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Challenges and enablers for large-scale CO₂ storage in chalk formations

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Abstract

The past two decades of research on Carbon Capture and Storage (CCS) seem to have finally become fruitful as global leaders and energy-intensive industries are cooperating to materialize CCS projects and reach the promised reduction in CO₂ emissions. Traditionally, CCS projects targeted mostly high permeability sandstone formations, despite the numerous carbonate fields undergoing CO₂ injection for Enhanced Oil Recovery (EOR) in the United States or Canada. Because of the reactivity between calcite minerals and CO₂ saturated water, chalk formations, characterized by high porosity and low permeability, have been previously portrayed as infeasible CO₂ storage sites. Although previous laboratory investigations were carried out to assess the performance of CO₂-EOR in North Sea chalk fields, these studies did not result in any field-scale demonstration projects; this may soon change since a positive movement towards CO₂ storage in depleted oil fields has been recently initiated. In this work, we reviewed existing studies on CO₂ injection in chalk to address the suitability of this type of formation for CCS. Although the evidence on the thermo-hydro-mechanical-chemical behaviour of chalk in the presence of CO₂-saturated aqueous solutions is mixed, the majority of flooding tests performed on reservoir core samples do not support further weakening relative to water injection conditions nor significant changes in the petrophysical properties. Along with the weakening effect and using the Danish North Sea chalk fields as a case study, we addressed events that impact the storage site safety such as fault reactivation, and caprock and well integrity. Furthermore, monitoring techniques relevant to offshore locations are also discussed. Based on studies on other types of carbonates, and considering the characteristics of chalk (e.g., permeability, wettability, and reactivity) we analysed the relevance of different trapping mechanisms (i.e., solution, capillary, and mineral) but also several effects (i.e., chemical, biological, mechanical) that can lead to loss of injectivity. The main observations and conclusions in this work can be easily extrapolated to other chalk formations worldwide.

Keywords: CO₂ storage, chalk, weakening, trapping mechanisms, fluid-rock interactions, depleted oil fields, decarbonisation, carbon neutrality, North Sea

1. Introduction

The climate change effects on the environment are driving the implementation of measures to reduce CO₂ emissions. In 2015, most nations showed their commitment to fighting global warming by ratifying the Paris Agreement, where they agreed to adopt measures to limit the temperature increase to 1.5°C above pre-industrial levels. The involved countries engaged in developing strategies to curtail their greenhouse gas (GHG) emissions. In 2018, the Intergovernmental Panel on Climate Change (IPCC) analysed different pathways that could limit the temperature increase to the desired target of 1.5°C and recognized the important role of CO₂ capture and storage (CCS) to reduce carbon dioxide emissions

during this century (de Coninck et al., 2018; Rogelj et al., 2018); a special report from the IPCC (Abanades et al., 2005) exhaustively described the CCS technology earlier in 2005. In recent years, the number of large-scale CCS facilities has increased considerably; the storage of CO₂ is combined with enhanced oil recovery (EOR) in approximately half of these projects (Global CCS Institute, 2020). However, existing facilities are spread around only a few countries (e.g., US, Norway, UK, Australia, Canada) and the fraction of CO₂ captured and stored is still insignificant compared to the emissions overall. Although responsible for only 10% of global greenhouse gas emissions, Europe is strongly supporting research and demonstration projects to promote the commercialization of CCS (European Commission, 2018); the European Commission has designated the Northern Lights (Norway) and Porthos (Netherlands/UK), among others, projects of common interest (PCI) as they may enable Europe to reach by 2030 a 40% reduction in GHG emissions compared to 1990.

Denmark aims at reducing greenhouse gas emissions by 70% compared to the levels in 1990 (Danish Ministry of Climate, Energy and Utilities, 2019) and to become carbon-neutral by 2050 (Danish Council on Climate Change, 2020). However, the 2020 Climate and Energy Outlook (Danish Energy Agency, 2020) showed that with the existing measures, Denmark could only reduce its emissions by 44% compared to 1990, therefore lacking 26% points to achieve the 70% reduction target. The Danish Council on Climate Change recommended imposing a tax of approximately DKK 150 /t of CO₂ and CCS as a transition element to reduce CO₂ emissions by 2030 (Danish Council on Climate Change, 2020). The Danish government climate partnership (Partnerskab for energitung industri, 2020) conveyed the same message, stressing the importance of developing a national strategy for carbon capture (utilization) and storage (CCS/CCUS) and encouraging collaboration among industry partners to demonstrate the feasibility of CCS/CCUS projects. The recent North Sea agreement, which sets 2050 as the end for the Danish oil and gas production and urges the CO₂ storage in the depleted hydrocarbon fields, represents an important step towards the deployment of CCS technology in Denmark (Danish Ministry of Climate, Energy and Utilities, 2021).

For a worldwide implementation of CCS, countries should begin by identifying suitable geological sites for CO₂ storage (Danish Council on Climate Change, 2020). At these sites, the CO₂ will be trapped/contained either (i) structurally due to the presence of a sealing unit/caprock (structural trapping), (ii) through the dissolution of the CO₂ in the formation water (solution trapping), (iii) as immobile disconnected blobs formed when water re-imbibes the pore space (occupied by CO₂) by the action of capillary forces (capillary trapping), and (iv) through mineralization reactions leading to the formation of carbonate-rich minerals (mineral trapping). The Danish CO₂ storage potential has been previously assessed in several European research initiatives (Christensen and Holloway, 2004; Holloway, 1996; Røkke et al., 2016; Vangkilde-Petersen, 2009a) (Figure 1).

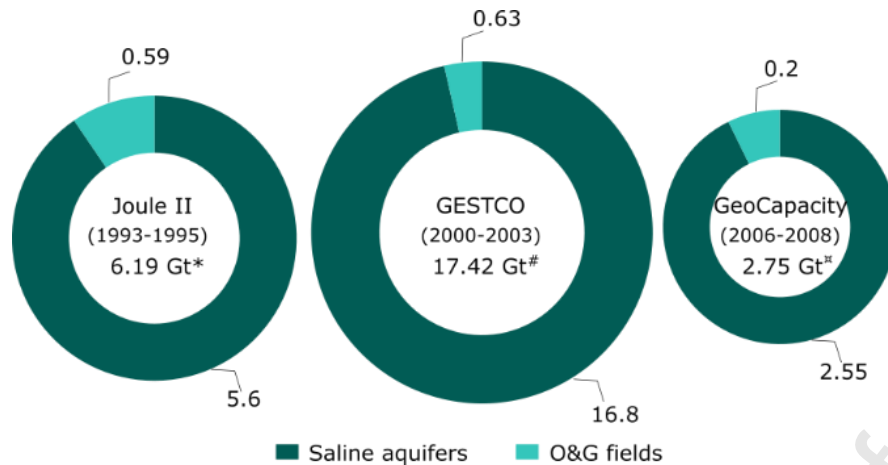


Figure 1. Denmark's CO₂ storage capacity estimated in the Joule (Holloway, 1996), GESTCO (Christensen and Holloway, 2004; Christensen and Larsen, 2004), and GeoCapacity (Vangkilde-Petersen, 2009a) projects. * The estimated capacity corresponds to 5 onshore saline aquifers and 16 hydrocarbon fields. A storage efficiency of 6% was assumed for the saline aquifers whereas for the O&G fields a volumetric replacement ratio of the produced hydrocarbon with CO₂ 1:1 was considered. # The estimated capacity corresponds to 11 onshore and nearshore saline aquifers and 16 O&G fields. A 40% storage efficiency for the saline aquifers was considered. # The estimated capacity corresponds to 11 onshore and nearshore saline aquifers and 17 O&G fields. A storage efficiency of 5-10% was assumed for the saline aquifers whereas the capacity estimation for the O&G fields considered that CO₂ can replace 25% of the volume of produced hydrocarbons.

Although the estimated storage capacity of oil fields is small compared to that of saline aquifers, the deployment of CCS in Denmark will most likely occur offshore in the depleted oil fields because of the strong opposition of both conservative and green parties against onshore storage (Rütters, H. and the CGS Europe partners, 2013). The storage capacity of Danish and Norwegian oil and gas (O&G) fields was assessed also in the NORDICCS initiative (Bergmo and Anthonsen, 2014). The calculation procedure followed the methodology used in the GeoCapacity project but assuming a 1:1 volumetric replacement ratio of hydrocarbons by CO₂. Figure 2 shows the estimated CO₂ storage capacity of the Danish O&G fields.

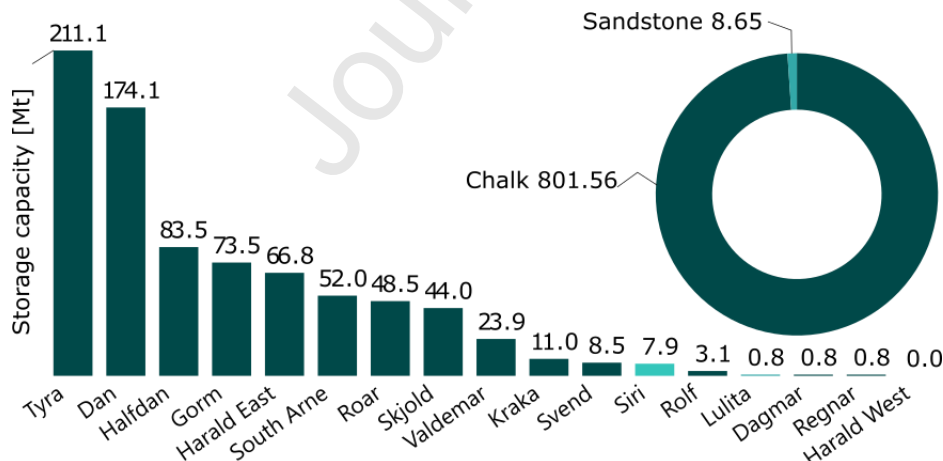


Figure 2. Storage capacity for different fields in the Danish North Sea (Bergmo and Anthonsen, 2014). The capacity was calculated by considering that the pore space availability for CO₂ storage corresponds to the volume of produced hydrocarbons. Although Harald West has available capacity for CO₂ storage, this was evaluated as 0, most likely because of a lack of data.

Since these estimations represent a static storage capacity, they are highly uncertain and may differ considerably from the (real) dynamic capacity (Anthonsen and Frykman, 2016; Lothe et al., 2015). Public Danish research institutions and industry partners were involved in several other European projects related to CCS (Jordanger and Røkke, 2009; Le Thiez, 2011; Poulsen et al., 2014). Despite the great involvement in research projects related to CCS, Denmark is still lacking demonstration/pilot projects related to storage. The country's new political strategy on climate and energy has resulted in allocating funds for the demonstration of CCS projects. Recently, the Danish Energy Technology Development and Demonstration Program (EUDP) has awarded nearly DKK 10 million to the Greensand project, which is a feasibility study of the storage of CO₂ captured at onshore industrial facilities into offshore depleted Danish oil fields. Although most hydrocarbon reservoirs in Denmark are chalk fields (Figure 2), CO₂ is planned to be stored (0.5-1 Mt/year) in a sandstone formation, the Nini field, and later expanded to the Nini East, Cecilie, and Siri fields (sandstone reservoirs) and store up to 4 Mt CO₂/year for the next 20 years (Nielsen, 2020). The concern on the reactive behaviour of chalk in presence of CO₂ (Christensen and Larsen, 2004; Korsnes et al., 2008; Vangkilde-Petersen, 2009b) and the lack of economic production from the mentioned reservoir may explain the selection of a sandstone field for this feasibility study, despite their lower storage capacity.

Upper Cretaceous to Palaeocene age chalk deposits are yet widely spread around the world representing economically important aquifers and hydrocarbon reservoirs in many places (Figure 3). In addition to the Tor and Ekofisk hydrocarbon-bearing formations, actively producing in the Dutch, UK, Norwegian, and Danish sectors of the North Sea, examples include the onshore chalk aquifers of southern and eastern England, northern France, northern Germany, Denmark, as well as hydrocarbon reservoirs located in the Austin and Niobrara chalk in the US.

