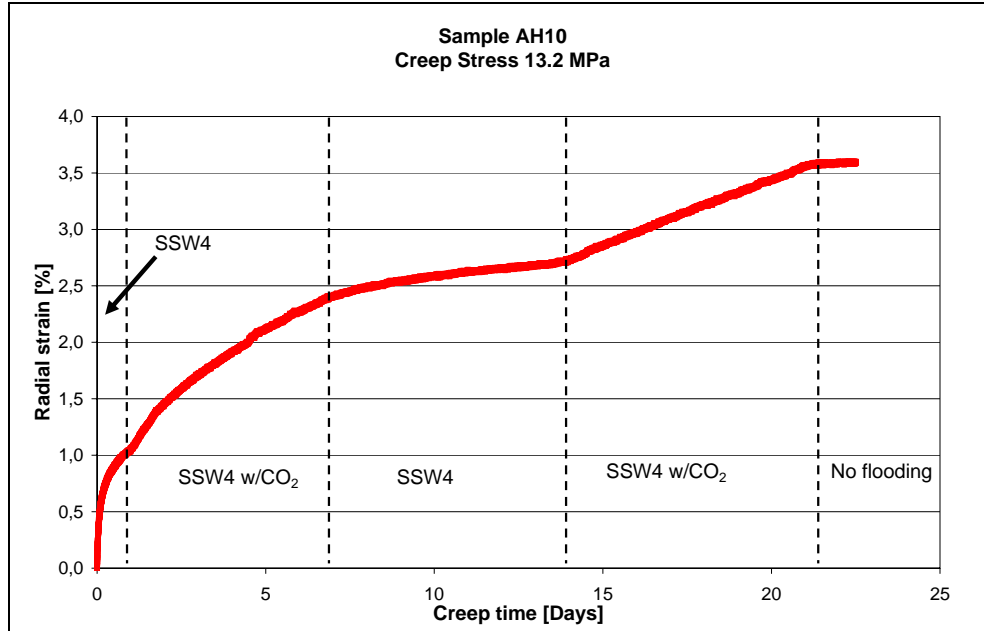


Figure 1. Strain vs. time for sample AH10 subsequently flooded with SSW4 and CO₂ enriched SSW4.

Creep from Flooding with Carbonated Distilled Water

One sample, AH15, was tested with alternating flooding by DW and carbonated DW. Creep stress was set to 14 MPa and the entire creep phase lasted more than 20 days. DW was flooded for 1.7 days, followed by 7 days of flooding by carbonated DW. A second flooding with DW for 5 days followed, before the test was ended with another flooding by carbonated DW that lasted 6.9 days, Table 3. Initially, in the primary creep phase of the first flooding period, strain increased rapidly, as shown in Figure 1 for sample AH10. The obtained strain values from the first flooding period will therefore not be used in this study when comparing strain rates on chalk cores flooded with and without CO₂ saturated brines. Axial creep strain during the first flooding phase with CO₂ was 0.09 percent per pore volume (%/PV). This strain rate was reduced by a factor of 2.2 when injection of DW without CO₂ was conducted (0.04 %/PV). The strain rate increased to 0.06 %/PV in the last flooding phase with CO₂. As shown in Table 3, strain rate from the second flooding period with CO₂ was less than that of the first flooding period with CO₂ (0.06 %/PV compared to 0.09 %/PV). This result is consistent with those of Madland et al. (2006).

Table 3. Experimental data from the creep phase with flooding by DW and CO₂ enriched DW.

Flooding period	Sample no.	Creep stress [MPa]	Flooding fluid	Flooding period [Days]	Pore volumes	Axial strain [%]	Axial strain [% per PV]
1	AH15	14.0	DW	1.7	3.7	0.89	0.24
2			DW ^w /CO ₂	7.0	15.1	1.36	0.09
3			DW	5.0	10.8	0.39	0.04
4			DW ^w /CO ₂	6.9	14.8	0.96	0.06

Creep from Flooding with Carbonated SSW

Two samples (AH13 and AH14) were tested with alternating flooding by SSW and carbonated SSW. For both samples, creep stress was set to 12 MPa and the entire creep phase lasted more than 20 days for sample AH13. During the test with sample AH14 several problems occurred: the data logging program stopped and an electrical power failure, lasting an entire weekend, made it impossible to interpret the results. In the creep phase for sample AH13, SSW was flooded for 1.12 days before the sample was flooded for 6.96 days by carbonated SSW. The next flooding period with SSW lasted for 4.79 days before the test was ended by a flooding period with carbonated SSW that lasted 6.35 days, Table 4. In the first flooding period with carbonated SSW, axial creep strain is 0.11 %/PV, which is an increase in creep deformation compared to the result obtained with carbonated DW, Table 3 and 4, even though the test with DW was performed at a higher creep stress, 14 MPa compared to 12 MPa with SSW. Since creep is stress dependent, one may anticipate that if these two samples had been tested with similar fluids, the sample tested at highest creep stress should have obtained highest creep strain values. The strain rate was reduced by a factor of 3.7 when the flooding fluid was changed from carbonated SSW (0.11 %/PV) to SSW (0.03 %/PV). The last flooding period with carbonated SSW increased the strain rate up to 0.07 %/PV, which again is a higher strain than obtained by carbonated DW, Table 3 and 4.

Table 4. Experimental data from the creep phase with flooding by SSW and CO₂ enriched SSW.

Flooding period	Sample no.	Creep stress [MPa]	Flooding fluid	Flooding period [Days]	Pore volumes	Axial strain [%]	Axial strain [%/PV]
1	AH13	12.0	SSW	1.12	2.47	0.70	0.28
2			SSW ^w /CO ₂	6.96	15.32	1.66	0.11
3			SSW	4.79	10.55	0.27	0.03
4			SSW ^w /CO ₂	6.35	13.97	0.93	0.07

Creep from Flooding with Carbonated SSW4

Two samples (AH10 and AH12) were tested with alternating flooding by SSW4 and carbonated SSW4. Creep stress was set to 13.2 MPa for both samples, and each was tested for more than 20 days. The duration of the different flooding periods varied: the first flooding period for sample AH10 lasted 0.86 days while it lasted 2.67 days for sample AH12, Table 5. The two subsequent flooding periods with carbonated SSW4 and SSW4 lasted approximately 2 days longer for sample AH10 than for sample AH12. In the last flooding period with carbonated SSW4, the flooding period for sample AH10 lasted 7.39 days, and 8.4 days for sample AH12, respectively, Table 5. Even though the primary creep period with SSW4 flooding was different for samples AH10 and AH12, the strain rate values obtained during the following flooding periods with carbonated SSW4 were similar. The strain rate was 0.10 %/PV for both samples during the first flooding period with carbonated SSW4, while it was somewhat lower, 0.07 (AH10) and 0.08 %/PV (AH12), during the second flooding period with carbonated SSW4, Table 5. Consider sample AH10, in the second flooding period with SSW, a problem occurred with the axial piston

movement, and the strain rate value had to be extrapolated. The axial strain rate was similar for both samples during flooding period 3 with SSW4 (0.03 %/PV), which is a reduction by a factor of 3.3 when comparing to the first flooding period with CO₂ enriched SSW4 (0.10 %/PV), Table 5.

Table 5. Experimental data from the creep phase with flooding by SSW4 and CO₂ enriched SSW4.

Flooding period	Sample no.	Creep stress [MPa]	Flooding fluid	Flooding period [Days]	Pore volumes	Axial strain [%]	Axial strain [%/PV]
1	AH10	13.2	SSW4	0.86	1.91	0.73	0.38
2			SSW4 ^w /CO ₂	5.94	13.43	1.35	0.10
3			SSW4	6.70	14.72	0.21	0.03*
4			SSW4 ^w /CO ₂	7.39	16.32	1.19	0.07
1	AH12	13.2	SSW4	2.67	5.99	1.60	0.27
2			SSW4 ^w /CO ₂	4.13	9.28	0.94	0.10
3			SSW4	4.71	10.57	0.29	0.03
4			SSW4 ^w /CO ₂	8.40	18.86	1.52	0.08

* Problem with axial piston movement.