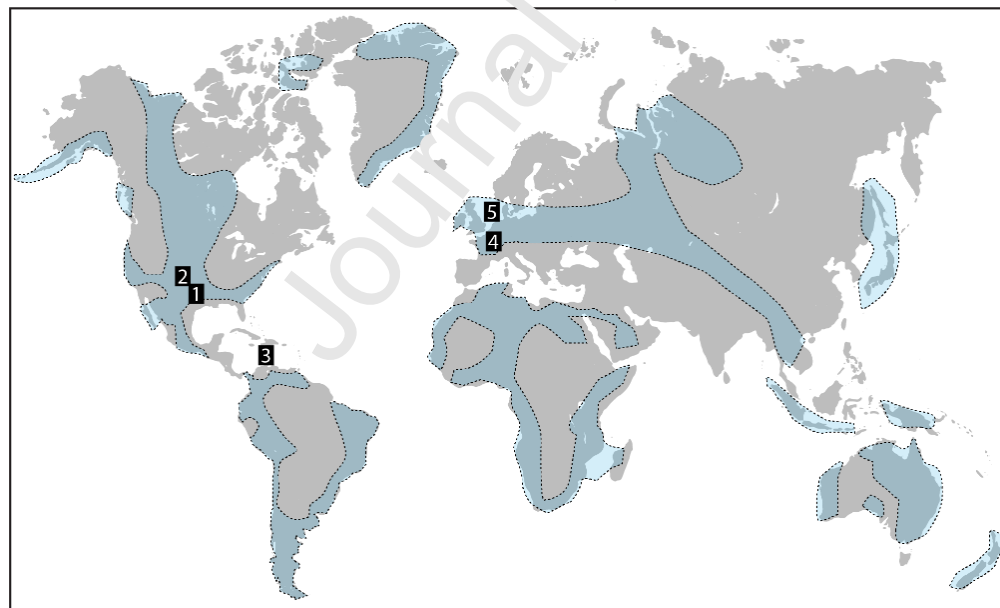


Figure 3. World map showing the overall distribution of Upper Cretaceous chalk deposits based on the palaeo-geographic map from (Mortimore, 2011). The locations of several onshore and offshore chalk deposits are also displayed and include: 1) the Austin chalk from the Pearsall-Gidding field ($\phi=3-10\%$, depth=2000m) (Pearson, 2012), 2) the Niobrara formation from the Silo, East Pony, and Redtail fields ($\phi=13-16\%$, depth=2100m) (Friedman, 1996), 3) chalk in the Colombian basin in the Caribbean sea from well DSDP 146 ($\phi=10\%$, depth=700m) (Carvajal-Arenas et al., 2020) 4) chalk from southern and eastern England, Northern France and northern Germany ($\phi=15-25\%$, depth=1500m) (Lasseur et al., 2009), 5) chalk from the Tor and Ekofisk Formation in

the Dutch, UK, Norwegian and Danish sectors of the North Sea (e.g. Dan, Tyra, Valhall, Ekofisk, Rembrandt and Vermeer fields with a typical porosity between 30-45% at a 2000-2900 m depth) (Hardman, 1982).

Sandstones also represent the predominant lithology for CO₂ storage projects worldwide. Because of the faster geochemical reactions in carbonates compared to sandstones, the former formation type has a great effect on the CO₂ uptake and mobility during the early injection time; however, their overall potential for mineral trapping was reported to be lower (Gunter et al., 1997). Moreover, calcite dissolution in presence of carbonated water may induce subsidence around the injection site, especially when the reservoir pressure near the injection well declines (Holloway, 1996). CO₂ injection in sandstone formations is perceived as safer since subsidence is less likely to occur in clastic formations. Nonetheless, CO₂ has already been injected in many carbonate formations for EOR or EOR + carbon sequestration purposes, e.g., at Weyburn (Canada). This storage site experienced no negative mechanical effects nor subsidence arising from carbonate dissolution. To assess the potential for CCS of different US regions and lithologies (including carbonates), Regional Carbon Sequestration Partnerships (RCSPs) were initiated. In Europe, a pilot test was undertaken in Hontomín (Spain) to progress the expertise of CCS in low permeability carbonates and demonstrate safe injection and storage (Kovacs et al., 2015). A successful demonstration project of CO₂ storage in a depleted hydrocarbon field (low permeability dolomite) was also carried out in Le Lacq (France) (TOTAL, 2015). Nonetheless, given the particularities of chalk (low permeability, high porosity, specific surface area, mechanical behaviour, reactivity, etc.), these demonstration projects did not seem to remove the stigma of unsafety when it comes to CO₂ storage in chalk. In this paper, we aim to give a perspective on the performance of CO₂ storage in chalk by reviewing existing experimental and modelling studies. We also identify and discuss the main gaps that should be addressed to ensure that depleted chalk fields in Denmark or elsewhere are suitable for CO₂ storage.

2. Storage site

2.1. Storage capacity

The injection of a lower density CO₂ phase into a reservoir leads to the formation of an ascending plume (Rochelle et al., 2004). By offering capillary resistance to the underlying gas (Kaldi et al., 2013), the caprock represents the first barrier for the CO₂ containment. Danish North Sea chalk fields are overlain by a thick cover of Tertiary shales and most traps in which oil occurs are elongated anticlines and salt-induced structures (see Figure 4) (Andersen and Doyle, 1990; Selley, 1976). Although the caprock capillary entry pressure of CO₂ is typically lower than that of hydrocarbons, the much lower viscosity of CO₂ means that the shock front height of the CO₂ plume (i.e., the CO₂ saturation immediately behind the leading edge of the plume) is low, typically 35% or less even in homogeneous reservoir rocks (Berg and Ott, 2012; Buckley and Leverett, 1942; Ott et al., 2012). Heterogeneity in the reservoir will reduce this even further, because CO₂ flow will become channelized along the most permeable layers, and much of the pore space will be bypassed. As a result, CO₂ storage efficiency may be as low as 5% in layered reservoir rocks (Matthai and Burney, 2018). The low permeability of chalk, however, will lead to more uniform piston-like displacement hindering the buoyant upward migration of the CO₂ which allows additional time for the CO₂ dissolution in the formation water, capillary trapping, and mineralization.

These trapping mechanisms are analysed hereafter in connection with chalk properties, whereas the structural trapping is discussed within the caprock integrity section.

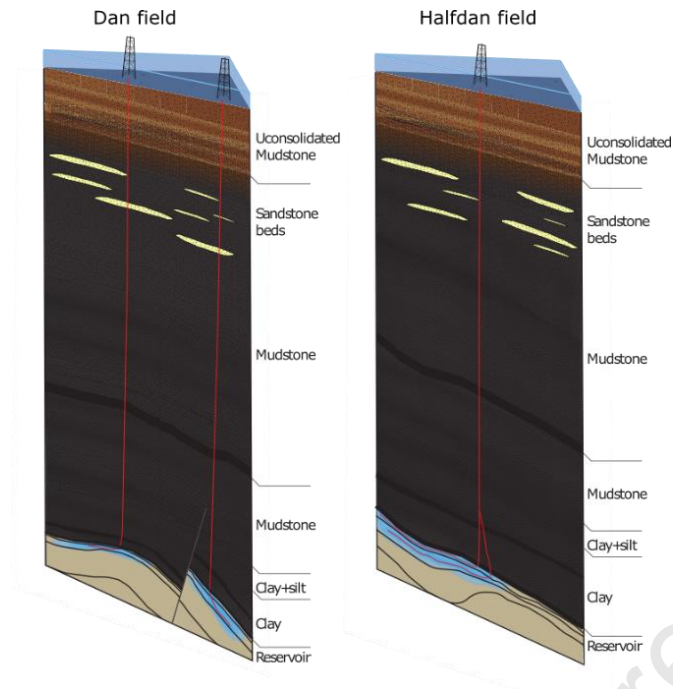


Figure 4. Two schematic stratigraphic columns illustrating the two characteristic geometries of chalk reservoirs from the Danish North Sea, an anticline structure (Dan field) and a continuous low-angle chalk succession (Halfdan field). The oil and gas infrastructures consist of three platforms in the Dan field and one in the Halfdan field, where wells are kilometres-long, parallel, and sub-horizontal. Three units are delimited in both reservoirs, from top to bottom, the Ekofisk Fm. and the oil- and water-bearing interval of the Tor formation. The large majority of the 2-2.5 km-thick overburden consists of claystone and mudstone with three shale and tuffaceous shale beds that are up to tens of metres-thick. Towards the top of the overburden, discontinuous sandstone bodies are present and were deposited by fluvial and turbiditic systems within shoreface to offshore depositional environments. To illustrate a potential CCS programme in the Danish North Sea, a sketch of CO₂ injection (blue) is shown in each field.

2.1.1. North Sea water chemistry and chalk permeability - Implications for solution trapping

The gradual dissolution of CO₂ reduces the pressure of the free gas phase acting on the caprock, hence the risk for leakage. The CO₂ solubility in the formation water is an important parameter for the accurate estimation of the storage capacity of a specific host rock. The solubility of CO₂ increases with pressure and decreases with temperature. A higher salinity reduces the solubility but the extent of the reduction depends also on the background electrolyte type (Liu et al., 2011). Different models have been proposed to predict CO₂ solubility in water (Spycher et al., 2003)/aqueous solutions (Duan et al., 2006; Duan and Sun, 2003; Li and Duan, 2007) and its phase behaviour (Pabsch et al., 2020; Tsivintzelis et al., 2011). After fitting the models to available experimental data, these can predict CO₂ solubility over a wide range of pressures, temperatures, and up to high ionic strength. (Yan et al., 2011) measured the solubility of CO₂ in water/brine and density of CO₂-NaCl mixtures up to high temperature (413 K), pressure (40 MPa), and salinity (5 M) (Figure 5). They showed that the solubility models and equations of state implemented in commercial simulators do not always predict satisfactorily the solubility neither the density at high salinity. At high salinity, the increasing ion-ion interactions introduce strong non-

idealities in the system; thermodynamic databases accounting for the aqueous speciation and models for activity coefficients are necessary to account for these deviations from ideal behaviour. While the models for the activity coefficients improve the description of real (non-ideal) systems, their implementation increases the computation cost. Because of this, commercial software usually includes empirical correlations or simpler activity coefficients models that are only valid up to salinities considerably lower (≈ 0.1 M) than that of saline aquifers or hydrocarbon fields.

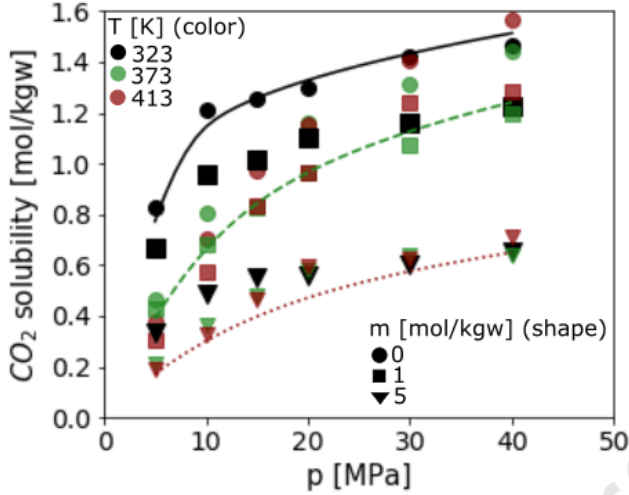


Figure 5. CO₂ solubility in aqueous solutions at different NaCl molality, pressure, and temperature; data from (Yan et al., 2011). The lines correspond to the calculated solubility using Pitzer model for activity coefficients.

When CO₂ dissolves in the formation water, the pH decreases according to reactions (1)-(4).



Eventually, the pH decrease impacts the reaction rates between the aqueous CO₂ with the host formation. The accurate description of the rock-fluid interactions requires reactive transport models/reservoir simulators that include thermodynamic models validated against experimental data to predict the pH at the in-situ conditions, i.e., temperature, pressure, and chemistry. These models should include a multicomponent description of the brine phase and a network of reactions to account for the CO₂ dissolution in water, aqueous speciation, and interactions of the aqueous species with the available mineral phases. All these interactions will impact the proton activity, thus, the pH. However, measuring pHs at high pressure, temperature, and salinity is challenging and makes it difficult to assess the consistency of the models. Several publications report pH measurements up to 15.3 MPa, 423 K, and 5 mol/kgw NaCl (Haghi et al., 2017; Xiaolu Li et al., 2018; Peng et al., 2013) and propose empirical correlations or test the performance of existing geochemical software (e.g., PHREEQC) to predict the pH. The existing models predict the pH value within the measurement uncertainty and the deviation

becomes more important (around 0.3 pH units) only at very high salinity (5 mol/kgw), which is not typical for the depleted hydrocarbon chalk fields in the Danish North Sea.

The total CO₂ trapped in the formation water is governed by the mixing through diffusive and, primarily, convective phenomena (Ennis-King and Paterson, 2005). Solution trapping can be enhanced by reservoir management strategies, such as injecting CO₂-depleted water/brine in zones with CO₂-saturated formation water (Leonenko and Keith, 2008) to shrink the CO₂ plume. However, the reservoir management possibilities will be restricted by the pore space availability and pressure constraints. The effective diffusion coefficient within a porous medium is usually inferred from the petrophysical properties of the formation and the molecular diffusion coefficient of CO₂ in water. The attempt of (Olivier, 2009) to assess the performance of existing models for chalk was unsuccessful because of the lack of experimental measurements at reservoir conditions of CO₂ diffusion coefficients (in both oil and water) available at that time in the literature. They later attempted to measure the CO₂ diffusion coefficient in water and oil (Lindeberg, 2017). However, instabilities caused by the density difference between water and CO₂ saturated water and the fast CO₂ dissolution in the oil led to convection patterns that challenged the diffusion coefficient acquisition. Several publications have, since then, published diffusion coefficients for CO₂ in water/aqueous solutions (Cadogan et al., 2015, 2014; Xingbo Li et al., 2018; Zarghami et al., 2017) at higher pressures and temperatures. The diffusion coefficient was shown to increase with increasing temperature and decreasing salinity (Zarghami et al., 2017) and to be less affected by pressure (Sell et al., 2013) as observed in Figure 6.

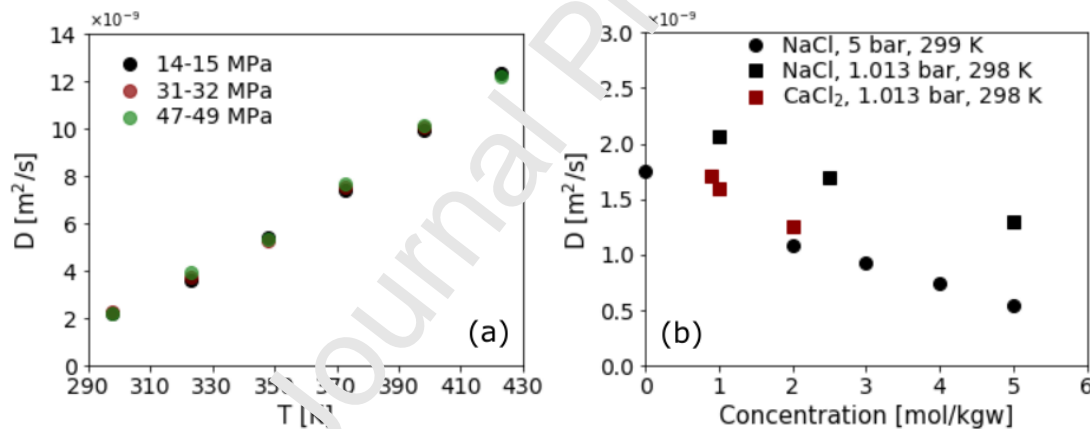


Figure 6. (a) CO₂ diffusion coefficient in water measured at different pressures and temperatures; data from (Cadogan et al., 2014). (b) CO₂ diffusion coefficient measured at different molalities; the data represented by black circles is taken from Sell et al., 2013 and the squares from (Cadogan et al., 2015).

Several papers (Farajzadeh et al., 2007; Xingbo Li et al., 2018; Zarghami et al., 2017) report a greater diffusion coefficient in the early stage of the experiment; this observation can, most likely, be attributed to the development of density-driven convection patterns caused by the dissolution of CO₂ in the brine/water. Thus, the measured diffusion coefficients in the initial stage of the experiment should be regarded as effective values as the transfer of CO₂ into the brine is, initially, not a purely diffusive process. Although expected to have a lower impact compared to the buoyant convection, the Marangoni convection may also contribute to the increased mass transfer rates recorded in the beginning of the experiment (Arendt et al., 2004). The predominant role of convective mixing over diffusion has been highlighted and described in previous publications (Ennis-King and Paterson, 2002,

2005). The CO₂ dissolution in the formation water increases its density (Figure 7). The descent of CO₂ saturated water and the rise of unsaturated lighter water result in convection patterns. This density-driven instability accelerates the mixing and speeds up the solution trapping within the reservoir eventually increasing the storage safety (because there is less CO₂ as a free phase). The research on this topic focuses on predicting the outbreak and the development of these instabilities. Understanding the behaviour of the fingers enables estimating the additional amount of CO₂ trapped in the solution as a consequence of the convective mixing.

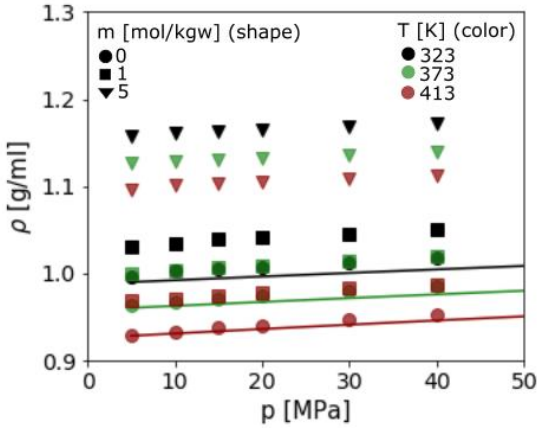


Figure 7. Density of CO₂-saturated NaCl solutions measured at different pressures and temperatures; data from (Yan et al., 2011). The lines correspond to the density of pure water determined with IAPWS online calculator.

The development of fingers is usually assessed through the Rayleigh (Ra) number, which indicates the relative importance of free convection to diffusion. The Ra can be calculated according to Eq. (5):

$$Ra = \frac{k \Delta \rho g H}{\phi \mu D} \quad (5)$$

where k is the effective permeability, $\Delta \rho$ is the density difference between the CO₂-saturated brine compared to formation brine, g is the gravitational acceleration, H is the depth of the reservoir, ϕ is the porosity, μ is the viscosity of CO₂-rich brine and D is the CO₂ diffusion coefficient. Theoretically, the development of fingers occurs at a Ra number greater than $4\pi^2$ (≈ 39.47) (Amarasinghe et al., 2020); the higher the Ra number, the more important the convection is. Since the properties of formation water, e.g., viscosity, density, are similar for many storage locations, the development of convective fingers will mostly be governed by the reservoir properties, especially permeability (Riaz et al., 2006) and thickness.

The convective fingers are usually studied through constant-pressure or pressure-decay experiments and visualization in a Hele-Shaw cell (Eftekhari et al., 2018; Emami-Meybodi et al., 2015). Performing these experiments with CO₂ at reservoir conditions is challenging and the use of analogues, e.g., water-glycol (Neufeld et al., 2010), have been proposed; however, these analogues may not necessarily be representative of the CO₂-water system (Jafari Raad and Hassanzadeh, 2015). Although visualization experiments are usually performed in the absence of porous media (Tang et al., 2019; Thomas et al., 2018), (Amarasinghe et al., 2020) included beads of different sizes to assess the impact of permeability. The decrease in the permeability (500 mD) leads to a diffusion-dominated regime and more piston-like displacement. (Soltanian et al., 2017, 2016) addressed the impact of permeability heterogeneity on the convective dissolution by carrying out simulations of advection-diffusion of CO₂. They showed that the

permeability fields and their connectivity strongly govern the density-driven instabilities and that heterogeneity can enhance the convective dissolution of CO_2 . Increasing the salinity not only retards the onset and growth of convective fingers but can even suppress them at salinities beyond 3 M (Thomas et al., 2018). Although in some cases higher temperature was reported to inhibit the convective dissolution (Jiang et al., 2020), others observed that increasing pressure and temperature promote fingering development (Tang et al., 2019). Depending on the pressure and salinity conditions, an increase in the temperature leads to either an increase or decrease of the CO_2 solubility in the aqueous solution (Figure 5), which may explain the observed contrasting effect of temperature on the convective fingers. The presence of impurities can also affect the CO_2 solubility in the formation brine and its density, thus the convective fingers. It has even been suggested that the CO_2 stream composition could be manipulated to intensify the convection and boost solution trapping (Jafari Raad and Hassanzadeh, 2017). The effect of pressure and salinity on the convective fingers development is sketched in Figure 8.

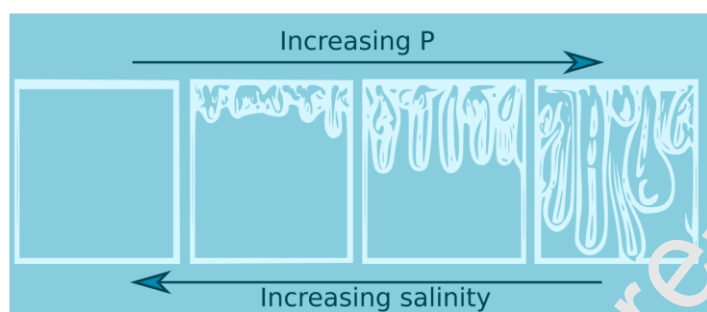


Figure 8. Effect of pressure and salinity on the development of convective fingers. The CO_2 solubility in the aqueous solution increases with pressure, promoting greater density gradients, higher Rayleigh number and, thus, the development of convective fingers. On the other hand, the CO_2 solubility in aqueous solutions decreases with increasing salinity, causing therefore the opposite effect. Temperature may either promote or inhibit the development of fingers, depending on the salinity and pressure conditions.

Traditionally, the convective mixing process is studied separately and disconnected from other interactions and phenomena occurring during CO_2 injection and storage (Emami-Meybodi et al., 2015). To bridge this gap, several authors assessed the effect of non-isothermal interactions (Eftekhar et al., 2018) and geochemical reactions (Babaei and Islam, 2018; Cardoso and Andres, 2014; Ghoshal et al., 2016; Sainz-Garcia et al., 2017) on the convective mixing. Geochemical reactions between the host formation and the aqueous CO_2 can reduce the convective mixing by removing CO_2 from the solution (Cardoso and Andres, 2014). Faster reactions will then have a greater potential for hindering the fingering because of an accelerated consumption of CO_2 (Babaei and Islam, 2018). Since the reaction rates for calcite with CO_2 are faster compared to other minerals, the convective flux in carbonate formations was reported to be reduced by 50% compared to other less reactive systems (Ghoshal et al., 2016). In contrast, (Sainz-Garcia et al., 2017) showed that fast dissolution reactions in carbonates translate into higher HCO_3^- content and porosity, leading to increased convection; this convection is also responsible for a non-uniform calcite dissolution.

Regardless of the effect of calcite reactivity, the very low permeability of chalk formations is expected to considerably hinder the convective mixing, thus the fast CO_2 trapping in solution. However, considering a gravity-override of the CO_2 plume, the presence of natural fractures in many chalk formations may increase the overall (vertical) permeability and essentially enhance the development of density-driven convective fingers. Additionally, the relatively low salinity of the formation water (≈ 1 M (Schovsbo et al., 2016)) compared to other potential storage sites candidates (>4.5 M, e.g., Gassum sandstone aquifer

(Kjøller et al., 2010)) will result in greater CO_2 solubility and diffusion coefficient, which will have a positive effect on the solution trapping. To resolve the temporal evolution of the solution trapping at potential storage sites, field-scale models are required to account for the convective-mixing and its effect on the dissolution rates; not including the effect of convective-mixing was shown to greatly affect the CO_2 plume features (Nordbotten et al., 2012). Nonetheless, to properly capture the effect of the convective-mixing at the field-scale, a computationally prohibitive fine mesh is required; to address this issue, (Gasda et al., 2011) accounted for the convective mixing through an upscaled mass transfer equation. Moreover, residual oil is another factor that should be taken into account when considering CO_2 storage in depleted chalk fields. Since the solubility of CO_2 in oil is greater than in formation water (Stewart et al., 2017), considering the presence of hydrocarbons will not only affect the storage capacity (Pingping et al., 2009) but also the onset and development of fingering. Considering the crude oil composition of the South Arne chalk field (Denmark), our rough estimations suggest that the amount of CO_2 dissolved in the oil phase will be significant even at low CO_2 molar fractions when the pressure is below the bubble point (around 30-38 MPa for the studied temperature range) and increases rapidly as the pressure reaches the bubble point and above (Figure 9). This behaviour cannot be extrapolated to other chalk fields as the amount of CO_2 that can dissolve in the oil will greatly depend on the specific composition of the crude oil.

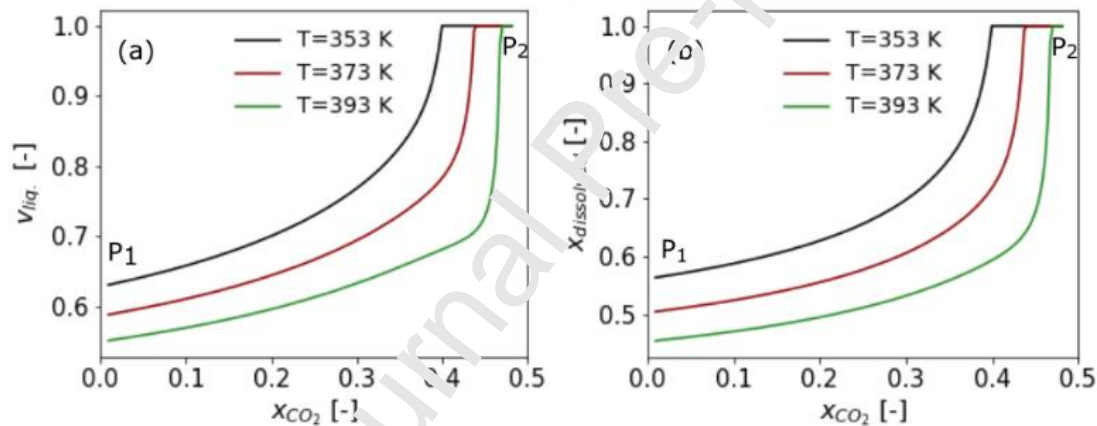


Figure 9. Phase behaviour of crude oil from the South Arne field (Denmark) in presence of CO_2 . The calculations were performed considering the specific crude oil composition and the SRK EoS with Peneloux volume correction reported by (Christensen and Pedersen, 2007). we considered an initial pressure $P_1=15$ MPa that increased up to $P_2=41$ MPa by the CO_2 supply. (a) Liquid fraction (volumetric) variation of CO_2 hydrocarbon mixture with increasing CO_2 molar fraction and pressure. At the initial pressure, the reservoir is below its bubble point and the hydrocarbon system consists of two phases. (b) CO_2 fraction dissolved in the hydrocarbon phase at increasing CO_2 content.