DISCUSSION

Reservoir Compaction

The experimental results show that flooding of CO₂ enriched brines into stressed chalk cores increases the strain rates significantly compared to flooding with brines without CO₂, a factor between 1.5 and 3.3 in increased strain rates are observed when comparing the different flooding periods with and without CO₂ enriched brines. In a chalk reservoir the injected CO₂ will dissolve into the formation water and form carbonic acid, which will enhance chalk dissolution and subsequently lead to enhanced reservoir compaction. It is, however, difficult to determine how large the compaction will be in a chalk reservoir if the reservoir is used for CO₂ storage or if CO₂ is used as an EOR technique. The effects will also be affected by the reservoir conditions, such as the amount of water inside the reservoir, brine composition, partial pressure, in-situ stresses, and temperature.

Creep Comparison with Different Flooding Brines

Axial strain per pore volume for each flooding period and brine, except for the first flooding period without CO₂, is listed in Table 6. All strain rates increased when flooding with CO₂ enriched SSW and SSW4 compared to CO₂ enriched DW, but the uncertainties are high since only one sample was tested with SSW and DW, and two samples with SSW4. The samples flooded with carbonated SSW and SSW4 obtained on average 17% and 21% more strain per pore volume in the second and fourth flooding period compared to the sample flooded with carbonated DW, Table 6. These results could have been different to some degree, if all tests had been performed at the same creep stress. Samples flooded with SSW and SSW4 were tested with a creep stress of 2.0 and 0.8 MPa, respectively, lower than the sample flooded with DW. In general an increase in creep stress will induce higher creep rates. The strain per pore volume flooded is lower for all brines in

the second flooding period with carbonated water, Table 6, which agrees with the results obtained by Madland et al. (2006).

Table 6. Creep per pore volumes flooded for all flooding fluids with and without CO₂.

Fluids	Average creep stress [MPa]	2 nd flooding period	3 rd flooding period	4 th flooding period
		Axial strain with CO ₂ [%/PV]	Axial strain without CO ₂ [%/PV]	Axial strain with CO ₂ [%/PV]
SSW4	13.2	0.10	0.03	0.075
SSW	12.0	0.11	0.03	0.07
DW	14.0	0.09	0.04	0.06

Madland et al. (2006) used the OLI System Stream Analyzer 1.3, which is a chemical model software based on thermodynamic equilibrium conditions using published experimental data, to determine the solubility of CO₂ and CaCO₃ as a function of CO₂ partial pressure and brine composition, as seen in Figure 2. SSW1 in the study by Madland et al. (2006) has the same brine composition as SSW4 in the present study. It is seen from Figure 2 that the solubility of CO₂ is higher in DW than in SSW and SSW4 at 8 bar partial pressure, but the solubility of CaCO₃ is highest with SSW4 and lowest with DW at 8 bar partial pressure. The difference in CO₂ solubility in the different waters are small, a factor of 1.1 between DW and SSW and SSW4 at 25 °C, Figure 2a. The difference is significantly higher regarding the solubility of CaCO₃; the solubility is increased by a factor of more than 2 at 8 bar partial pressure when comparing SSW4 and DW, Figure 2b.

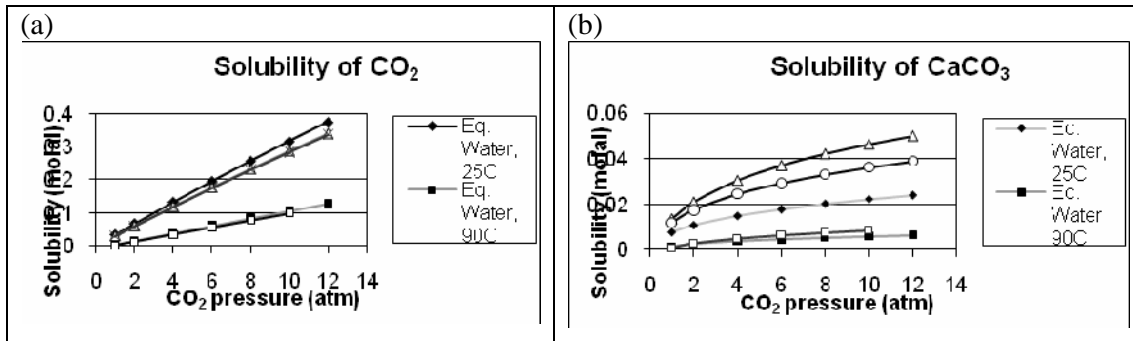


Figure 2. (a) Solubility of CO₂(g) and (b) Solubility of CaCO₃(s) at different partial pressures in different brines and temperatures (Madland et al., 2006).