2.1.2. Chalk wettability – Implications for capillary trapping

During its upward migration, the CO_2 plume can be fragmented by water re-imbibing the pore space. This re-imbibition process breaks the continuous CO_2 phase into disconnected residual drops. The capillary-trapped CO_2 is immobile and cannot migrate, reducing the risk for leakage. Capillary trapping was shown to be more efficient at high initial water saturation (Akbarabadi and Piri, 2015) and to strongly depend on the injection scheme. WAG (Al-Khdeewi et al., 2018; Juanes et al., 2006), cyclic injection (Herring et al., 2016), and surfactants/foam alternating gas (Adebayo, 2018; Adebayo et al.,

2017) are some of the methods proposed to increase the CO₂ storage through this mechanism. Probabilistic geology models estimate that up to 95% of the total CO₂ injected may be stored by capillary trapping (Krevor et al., 2015); however, injection and reservoir management strategies may be needed to achieve this high percentage. Thus, the modelling of the capillary trapping will have great implications on the predicted CO₂ migration patterns. Hysteresis also considerably impacts the CO₂ plume dynamics (Akbarabadi and Piri, 2015; Juanes et al., 2006). This means that the partial brine displacement caused by CO₂ injection (drainage process if the CO₂ is considered non-wetting phase) cannot be described by the same relative permeabilities as when water flows back in the pores occupied by CO₂ (imbibition process). Hysteresis was found necessary to match the water production during CO₂ flooding in chalk samples (Ghasemi et al., 2018a, 2018c). Dynamic imaging methods can facilitate the determination of relative permeability and capillary pressure in such complex processes by in-situ imaging at representative conditions (Brautaset et al., 2010; Nørgaard et al., 1999; Zamiri et al., 2019).

The extent of CO₂ capillary trapping is eventually dictated by the residual gas saturation (Kumar et al., 2005). The higher the residual gas saturation, the greater the amount of CO₂ that can be capillary trapped. The gas trapped upon an imbibition episode is usually related to the initial gas saturation at the onset of imbibition as given by the Land model (Land, 1968). Commercial software generally accounts for the capillary trapping by considering the effect of hysteresis on the capillary pressure- and relative permeability-saturation functions. Different hysteresis models and formulations for the capillary pressure-saturation function may result in contrasting estimations of the potential for capillary trapping (Gershenzon et al., 2016). Besides hysteresis, local differences in the capillary pressure-saturation relationships can also assist in the immobilization of CO₂ (Li and Benson, 2015). To estimate the potential for capillary trapping, measurements of the residual gas saturation for different lithologies were reported. Some of these measurements were gathered and reviewed by (Krevor et al., 2015). The potential of capillary trapping in carbonate formations is a controversial topic. Evidence from core flooding experiments (Akbarabadi and Piri, 2015; El-Maghraby and Blunt, 2013) and pore-scale visualizations (Andrew et al., 2014) showed that the residual CO₂ saturation in carbonates is lower compared to sandstones. In contrast, the capillary pressure-saturation curves and residual gas measurements on cores of limestone and dolomite aggregates performed by (Wang and Tokunaga, 2015) suggest that carbonate-rich reservoirs may have a larger capillary trapping potential compared to quartz-rich reservoirs. The difference in the residual gas saturation of carbonate and sandstone samples are generally attributed to their variations in heterogeneity and wettability (Akbarabadi and Piri, 2015; Krevor et al., 2015). Since the capillary trapping depends on the wetting conditions, it will also be strongly influenced by temperature and water chemistry (Iglaue, 2017). Generally, greater capillary trapping potential is associated with water-wet conditions (Al-Khdheawi et al., 2017). (Al-Menhali and Krevor, 2016) altered the wettability of a limestone sample to mixed-wet conditions and showed that the residual CO₂ trapped by the sample at its original wetting state (water-wet) was greater than for the altered sample (Figure 10-a). As the surface became less water-wet, the CO₂ could form larger connected drops facilitating the flow of the CO₂ (Al-Menhali et al., 2016). Since only a tiny amount of organic acids can have a great (negative) effect on the residual trapping (Ali et al., 2019), depleted oil fields, which are most likely oil-wet or mixed wet, are expected to have a lower potential to capillary trap the CO₂ compared with saline aquifers. These studies (Ali et al., 2019; Al-Menhali et al., 2016; Al-Menhali and Krevor, 2016) imply that in depleted oil fields, CO₂ will migrate more freely, decreasing the overall storage safety. However, it was also recently shown that in a system composed of a preferentially oil-wet carbonate and three phases (i.e., water, oil, and CO₂), the CO₂ may be an

intermediate-wetting phase. Because of this intermediate affinity of the mineral surface for the CO_2 , the CO_2 was constricted to the corners of the pores, retarding its mobility (Alhosani et al., 2020).

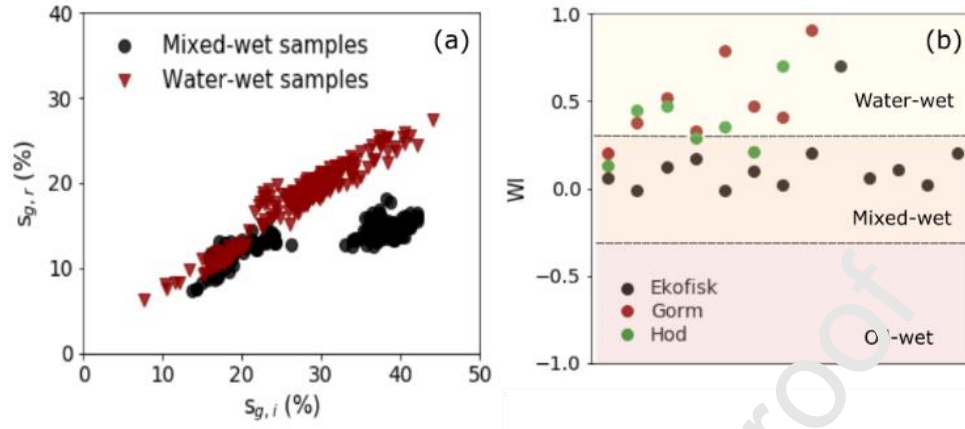


Figure 10. (a) Residual CO_2 saturation of water-wet and mixed-wet carbonate samples; data from (Al-Menhali and Krevor, 2016) (b) Measured Amott-Harvey index for chalk from different formations; data from (Ringheim and Hjelmeland, 1992).

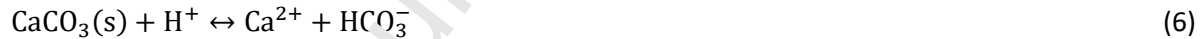
The potential of CO_2 capillary trapping in chalk has received less attention. Published studies on CO_2 floodings performed on chalk focused mostly on assessing the additional oil that could be recovered by CO_2 injection. (Gooya et al., 2019) estimated a maximum of 70% gas saturation by numerical simulation in nanotomographic image of chalk from Hod formation. One study reports a gas saturation after the flooding of about 60% (Høgnesen and Johansen, 2007). However, it is not clear how much of this CO_2 was capillary trapped. A recent micro-Positron emission tomography (μ -PET) imaging study (Brattekkås and Haugen, 2020) showed that after the injection of 2 pore volumes (PV) of CO_2 in a brine-saturated chalk sample, the CO_2 saturation reached 0.56. Injecting additional brine left behind capillary trapped CO_2 in around 40% of the pore volume; this CO_2 residual saturation was recorded at the breakthrough of the injected water. This is higher than the average 30% reported for sandstones (Krevor et al., 2015). However, the reviewed literature reveals a strong effect of wettability on the capillary trapping potential, which emphasizes that the residual CO_2 observed in one study cannot be extrapolated to all chalk formations. The wettability of chalk has been previously characterized as fractional with alternating hydrophilic and hydrophobic patches (Hassenkam et al., 2009). The chalk shielded by hydrocarbons may also have different wetting conditions depending on the formation brine compositions and the crude oil properties (Bonto et al., 2020); this is important for the North Sea chalk formations where the water chemistry is highly reservoir-specific (Bergfors et al., 2020). All these factors may explain the differences in the wettability index measured for chalk from different formations (Figure 10-b). CO_2 injection was also shown to induce time-dependent changes in the wetting conditions of both sandstones (Ott et al., 2011) and carbonates (Wang and Tokunaga, 2015). Although (Alam et al., 2014) observed no wettability change of reservoir chalk samples after CO_2 injection, geochemical reactions between chalk and CO_2 can lead to changes in the pore structure, shape, and roughness (Seyyedi et al., 2020) having implications for its capillary trapping potential. Thus, to accurately assess the amount of CO_2 that can be capillary trapped in chalk fields, models that can account for the effect of time-dependent wettability alteration (Kassa et al., 2020) and also for heterogeneous (fractional) wettability conditions (Yekeen et al., 2020) would be highly beneficial. Moreover, acknowledging that

there is no such storage site that can be characterized as overall homogeneous, upscaling procedures are necessary to account for the effect of small-scale heterogeneity on the local capillary trapping in (coarse-grid) field-scale models. Different approaches (Cheng and Rabinovich, 2020; Gasda et al., 2012; Nordbotten and Dahle, 2011; Rabinovich et al., 2015; Saadatpoor et al., 2011) have been proposed to upscale the capillary pressure. Since the methodologies for the upscaling often rely on assumptions regarding the type of forces dominating the flow and on the boundary conditions (Hassan and Jiang, 2012), their applicability may be site-specific.

2.1.3. Reactivity – Implications for mineral trapping

The dissolution of CO₂ in the formation water decreases the pH and triggers the dissolution of chalk. The transport of the dissolved species can lead to local oversaturation giving rise to calcite precipitation. An understanding of the calcite dissolution/precipitation is crucial to assess the potential for mineral trapping storage of chalk reservoirs. The potential for mineral trapping of carbonates is lower compared to sandstones (Rochelle et al., 2004; Rosenbauer et al., 2005). The monitoring implemented at the Weyburn field enabled the assessment of precipitation of secondary phases upon CO₂ injection by using geochemical software or reactive transport models (Cantucci et al., 2009; Perkins et al., 2005). Mineral trapping was found insignificant by 2005 (Riding and Rochelle, 2005) but estimated to increase within 100 years mostly by precipitation of dawsonite (Cantucci et al., 2009), resulting from the reaction between feldspars in the formation with dissolved CO₂. To predict the potential for mineral trapping within a reservoir, the kinetics of precipitation/dissolution of the minerals that constitute the reservoir need to be included. Chalk consists predominantly of calcite and to a lower extent of quartz and clays. The dissolution/precipitation of chalk is not only important to assess the mineral or solution trapping but will also have an impact on the porosity, permeability, and mechanical properties of the host rock; these factors will eventually affect the migration of the CO₂ plume and the integrity of the storage site.

The dissolution of calcite has been extensively investigated. Plummer et al. (Plummer et al., 1978) described the calcite dissolution process to occur through three independent but simultaneous reactions (Eqs. 6-8).



They measured dissolution rates on Iceland spar calcite and showed that the net rate can be expressed as shown in Eq. 9:

$$r = k_1 a_{\text{H}^+} + k_2 a_{\text{H}_2\text{CO}_3^*} + k_3 a_{\text{H}_2\text{O}} - k_4 a_{\text{Ca}^{2+}} a_{\text{HCO}_3^-} \quad (9)$$

where k_1 , k_2 , k_3 represent the constants for the kinetic rates of the forward reactions described in Eqs. 6 to 8, and k_4 is the constant of the backward reaction, a represents activities, and H_2CO_3^* stands for $\text{CO}_{2,\text{aq}} + \text{H}_2\text{CO}_3$; k_1 , k_2 , and k_3 depend on the temperature, whereas k_4 depends on both CO₂ partial pressure and temperature.

The rate law given in eq. (9), was found suitable to describe both forward (dissolution) and backward (precipitation) reactions (Busenberg and Plummer, 1986). However, others (Dreybrodt et al., 1997)

found the PWP rate law proposed by (Plummer et al., 1978) can describe the precipitation rate provided it is corrected for surface-controlled inhibition effects.

Since the dissolution involves a heterogeneous reaction, the rate-limiting step for this reaction could be the mass transfer between the surface and the bulk solution and not the chemical reaction. The fact that some experiments reported identical rates for marble, limestone, and synthetic calcite showed that CO_2 conversion into H^+ and HCO_3^- within the solution (Kaufmann and Dreybrodt, 2007; Liu and Dreybrodt, 1997) may also govern the dissolution kinetics. Different configurations or conditions used in the experimental setup may result in a different limiting step controlling the overall rate of dissolution, resulting in very different equilibrium constants (Alkattan et al., 1998); thus, the reported reaction rate constants should be understood as apparent constants.

With the publication of the IPCC special report on CCS (Abanades et al., 2005), the number of studies on calcite dissolution at high CO_2 pressures increased. (Pokrovsky et al., 2005) measured dissolution rates of calcite (Iceland spar), dolomite, and magnesite at 25°C , pH between 3 to 4, and high CO_2 partial pressures (up to 5.58 MPa). The calcite dissolution rates were strongly influenced by the speed of stirring but less by the CO_2 partial pressure; this highlights the impact of the hydrodynamic conditions on the experimental outcome. (Peng et al., 2015) studied the kinetics of calcite dissolution at high temperatures ($50\text{--}100^\circ\text{C}$) and CO_2 partial pressures (up to 13.2 MPa) using the rotating disk technique. They first found the range of the stirring speed that would yield a constant reaction rate, ensuring in this way the study of the kinetics under surface reaction controlled conditions. Then, they proposed a new kinetic mechanism consisting of two parallel first order reactions, i.e., capturing the effect of pH and that of carbonic acid. To understand the effect of surface morphology on the dissolution rates, they performed experiments on samples that had undergone surface treatment. Samples with different initial defect density did not show considerable reactivity differences. In a separate work (Peng et al., 2016), they found that salinity impacted only weakly the calcite dissolution rates. In contrast, an AFM study (Ruiz-Agudo et al., 2010) showed the impact of both the background electrolyte type and its concentration on the calcite dissolution rates. For instance, Mg^{2+} was shown to increase dissolution rates (Ruiz-Agudo et al., 2009) and to inhibit calcite growth (Nielsen et al., 2013). (Anabaraonye et al., 2019) demonstrated that the dissolution rate of Iceland spar calcite in a NaCl and NaHCO_3 brine is only slightly lower compared to a multicomponent system containing Na^+ , Mg^{2+} , K^+ , Cl^- , SO_4^{2-} , HCO_3^- and that an increase in the molality leads to a slight increase in dissolution rates.

However, the aqueous chemistry effect on reactivity may not necessarily be the same for all calcite samples. Through CT imaging of a Rørdal chalk sample at high pressure, (Yang et al., 2018) observed that heterogeneities, such as fossil remains, induce different dissolution behaviour compared to pure calcite. (Svensson and Dreybrodt, 1992) pointed out the different dissolution rates of natural samples (marbles, limestone, and marine sediments) and synthetic calcite in a $\text{CO}_2\text{--H}_2\text{O}$ system; impurities such as phosphate and organic material incorporated during diagenesis may inhibit dissolution. (Ricci et al., 2015) showed through AFM that stearic acid slows down calcite dissolution rates and that, depending on the amount, the organic molecules could even become embedded in the growing crystal. Organic material can also inhibit calcite precipitation rates (Lin et al., 2005); the extent of the inhibiting effect depends on the carbonate/calcium ratios, pH values, and organic matter characteristics (e.g., molecular weight, functional groups). Contrary to these studies, (Pokrovsky et al., 2009) showed only a weak effect of several organic (acetate, oxalate, malonate, succinate, phthalate, citrate, EDTA) and inorganic ligands

(sulphate, phosphate, borate, silicate) on the calcite and magnesite dissolution rates at 60°C, 3 MPa CO₂, and pH of 5.

Despite the similar dissolution rates reported for limestone and chalk by (Peng et al., 2016), increasing evidence suggests that the latter type has a different reactive behaviour compared to synthetic or other natural calcite samples (Figure 11). (Hao et al., 2020) observed comparable dissolution behaviour for synthetic calcite and chalk (both reservoir and outcrop samples) in presence of deionized water. However, when MgCl₂ was added to the water, the calcium concentration with time progressed differently for the chalk samples. (Yang et al., 2018) showed that, given its high specific surface area, chalk dissolution is faster.

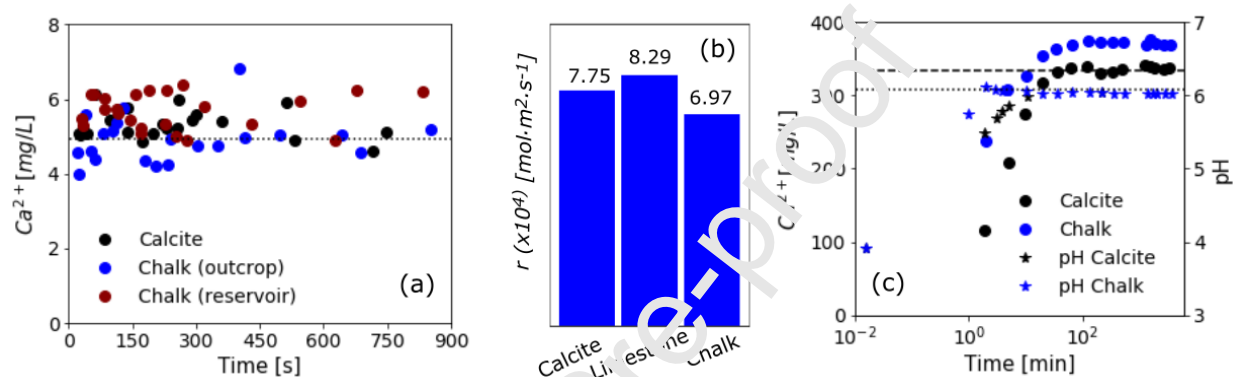


Figure 11. Comparison between chalk and calcite reactivity. (a) Calcium concentration profile in a system containing water and chalk or calcite particles (in absence of CO₂); data from (Hao et al., 2020). The horizontal line represents the Ca²⁺ equilibrium concentration predicted by Phreeqc using “phreeqc.dat” database. (b) Dissolution rates for different calcite samples inferred at T=353K and P=6 MPa CO₂ by (Peng et al., 2016) c) Calcium concentration profile in a system at room temperature containing water equilibrated with 1 atm CO₂ and chalk or calcite particles; data from (Yang et al., 2018). The horizontal lines represent the Ca²⁺ equilibrium concentration and equilibrium pH predicted by PHREEQC using “phreeqc.dat” database.

(Lakshtanov et al., 2018) showed that the growth of calcite and chalk obeyed different laws and that the rates for chalk precipitation were three times lower compared to calcite. Polysaccharide layers on the chalk surface were suspected to inhibit re-crystallization and govern calcite precipitation on chalk. To further corroborate this suppressing effect, (Sand et al., 2014) extracted ancient polysaccharides from North Sea chalk and added them to Iceland spar calcite. They showed that the polysaccharide extracted from Maastrichtian chalk was still very active and significantly hindered the dissolution and precipitation of calcite. Applying surface treatments to reduce the amount of organic material on the chalk resulted in higher recrystallization rates (Belova et al., 2012). Other characteristic features, such as the presence of nanoclays, may also explain the different surface properties and reactive behaviour of chalk compared to synthetic calcite samples (Okhrimenko et al., 2014; Skovbjerg et al., 2012).

Depending on the hydrodynamic conditions, the kinetics may become less relevant. Calcite dissolution/precipitation is faster compared to other minerals. When the time for reaction is shorter than for advection, the interactions between calcite and carbonated water can be described considering an equilibrium approach. In this case, the slow transport results in a residence time that is sufficient for the fluid to reach equilibrium with the surrounding minerals. Thus, the calcium concentration in the solution will be governed by the solubility limit. Many publications reported CaCO₃ solubility measurements at different pressures, temperatures, and CO₂ partial pressures. However, many of these

measurements are performed in deionized water or simple NaCl solutions. (Coto et al., 2012) measured calcite solubility and reviewed some of the existing measurements. They also developed a simulation tool in Aspen Plus to predict the solubility behaviour of CaCO_3 at different conditions using the ELEC-NRTL model. Additional solubility datasets are gathered in the work of (García et al., 2006), which shows the performance of the Extended UNIQUAC model for predicting and quantifying calcite precipitation in aqueous systems. However, existing thermodynamic models can only be labeled as reliable for describing CaCO_3 solubility in CO_2 storage applications once they are tested against extensive solubility experimental evidence at relevant temperatures, pressures, and brine compositions.

The revised literature points out the differences in the surface reactivity behaviour of chalk compared to calcite. This means that the models or equilibrium constants inferred for calcite dissolution/precipitation may not be directly applicable to chalk. Besides, the hydrodynamic conditions, surface reactive area, and degree of heterogeneity in the experiments may differ from those at the field scale (Li et al., 2008). Thus, an increasing number of studies (Noiriel et al., 2012; Wen and Li, 2008, 2017) focused on providing upscaled models for mineral kinetics. Pore-scale modelling has also been recognized as a valuable tool in extrapolating kinetics obtained on the micrometer scale (i.e., crushed samples) to bigger scales (Li et al., 2006; Raoof et al., 2013). In the case of depleted oil fields, it should be considered that the presence of an oil phase coating the chalk surface may impact the wettability and the overall dissolution and precipitation processes (Singh et al., 2018).

2.1.4. *Effect of reservoir heterogeneity on CO_2 storage efficiency*

CO_2 storage efficiency is also affected by vertical heterogeneity in the flood front as it propagates horizontally away from the injection site. The vertical heterogeneity (or stratification) of a CO_2 flood front will be enhanced by the high contrast in viscosity between CO_2 and water (compared for example to oil and water). A series of flooding experiments (on Berea sandstone core plugs) and numerical models (Berg and Ott, 2012; Ott et al., 2012) identified two processes causing stratification of the CO_2 front: horizontal viscous fingering and channelization. Horizontal viscous fingering results from instability in the CO_2 shock front caused when the mobility of the CO_2 phase immediately behind the front is greater than the mobility of the water or brine phase in front of it. It can occur even in a perfectly homogeneous reservoir and is scale-dependent (Berg and Ott, 2012); for typical CO_2 and brine mobility in the depleted hydrocarbon reservoirs, viscous fingering is not observed on the cm-scale but is likely to occur on a metre-scale and greater. Its effects will be enhanced by gravity, due to the high contrast in density between CO_2 and brine, leading to an extended “tongue” of CO_2 at the top of the reservoir extending far beyond the main flood front. However, the models also showed that a high capillary entry pressure can reduce viscous fingering by “spreading” the CO_2 shock front. Since chalk has a higher capillary entry pressure than sandstone, it is possible that viscous fingering will not be a significant issue in chalk reservoirs.

Channelization refers to the stratification of the CO_2 due to contrasts in the petrophysical properties (specifically the permeability and capillary entry pressure) of different layers in the rock. Unlike viscous fingering, it requires heterogeneity in the reservoir; however, both experimental and numerical modelling results show that a very slight contrast in petrophysical properties (a c.10% variation in permeability and <10% in capillary entry pressure) is sufficient to generate significant stratification of the CO_2 . This can occur at any scale, from the cm-scale up to the formation scale.

Modelling studies (Matthai and Burney, 2018) showed that stratification of the CO₂ flood front can reduce storage efficiency significantly by causing bypass of the lower permeability layers in the reservoir. In addition, by increasing the rate of propagation of the CO₂ flood front in the high permeability layers, it increases the risk that the CO₂ may pass a spill point or a break in the topseal (e.g., a fault), and thus leak from the reservoir. The chalk reservoirs of the Danish North Sea show vertical heterogeneity on a range of scales, from the formation scale (for example, the Ekofisk Formation has considerably different petrophysical properties to the Tor Formation (Megson and Hardman, 2001; Megson and Tygesen, 2005), through the metre-scale (with the identification of stratigraphic and sedimentary cycles (Scholle et al., 1998) of varying porosity and permeability, reflecting variations in clay/silica content, grain size, and early lithification resulting from subtle changes in depositional environments (Klinkby et al., 2005; Kristensen et al., 1995; Lindgreen et al., 2012; Stage, 2001; Vejbæk and Kristensen, 2000), down to the centimetre scale (with contrasts in petrophysical properties resulting from local diagenetic effects around stylolites or chert nodules (Lind et al., 1994)). It is therefore important that the occurrence and effects of such heterogeneity are understood when planning optimal CO₂ injection strategies.

A further source of heterogeneity in the reservoirs is provided by natural fractures. Many of the chalk hydrocarbon fields in the Danish North Sea contain extensive networks of natural fractures (Aabø et al., 2020) (and some also contain networks of artificial fractures, induced to assist hydrocarbon production). Models by (Matthai and Burney, 2018) showed that the presence of very thin, planar high permeability elements could have a dramatic effect on the CO₂ flow through the reservoir, leading to extreme channelization of flow through these elements and the horizons in which they were concentrated, rapid CO₂ breakthrough, and bypass of much of the rest of the reservoir. Although the elements in their model were intended to represent high permeability bedform surfaces, the same behaviour would be expected of open natural fractures (or indeed induced fractures left over from hydrocarbon production). It is thus important to understand the distribution of these fractures when planning optimal CO₂ injection strategies.

2.1.5. CO₂ storage with EOR - an opportunity for increasing storage capacity

The existing knowledge on the chalk behaviour exposed to CO₂ comes primarily from CO₂-EOR studies. Mærsk Oil and Gas considered the option of performing CO₂-EOR in a sector of the Dan field (Wishart and Pedersen, 2012) and engaged in important research for several years. The project was finally discontinued in 2011 because captured CO₂ was not available on time for the planned project start date and the project was economically unfeasible even if the gas was provided free of charge at the injection site. Recently, (Suicmez, 2019) published a paper that assesses the feasibility of CCUS within the Danish North Sea. CO₂ flooding was identified as a promising option to recover the remaining oil reserves and assist in reaching the required CO₂ emissions reduction target. According to their full-field scale simulations, approximately 5.4% additional oil recovery (104 million barrels) could be obtained by injecting approximately 750 MMscf CO₂ over 30 years (Figure 12). However, the cost of the facilities and that of supplying the CO₂ to an offshore location made the project very expensive. The NPV and profitability index (NPV/Discounted CAPEX of the project) were found to be very sensitive to the crude oil price, discount rate, and CO₂ cost. These findings are susceptible to uncertainties related to subsurface data such as hydrocarbon properties that may lead to different recovery forecasts. However, several publications (Johns and Dindoruk, 2013; Lindeberg et al., 2017; Suicmez, 2019; Vuillaume et al.,

2011) confidently suggest that the oil recovery increase by injecting CO_2 into the North Sea chalk fields ranges between 5-15%. Although CO_2 storage projects are more feasible in combination with EOR because of the revenues coming from selling the produced oil, the study points out that government subsidies are necessary to make this type of projects possible.