From these chemical aspects, it could be anticipated that highest mechanical weakening of chalk by CO₂ flooding should take place when SSW4 is used as carbonated flooding fluid at 25 °C, Figure 2. The reaction rate, i.e., kinetics, is however important when discussing chemical weakening of chalk. Chalk dissolution increases with decreasing temperature, but due to kinetics, the dissolution rate is reduced when the temperature is lowered. More chalk is dissolved at ambient temperature than at 90°C, but the dissolution rate is faster at 90°C. The solubility of CaSO₄ decreases with increasing temperature, i.e., CaSO₄ will precipitate more easily from SSW and especially SSW4 when the temperature increases.

Precipitation of CaSO_4 will enhance chalk dissolution since one of the common ions, Ca^{2+} , is removed from the equilibrium solution. The difference in CO_2 and CaCO_3 solubility is smaller between the different brines at 90°C , Figure 2, but the additional process of CaSO_4 precipitation could enhance the chemical weakening of chalk by flooding CO_2 enriched SSW and SSW4. It is difficult to conclude if the observed increase in strain rates is due to CaSO_4 precipitation, but the acidic brine leads to a higher equilibrium concentration of Ca^{2+} , and SSW and SSW4 can be supersaturated for CaSO_4 at ambient temperatures. At ambient temperatures, CaSO_4 will precipitate as gypsum ($\text{CaSO}_4 \times 2\text{H}_2\text{O}$) (Carlberg and Matthews, 1973).

Comparison of CO_2 Floods with and without Backpressure

Madland et al. (2006) performed CO_2 floods with a chalk type, fluid compositions, temperature, and creep stress comparable to the present study, but the tests were performed without backpressure and with higher flooding rates. Without backpressure, the partial pressure of CO_2 will gradually reduce when carbonated water is flooded through the core. A comparison between average axial strain values per pore volume with CO_2 saturated injection fluids from the present study, and some randomly selected tests from the study by Madland et al. (2006) are presented in Table 7. The average strain value without backpressure is 0.053 %/PV while the average value is 0.084 %/PV with backpressure, which means that the strain values were higher with backpressure by an average factor of 1.6. Without backpressure, the gradual reduction in CO_2 partial pressure will reduce the solubility of CO_2 in the flooding brines, and thus the solubility of CaCO_3 . The pH will also increase with decreasing partial pressure. The results presented in Table 7 are therefore in line with the chemical modelling in Figure 2.

Table 7. Average axial strain values per pore volume with CO_2 saturated injection fluids.

Sample no.	[%/PV]	Flooding fluid
AM4*	0.054	Tap-water
AM5*	0.052	Tap-water
AM6*	0.077	Tap-water
A24*	0.042	Tap-water
B13*	0.044	Tap-water
B14*	0.047	Tap-water
B17*	0.058	SSW4
AH10 & 12	0.087	SSW4
AH13	0.087	SSW
AH15	0.078	DW

* Results from Madland et al. (2006)

CONCLUSIONS

Even though definitive conclusions are problematic to draw because of the limited number of tested samples, some important results are summarized as follow:

- Creep strain rates per pore volume of flooded carbonated water increased in average by a factor of 2.5 compared to non-carbonated water.
- Results point towards an enhanced chemical weakening by flooding of carbonated SSW and carbonated SSW4. Compared to carbonated DW, a 19% average increase in creep strain rates was detected from the two flooding periods with carbonated brines.
- Enhanced weakening by CO₂ enriched seawater at ambient temperature is probably linked to higher CaCO₃ solubility and precipitation of CaSO₄ (s), which will promote enhanced chalk dissolution.
- CO₂ flooding with backpressure increased strain rates per pore volume by a factor of 1.6 compared to without flooding backpressure.
- To predict the impact of CO₂ injection into chalk reservoirs the tests need to be performed at in-situ stress, pressure and temperature.

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