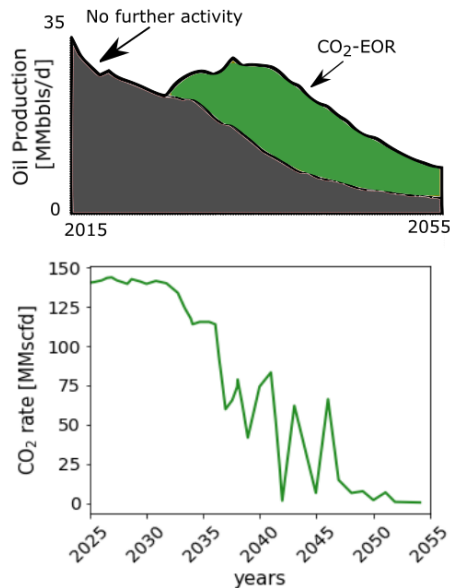


Figure 12. Oil production forecast for a CO_2 -EOR scenario for a Danish North Sea field (upper panel). CO_2 import rate required for injection (lower panel). The import rate decreases towards the latter stage of the project as some of the CO_2 that breaks through is separated from the hydrocarbon stream and reinjected. Adapted from (Suicmez, 2019).

In conventional CO_2 -EOR projects, the main goal is to maximize oil recovery with the minimum amount of injected CO_2 . However, CCUS projects could be engineered in such a way to either optimize both the oil recovery and the CO_2 storage (“advanced EOR”) or simply maximizing the CO_2 storage (“maximum storage EOR”). It was shown that both “advanced EOR” and “maximum storage EOR” result in negative net emissions even when considering the CO_2 emissions from the produced oil (Ampomah et al., 2017; Mokhtari et al., 2016; OECD/IEA, 2015). The largest CCUS project that focuses on maximizing CO_2 storage during oil production is the Weyburn project; many similar projects are now under construction. WAG injection, use of horizontal wells, or injection in the aquifer underlying the reservoir (if any) are some of the strategies that are recommended to co-optimize or maximize the CO_2 storage (Forooghi et al., 2009; Jessen et al., 2005; Karimaie et al., 2017). CO_2 -EOR projects are also more sustainable (thermodynamically speaking) than CCS projects because the energy required to inject and store the CO_2 is partially counterbalanced by that produced from the recovered oil. However, the output energy from a CO_2 -EOR project was shown to still be inferior to that required for storage, making the overall project unsustainable (Farajzadeh et al., 2020).

CO_2 -EOR performance depends heavily on the properties of the crude oil. These properties will determine its phase behaviour in presence of CO_2 and the development of miscibility. The pressure at which the oil and the CO_2 become miscible is called minimum miscibility pressure (MMP). (Christensen and Pedersen, 2005) assessed the phase behaviour of North Sea fluids from the South Arne and Ekofisk reservoirs in presence of CO_2 and developed EoS models compatible with ECLIPSE reservoir simulator software. Mærsk Oil and Gas also carried out a PVT study to investigate the behaviour of fluids from

Dan, Halfdan, and Tyra with CO₂ (Mærsk Oil and Gas, 2008). The established EoS from the PVT study was used for the determination of the MMP using software such as PVTsim and PhaseComp. At 350 K, the MMP was estimated to be 25 MPa.

Several papers addressed the CO₂-EOR performance in chalk by quantifying the oil recovery (Ghasemi et al., 2020; M. Ghasemi et al., 2018b, 2018a, 2018c; Ghasemi et al., 2018; Høgnesen and Johansen, 2007; Karimaie et al., 2007; Olsen, 2011, 2010, 2009, 2007) of CO₂ displacement tests on both outcrop and reservoir samples (Table 1). Other studies characterized the displacement dynamics by using non-invasive imaging methods such as X-ray Computed Tomography (CT) (Eide et al., 2016, 2013; Niu, 2010; Niu et al., 2010) and Magnetic Resonance Imaging (MRI) (Brautaset et al., 2008). These flooding tests showed the importance of pressure and temperature (Ghasemi et al., 2018c; Hamouda and Tabrizy, 2013), wettability (Chen et al., 2019; Ghasemi et al., 2020), asphaltene content (Hamouda et al., 2009), water composition, and saturation (Fernø et al., 2015b; Ghasemi et al., 2018b) on the outcome of CO₂ tertiary recovery. Since some North Sea chalk reservoirs are fractured, many of the experimental studies were carried out on specimens with artificially induced fractures (Eide et al., 2016; Fernø et al., 2015b; Ghasemi et al., 2020; Ghasemi et al., 2018b, 2018a, 2018c; Ghasemi et al., 2018; Ghasemi and Suicmez, 2019; Karimaie et al., 2007). To further improve the displacement efficiency and avoid fingering and preferential flow of CO₂ through fractures, foam (Fernø et al., 2015a; Fjelde et al., 2011; Zuta et al., 2010; Zuta and Fjelde, 2011) and switchable surfactants (Elmag et al., 2014; Sun et al., 2019) were proposed as suitable blocking agents. Diffusion was reported as the main recovery mechanism during CO₂-EOR in fractured chalk. The diffusion coefficients for oil and CO₂ are usually obtained either by fitting saturations obtained by imaging (e.g., X-ray CT (Eide et al., 2016)) and recovery data obtained from CO₂ floods performed on chalk samples or constant-volume-diffusion method (Ghasemi et al., 2017). For some experiments, Ghasemi et al. (Ghasemi et al., 2020; Ghasemi et al., 2018b, 2018c) defined two sets of diffusion coefficients, i.e., one set for the initial stage of the CO₂ injection and a second set for the later stage; each set contained diffusion coefficients for the oil pseudocomponents and CO₂ diffusion coefficients. Thus, for an oil composition described by 10 pseudocomponents, a total of 20 diffusion coefficients were necessary to fit the experimental data with a model implemented in ECLIPSE. Lastly, the oil recoveries expected at the field-scale cannot be assumed equivalent to those at the core-scale as the molecular diffusion depends on the length-scale. (Ghasemi and Suicmez, 2019) developed a 2D numerical model implemented in a commercial software to upscale at the field-scale their previous modeling and experimental results obtained on North Sea chalk core samples.

Table 1. CO₂ flooding experiments reported for chalk. *-reservoir cores; †- outcrop cores; ‡ cores with fractures induced artificially; Sor, Swi- residual oil and initial water saturation, respectively.

Ref.	Core type	k[mD]/ φ[%]	P[MPa] /T[K]	Flooding sequence	S _{or}	Observations
(Olsen, 2007)	South Arne*	6/42	41.4/388	FW→CO ₂ sat water	24→20.5	Dissolution at the inlet; slight weight loss, reduced porosity and gas permeability
		5/40			30.3→23.4	
(Høgnesen and Johansen,	South Arne*	4/43	41.4/388	FW→ CO ₂	27.8→13.4	No alterations/weakening were observed
		4/43			20.5→14.2	

2007)						
(Olsen, 2009)	Halfdan*	0.6/28.8	28.2/358	FW→CO ₂ sat water	28.1→23.3	Bulk volume reduction; dissolution at the inlet; increase in porosity (1.1%) and permeability (23%)
(Olsen, 2010)	South Arne* (Tor)	1.6/29.4	38/388	FW→CO ₂	25.6→5.9	No dissolution, compaction, nor permeability changes
(Olsen, 2011)	South Arne* (Ekofisk)	0.5/30.2	38/388	FW→CO ₂	37.7→4.6	No dissolution nor compaction
(Ghasemi et al., 2018a)	Stevns Klint ^{†y}	4.1/47.8 4.1/48.2	25.8/383	FW→CO ₂	25.4→22.0 26.3→24.2	Cores saturated with either North Sea tank oil or live oil
(Ghasemi et al., 2018)	Stevns Klint ^{†y}	2.5/46.8 2.6/47.1	25.8/383 33.6/373	CO ₂	44.4 10	Cores without S _{wi} and saturated with stock-tank oil or n-decane No core alterations observed
(Ghasemi et al., 2018b)	Stevns Klint ^{†y}	3.7/46.9 4.5/46.8	25.8/383	SW→CO ₂ FW→CO ₂	23.7→15.2 39.5→20.4	SW contains sulfate whereas FW is depleted from sulfate
(Ghasemi et al., 2018c)	Stevns Klint ^{†y}	4.1/46.8	30/383	FW→CO ₂	28.4→21.2	Oil dissolves up to 40% CO ₂
(Ghasemi et al., 2020)	Stevns Klint ^{†y} Reservoir ^y	4.4/48	25.8/383	SW→CO ₂	39.5→19.4 39.8→13.2	Effect of rock properties and wettability on CO ₂ -EOR; more water-wet conditions result in worse CO ₂ flood performance
(Hamouda and Tabrizy, 2013)	Stevns Klint [†]	1.5-6.5/ 10-48	9-14/ 323-353	Miscible CO ₂	NA	Better performance of CO ₂ in chalk compared to sandstones because of less fingering and greater surface area exposed.
(Hamouda et al., 2009)	Stevns Klint [†]	3-5/ 44-48	9-14/ 323-353	Miscible/ Immiscible CO ₂	10.94/ 64.44	Effect of asphaltenes precipitation on the oil recovery.
(Alam et al., 2014)	South Arne* (Ekofisk)	0.02- 4.8/ 18-42	38/388	Sc.CO ₂	6-18	No major alterations of the petrophysical properties. Slightly greater alterations for the sample with higher CaCO ₃ content (Tor).
(Karimaie et al., 2007)	Chalk ^{†y}	5/44	17/358	CO ₂ FW→CO ₂	27 47→31.2	Cores saturated with a mixture of n-heptane and methane(no S _{wi})
(Fernø et	Chalk ^{†y}	2.7-4.4/	9-14/	CO ₂	0-71	Effect of fracture

al., 2015b)	46-49	323-353	FW→CO ₂	29-46→ 22-36	permeability, diffusion length and S_{wi} on the oil recovery.
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2.2. Storage site safety

2.2.1. Chalk weakening

The mechanical properties of chalk are not only affected by the saturation state, but also by the type of fluid in its pores; this means, for instance, that chalk is weaker when saturated with water compared with organic fluids such as crude oil or glycol (Risnes et al., 2005, 2003). Several studies have also shown that the specific brine composition or the presence of certain ions can enhance the degree of weakening by inducing chemical interactions with the surface of chalk (Heggheim et al., 2005; Madland et al., 2011; Megawati et al., 2013). The presence of CO₂ can further accelerate chalk reactivity (Guen et al., 2007) and impact the mechanical properties of the formation. The popularity of waterflooding in oil production and the observed compaction in the Ekofisk (Liteanu et al., 2013) have driven extensive research on the water-weakening of chalk. Failure of grains and contact cement due to stress corrosion cracking and/or dissolution, pressure solution creep, and ion adsorption are some of the reported mechanisms behind the weakening (Liteanu et al., 2012).

Despite the existence of numerous CO₂-EOR projects in the US, the mechanical response of carbonates upon CO₂ injection is still raising concerns. Numerical (Kim et al., 2018; Renard et al., 2005) and experimental studies (Guen et al., 2007; Liteanu et al., 2012; Liteanu and Spiers, 2009; Vanorio, 2015; Vanorio et al., 2011; Vialle and Vanorio, 2011) have shown increasing compaction and strain rates when CO₂-saturated water was injected in carbonate cores. The extent of mechanical weakening depended on the type of carbonates (Guen et al., 2007), grain size (Liteanu et al., 2012), texture (Vanorio, 2015; Vanorio et al., 2011; Vialle and Vanorio, 2011), impurities in the CO₂ stream (Renard et al., 2011), and salinity (Liteanu and Spiers, 2009); experimental factors, e.g., flow conditions, also affected the mechanical response of the samples. Despite these studies, (Grgic, 2011) showed that CO₂/CO₂-saturated solutions led to compaction only when the flooding solution was not pre-equilibrated with the rock. At field conditions, the equilibration of the brine with the formation is expected to be fast and the storage capacity should not be affected by compaction.

Given the high porosity of chalk, reactivity, and compaction behaviour, studies were undertaken to understand the implications of CO₂ injection into chalk. (Madland et al., 2006) studied the mechanical weakening of outcrop Rørdal chalk by performing quasi-hydrostatic tests at 20 and 90°C on cores initially saturated with equilibrated water and then flooded either with equilibrated water, carbonated equilibrated water, or CO₂ gas. To assess the role of brine composition and initial wettability conditions on the weakening effect, a few tests were performed with seawater and on samples aged in crude oil. The experiments revealed that the axial strain was only minor when equilibrated water or CO₂ gas was injected in the sample but increased considerably for the sample treated with CO₂-saturated water. A temperature increase or more oil-wet conditions had a positive effect on chalk strength. Contrarily, during creep under hydrostatic stress slightly beyond pore collapse, oil-wet chalk samples registered greater strain per volume of fluid injected. (Korsnes et al., 2008) performed hydrostatic tests at room temperature and flow conditions on Rørdal chalk with similar properties. They flooded the cores with different types of brines (i.e., deionized water, seawater, and seawater enriched in sulfate), both

depleted and saturated with CO₂. After loading the samples hydrostatically beyond yield (12 to 14 MPa), these were left to creep while alternating flooding stages with water and CO₂-saturated water. No differences were observed in the hydrostatic yield strength for different aqueous solution types at room temperature. Then, they alternated creep phases followed by flooding phases with the brines and the CO₂-saturated brines. The creep tests showed that the strain rate/volume of fluid injected was greater in the presence of CO₂-saturated brines and in brines with high sulphate content. (Alam et al., 2014) investigated the effect of CO₂ on the mechanical and petrophysical properties of reservoir chalk samples obtained from the Ekofisk and Tor formations in the Danish North Sea. The chalk samples were conditioned to become representative of a waterflooded oil reservoir undergoing tertiary recovery by CO₂ flooding. After CO₂ injection, the samples were flooded again with formation water to remove the remaining CO₂. Then, the cores were unloaded from the flooding setup and stored in formation brine until being brought under the triaxial tests. CO₂-flooded samples were slightly less stiff and showed only a marginal decrease in the pore collapse stresses (Figure 13-a). The changes in the mechanical behaviour of Ekofisk chalk samples after CO₂ flooding were more insignificant compared to those from the Tor formation. The different impact of CO₂ on these samples was linked to their CaCO₃ content; Ekofisk samples contained 20-25% quartz and clay, whereas the non-carbonate percentage in the Tor samples was less than 5%. Thus, the higher mineralogical heterogeneity in the Ekofisk formation was understood as less amount of contact cement available to interact with the CO₂. The effect of CO₂ saturated water (equilibrated/not-equilibrated with calcite) on the strength, deformation, and creep properties of 24 reservoir chalk core samples from the Tor formation was also studied by (Ditlevsen and Christensen, 2010). The sample preparation procedure was similar to that in (Alam et al., 2014), i.e., to replicate the reservoir conditions after CO₂-EOR. Then, the experiments simulated four scenarios: (i) waterflooding - samples flooded with seawater equilibrated with CaCO₃ (reference samples), (ii) near-well/injection conditions - samples flooded with 6-7 PV of fluid CO₂ at 25°C, 38 MPa pore pressure, and 43 MPa confining pressure, (iii) far from injector conditions - samples flooded with CO₂ saturated seawater in equilibrium with CaCO₃ at 10 MPa (pore pressure) and 15.8 MPa confining pressure, (iv) intermediate distance from injector – samples flooded with CO₂ saturated seawater (no pre-equilibration with CaCO₃) at 75°C, 10 MPa pore pressure, and 15.9 MPa confining pressure. Scenarios (ii)-(iv) were followed by CaCO₃ equilibrated seawater to remove the CO₂ from the sample before unloading the cores from the coreflooding setup. After unloading and before the mechanical tests, the cores were again vacuum-saturated with CaCO₃-equilibrated seawater at 22°C. Several triaxial tests and one uniaxial compaction test were performed for each scenario and these revealed that chalk was not affected by exposure to CO₂ (Figure 13-b). (Liteanu et al., 2013) studied the effect of CO₂ on Sibbergroeve Maastrichtian chalk (outcrop) by performing mechanical tests on both dry and wet samples at 20 and 80°C, effective confining pressures in the range 0.5 to 7 MPa, and pore fluid pressures up to 10 MPa. The wet tests were carried out with pre-equilibrated water with CaCO₃ particles and with pre-equilibrated water saturated with CO₂. Saturated chalk showed a decrease in the compressive strength between 15-65% but injecting CO₂ or increasing the temperature caused no further weakening effect (Figure 13-c).

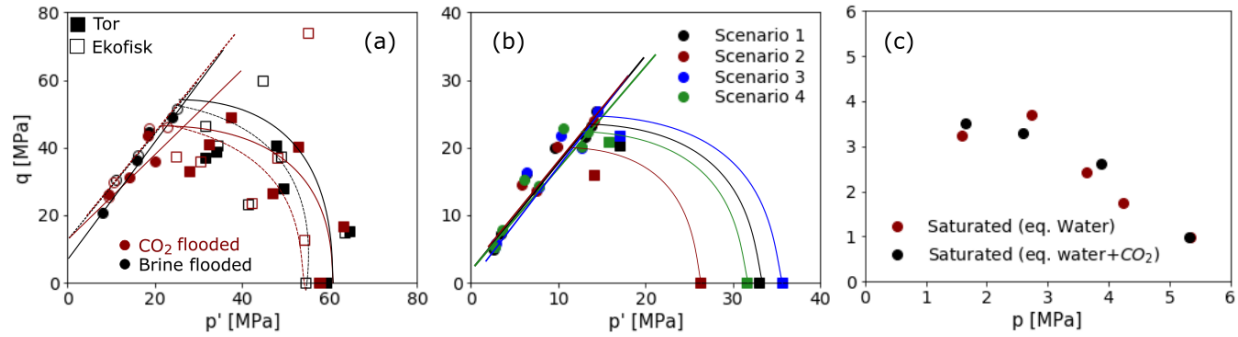


Figure 13. Failure envelope of CO₂-treated compared to waterflooded (reference) samples. (a) Data from (Alam et al., 2014). The dots indicate shear strength and the squares compaction strength. (b) Data from (Fitlevsen and Christensen, 2010). The dots indicate shear strength and the squares compaction strength. The different entries in the legend refer to the experimental conditions described in the main text. The unexpectedly low pore collapse stresses for the second scenario were qualified as uncertain and the authors recommended further investigation. (c) Data (Liteanu et al., 2013). The weaker nature of this chalk compared to Danish samples was attributed to the high degree of meteoric weathering.

The existing studies show that the effect of CO₂ on the mechanical properties of carbonates is conflicting. The observations strongly depend on the experimental conditions: saturation state at loading, aqueous solution pre-equilibration, pore fluid composition, backpressure, flow conditions, specimen grain size, and mineralogy are only some of the factors determining the experimental outcome. Specifically for chalk, the laboratory evidence indicates that reservoir samples are less affected than outcrop ones. However, the mechanical tests on the reservoir chalk were performed at no-flow conditions (after flooding) and room temperature, whereas dissolution and precipitation were shown to be affected by the effective stress and test temperature; this means that mechanical deformation and flooding are coupled processes (Nermoen et al., 2016) that should not be studied separately. When the cores were cooled and depressurized during the unloading from the flooding setup, the change in pressure and temperature conditions could have promoted calcite precipitation. Precipitation at the intergranular contacts can eventually affect the strength of chalk. These induced artifacts should be avoided by designing the experiments in closer agreement with the reservoir conditions. Moreover, chemical effects (e.g., dissolution) leading to weakening are more relevant at non-equilibrium conditions. At the field scale, these conditions will most likely occur near the wellbore; thus, chalk weakening may become especially relevant for wellbore stability. Yet, injection of dry supercritical/gas CO₂ has a limited effect on the mechanical properties compared to carbonated brine. Close to the injector, CO₂ may even dry the formation, which may induce a positive effect on the chalk strength.

Despite considerable efforts, defining a general mechanical behaviour when CO₂ is injected into chalk is not straightforward; the complex behaviour of chalk arises from its heterogeneous internal structure (Risnes et al., 2003) and surface properties (Hassenkam et al., 2009). Furthermore, the chalk wetting conditions (Sachdeva et al., 2019) and chemical/mineral heterogeneity (Minde et al., 2018) control the geomechanical response in mysterious ways. A recent study has shown that mixed-wet chalk samples are stiffer and stronger than water-wet samples (Sachdeva et al., 2019) but the creep did not seem influenced by wettability (Sachdeva, 2020); however, there is also evidence that oil-wet samples exposed to CO₂-saturated water showed higher strain rates compared to water-wet samples (Madland et al., 2006). Moreover, while (Renard et al., 2005) predict an increasing mechanical deformation with increasing chemical/mineral heterogeneity (thus, lower CaCO₃ content), the experiments performed by

(Alam et al., 2014) showed the opposite effect, i.e., slightly greater weakening for the cores with greater CaCO_3 content. All these greatly site-specific factors should be considered when assessing the effect of CO_2 on the mechanical behaviour of a chalk reservoir. Considering that many of the experimental observations are sample- or protocol-dependent, it may prevent the upscaling of the geomechanical behaviour from core- to field-scale (Rohmer et al., 2016).

2.2.2. Caprock integrity

In order to evaluate the risks to caprock integrity, it is important to understand the mechanism by which caprocks prevent hydrocarbons from leaking from oil and gas fields. It should be stressed that caprocks do not act as seals because they only have negligible or zero absolute permeability, but because they have a very high capillary entry pressure, so the hydrocarbons cannot enter their pore network and their relative permeability to the hydrocarbon phase is zero. Caprock failure can therefore occur by two methods (Watts, 1987): i) if hydrocarbon phase pressure is sufficient to overcome the capillary entry pressure it will be able to flow through the caprock pore system: this is known as membrane seal failure. It is controlled by the density of the hydrocarbon phase and the height of the caprock above the free water level; thus for a specific hydrocarbon composition, we can define a maximum column height that the caprock can contain; ii) if the hydrocarbon phase pressure exceeds the fracture pressure in the caprock, fluid-driven fractures may start to propagate through the caprock, allowing hydrocarbons to escape; this is known as hydraulic seal failure. This is also controlled by the height of the caprock above the free water level, but also by the effective horizontal stress and the mechanical strength of the caprock, and may be different from the column height at which membrane seal failure occurs.

Since supercritical CO_2 is also a buoyant immiscible fluid, the same seal failure mechanisms will apply. However, several additional factors must be considered when evaluating the caprock of a CCS prospect.

Firstly, the capillary entry pressure is a function of the interfacial tension between the wetting and non-wetting fluid phases, as well as the contact angle between the fluids and the grain surfaces, and so will vary for different combinations of fluid and caprock lithology. Numerous experimental studies on shale, marl, and evaporite caprocks (Chiqui et al., 2007; Hildenbrand et al., 2004; Li et al., 2005; Tonnet et al., 2011; Wollenweber et al., 2010), suggest that the capillary entry pressure of CO_2 is approximately half that of methane or oil, due to the lower interfacial tension between CO_2 and water, and therefore we cannot assume that, because a particular field was able to hold a certain volume of hydrocarbons, it will be able to hold the same volume of CO_2 .

Secondly, as noted in *Section 2.1*, the storage efficiency is typically much lower than for hydrocarbons. One way to increase the storage capacity is to increase the CO_2 phase pressure, by increasing the column height. The CO_2 phase pressure at the top of the structure is therefore likely to be higher than in a hydrocarbon field with equivalent volume, leading to a greater risk of both membrane and hydraulic seal failure (Streit et al., 2005; Streit and Siggins, 2005; Vilarrasa et al., 2011).

A third factor is that, unlike hydrocarbons, CO_2 may react chemically with the grains or cement in the caprock (Berthe et al., 2011; Dalkhaa and Okandan, 2011; Korsnes et al., 2008; Lima et al., 2011; Madland et al., 2006; Rathnaweera et al., 2015). This is a particular risk if the caprock is a carbonate or contains carbonate cement, which may be dissolved by CO_2 (Ellis et al., 2011). This can have two effects: i) an increase in the pore (and especially pore throat) size, which will reduce the capillary entry pressure

and promote membrane seal failure; ii) geomechanical weakening of the caprock, allowing fracture propagation and hydraulic seal failure.

Experimental evidence suggests that supercritical or gaseous CO₂ is relatively inert, and has little effect on the host rock (André et al., 2007; Madland et al., 2006). However, when it dissolves in the water phase it leads to acidic conditions making the overall environment much more reactive. This is likely to occur near the edge of the CO₂ plume – i.e. at the interface of the reservoir rock and the caprock. The experimental evidence is mixed about the immediate impact of CO₂ saturated water on carbonate-rich caprocks: some studies suggest it may increase the porosity and decrease the capillary entry pressure, and/or reduce the mechanical strength of carbonate-rich shales (Amann et al., 2011) while others suggest that the immediate effect is negligible (Ojala, 2011).

When assessing risks for a CCS prospect, however, it is also important to consider the medium to long-term effects of dissolved CO₂ on the caprock; it is necessary to demonstrate seal integrity over periods of hundreds to thousands of years. This is difficult to do as it necessitates extrapolation from experimental timescales; moreover, relatively few such experiments have been carried out on caprocks, since most experimental studies have focused on reservoir rock. However, the results of several extended duration experiments on shales and marls carried out over periods of days to months, suggest that even over these timescales, dissolved CO₂ can have multiple effects on the petrophysical and mechanical properties of the caprock, including an increase in porosity (Dalkhaa and Okandan, 2011), a decrease in caprock strength (Lima et al., 2011), and enhanced subcritical propagation of micro-cracks and fractures (Ellis et al., 2011a).

Once dissolved in the water, the CO₂ is no longer constrained by the membrane sealing properties of the caprock, but can permeate the caprock pore system through the water phase (which forms the wetting phase). Fortunately, dissolved CO₂ increases the density of water, so there is no buoyancy or convection drive assisting the escape of the CO₂-saturated water through the caprock; however, CO₂ can still permeate through the caprock by diffusion, driven by the concentration gradient between the reservoir rock and the overburden (Amann et al., 2011; Berthe et al., 2011). This however may not be sufficient to cause significant CO₂ leakage over long timescales.

The final factor that must be considered is that supercritical CO₂ is a powerful dehydrating agent, and this can also affect the clay minerals in the caprock. For example, dissolution of bound water in the CO₂ phase will cause shrinkage of the clay minerals, allowing more CO₂ to invade the caprock pore system, and generating tensile strain that will enhance fracture (André et al., 2007; Espinoza and Santamarina, 2012). In addition, as (Espinoza and Santamarina, 2012) pointed out, the dissolution of water in the CO₂ phase will reduce the water phase pressure; this will increase the capillary pressure of the CO₂ phase and thus promote hydraulic seal failure. This risk however may be limited as the CO₂ phase in contact with the seal is already fully wet and saturated with water. Moreover, the dissolution of water in the CO₂ phase can also lead to salt precipitation, which may increase the sealing capacity.

The caprocks of the chalk fields of the Danish North Sea comprise Palaeocene age clays of the Våle, Lista, and Sele Formations. As noted above, experimental studies of the effects of CO₂ have generally focussed on the reservoir rocks, and few studies have been carried out on the caprocks. We, therefore, have very limited direct data regarding the chemical and mechanical properties of these formations in the Danish sector of the North Sea. This may be partly due to the lack of core material (it is uncommon for caprock to be cored), and partly because the sealing capacity of these units is not the limiting factor

on the size of hydrocarbon accumulations in this area. However, for the reasons described above, it should not simply be assumed that a caprock capable of sealing a hydrocarbon accumulation is also capable of sealing CO₂.

Some studies have been carried to assess the possibility of CCS onshore Denmark, in particular in the Vedsted structure of northern Jutland (Mbia et al., 2014a, 2014b). These studies concluded that the caprocks for this proposed site would be effective seals for CO₂. However, the reservoirs and caprocks of this proposed site are Triassic to Jurassic in age, and thus have limited relevance to the offshore chalk structures.

More relevant data is available from the UK sector of the North Sea, where Worden et al. (Worden et al., 2020) examined the Lista Formation, to assess its effectiveness as a CO₂ seal for the Acorn CCS project. This study carried out a detailed mineralogical and petrophysical analysis of the Lista Formation clays, including Mercury Injection analysis, and concluded that it would be capable of sealing a CO₂ column between 166 and 470m in height. Furthermore, they suggested that the Lista Formation has a very low calcite composition, and is thus unlikely to react chemically with the CO₂.

2.2.3. *Fault reactivation*

Many of the considerations discussed regarding caprock seals also apply to fault seals, but there are also some additional factors, specific to fault seals, that must be taken into account.

When considering fault seals, it is important to distinguish between: i) juxtaposition seals, where fault juxtaposes the reservoir rock against the caprock or some other sealing lithology, such as shale (Knipe, 1997; Knipe et al., 1998), and; ii) fault rock seals, where the fault juxtaposes reservoir rock against reservoir rock, or some other permeable lithology such as sandstone, and the seal is provided by the fault rock itself. Fault rock seals may be provided by clay smears, shale gouge, cataclasite, or cement (in approximately descending order of seal effectiveness (Fisher et al., 2003; Fisher and Knipe, 1998)).

Fault rock seals may have a much greater risk of leakage since the trapped fluid only needs to break through a relatively thin membrane seal layer to escape. As discussed above, a greater CO₂ column height may be required to attain the same storage efficiency, and this leads to a greater phase pressure for the CO₂ phase compared with the hydrocarbon phase. This may cause leakage through the fault due to membrane seal failure. However there is also the possibility that it will reveal some leak point in the fault below the level of the Oil-Water Contact; for example the pinchout of a clay smear (Egholm et al., 2008; Hesthammer and Fossen, 2000; Sperrevik et al., 2000), or a lens structure providing a fluid escape pathway (Foxford et al., 1998). Even a small leak point would be sufficient to cause significant CO₂ leakage over long timescales, and may not be visible on seismic data. This risk can be limited if most of the injected CO₂ is either trapped within the pores by capillarity or dissolved in the remaining oil phase.

Chemical reactions between the CO₂ and the grains or cement in the fault rock may also cause leakage through the fault, either by reducing the capillary entry pressure and causing membrane seal failure or by weakening the fault rock, allowing fractures to grow and causing hydraulic failure. These processes have already been described with respect to caprock leakage, but are a greater risk for thin fault rock seals than for thick caprocks. It is important to note that, as observed in the Little Grand Wash Fault (Utah) zone (Patil et al., 2017), faults can be sealed due to CO₂ leakage.

Finally, we need to consider the risk of fault reactivation due to the elevated pressure of the CO₂ phase (Konstantinovskaya et al., 2012; Orlic et al., 2011; Ruth et al., 2006; Streit et al., 2005; Streit and Siggins, 2005) which can induce micro-seismic events. Fault reactivation can result in seal breach and fluid escape (Sibson, 2000, 1994, 1990), so may cause leakage of the CO₂ as well as presenting a public safety risk in its own right. The risk of fault reactivation may also be enhanced by CO₂-related dissolution and weakening of the fault rock, thus reducing its cohesion or friction coefficient (Orlic et al., 2011) and also by non-isothermal effects, shear slip stress transfer, and heterogeneity (Vilarrasa et al., 2019). For instance, at the In Salah (Algeria) storage site, the recorded micro-seismicity was partially ascribed to the 45°C difference between the formation and the CO₂ stream (Vilarrasa et al., 2015). A full evaluation of the fault reactivation risk requires 3D geomechanical modelling, incorporating the in situ stress state and the mechanical properties of surrounding rocks as well as fault rocks (Chang et al., 2011; Ducellier et al., 2011; Ouellet et al., 2011).

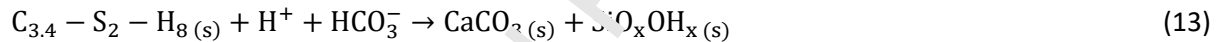
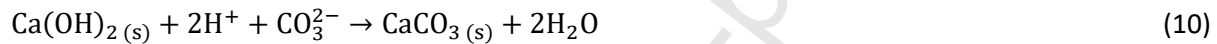
2.2.4. *Well integrity*

Wells are a primary element in CCS projects that can lead to CO₂ leakage (Barlet-Gouédard et al., 2009). After drilling a well, it is completed by running a casing inside the borehole, which is then cemented to the surrounding formation. A good cementing operation does not only provide isolation to the metal casing but also prevents fluid vertical migration through the wellbore annular space or caprock penetrations (Zhang and Bachu, 2011). To better understand and quantify the potential for CO₂ leakage through injection wells, existing data related to the integrity of O&G wells were analysed in several publications. Leakage incidents caused, most commonly, by sub-optimal cementation (IEAGHG, 2006) were shown to be quite typical (Bachu and Bennion, 2009a; IEAGHG, 2006; Watson and Bachu, 2009; Zhang and Bachu, 2011) and not related to the age of the wells. Factors such as well stimulation (Bachu and Bennion, 2009b) also increased the risk of leakage and in Alberta, wells exclusively built for CO₂ operations failed less compared to repurposed ones (IEAGHG, 2007). Cement with silica additives, used in many wells in the North Sea to maintain the cement strength above 110°C, has been shown to deteriorate more easily than ordinary Portland cement when exposed to CO₂ (Bjørge et al., 2019). These statistics may discourage the reuse of existing O&G wells for CO₂ injection and storage, despite their perceived financial advantage (IEAGHG, 2018).

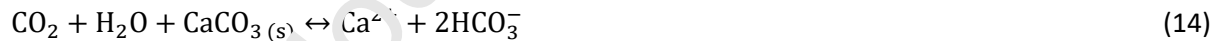
Besides the injectors, abandoned wells intersecting the CO₂ plume may also represent an important leakage threat (Scherer et al., 2005). Several articles reported methane leaks from abandoned wells in the Central North Sea (Böttner et al., 2020; Vielstädte et al., 2017, 2015). One-third of the wells in the North Sea were estimated to result in leaks, yet, the hydrocarbon gas emissions from these wells were found insignificant for the overall balance of global greenhouse gas (Böttner et al., 2020). It was also estimated that from 1998 to 2007 around 20-30% of wells in the Norwegian Continental shelf suffered at least one leak (IEAGHG, 2008) caused mostly by internal failures, seal, and steel issues rather than cementing problems. For the specific case of Denmark, between 1980-2009, only two abandoned wells were reported to experience leaks (IEAGHG, 2009a). The abandonment procedure may dictate the prospects of a project. For instance, abandonment plugs may contain elastomers (not in the North Sea) that can negatively interact with the CO₂ (Bachu and Bennion, 2009b); these wells would need to be re-plugged if they were anticipated to be exposed to CO₂. Although successful replugging was achieved in the Salt Creek Project (IEAGHG, 2009b), in some cases, workover operations are too costly and can lead to project cancellation (e.g., CO₂ injection in the Dutch De Lier field) (IEAGHG, 2009a). In the North Sea,

the remediation of leaky abandoned wells may become challenging because the wellhead and surface casings will have been permanently removed. Thus, well selection for CO₂ injection and storage in depleted oil fields should be done with caution, such that the CO₂ migration does not intersect unwanted abandoned “leaky” wells.

The experience from the O&G industry confirms the tight connection between leakage from wells and the cement quality. The function of the cement to provide zonal isolation and avoid any leakage can be compromised either by the consequences of sub-optimal cementation techniques (e.g., poor bonding with the formation, cracks, channelling, micro-annulus)(Bachu and Bennion, 2009b; Zhang and Bachu, 2011) or by the cement degradation, induced by the interaction with supercritical CO₂/carbonated brine (Barlet-Gouédard et al., 2009). As discussed in Section 2.1.1, CO₂ dissolution in formation water leads to different carbon-containing species distribution depending on the pH. The (bi)carbonate species react with the portlandite (Ca(OH)₂) and calcium silicate hydrate (C-S-H) from the cement (reactions 10-13); the former reactions results in one of the three CaCO₃ polymorphs, i.e., calcite, aragonite, or vaterite, depending on the pressure and temperature conditions (Zhang and Bachu, 2011) whereas the latter interaction, although slower (Duguid and Scherer, 2010), leads to the formation of a porous silica gel layer.



Cement carbonation does not impair cement properties as it results in lower porosity and permeability (Zhang and Bachu, 2011). However, as Ca(OH)₂ and C-S-H phases are depleted, the pH drops, and the calcium carbonate layer can undergo dissolution (reactions 14-15), leaving behind only the remaining porous silica layer (Duguid and Scherer, 2010). This bicarbonation process degrades the cement, i.e., increases its porosity and permeability.



While newly drilled wells can be cemented with CO₂-resistant cement (Barlet-Gouédard et al., 2009), CO₂ storage in depleted oil fields requires a guarantee that the cement in the existing wells is suitable for CO₂ applications. Although the cement obtained from a hydrocarbon field undergoing CO₂-EOR (Carey et al., 2007) and a natural CO₂ reservoir (Crow et al., 2010) showed poorer properties compared to pristine cement, it still prevented CO₂ migration and maintained structural integrity. Evidence of cement carbonation above the caprock in natural CO₂ does not necessarily imply CO₂ migration; over geological time scales the CO₂ could have diffused through the caprock, saturated the pore water, which eventually led to carbonation depths up to cm (Scherer and Huet, 2009). A research programme led by TOTAL concluded that Portland cement preserves its properties and is a suitable barrier to CO₂ migration (Garnier et al., 2010, 2012a; TOTAL, 2015). (Garnier et al., 2010) performed time-lapse X-ray CT measurements on samples stored at representative pressure and temperature. They observed that the carbonation front progression in class-G cement is controlled by diffusion, whereas in class G + silica

cement is controlled by kinetics. They concluded that the cement matrix reaction in CO₂ environments does not appear to be the driving phenomenon for the loss of well integrity. Mechanically-induced damage to the cement sheaths, e.g., due to hydro-fracturing, may pose a greater concern. Similar imaging methods were employed to test new types of cement that were resistant to CO₂ and H₂S (Garnier et al., 2012b). (Bachu and Bennion, 2009a) showed that injecting CO₂ saturated brine through class G cement samples for 90 days at the storage conditions expected at 1 km depth can lead to improved cement properties, i.e., decreased permeability. Others (Duguid et al., 2011; Duguid and Scherer, 2010) have revealed that the degradation extent depends also on the formation type the cement is embedded in; cement exposed to CO₂ flow at conditions representative of sandstone formations exhibited a temperature-dependent degradation, whereas no alterations were observed for the samples at the experimental conditions representative of limestone formations (Figure 14). Thus, for the case of carbonates (e.g. limestone, chalk), the formation water is saturated with respect to calcite (contains both Ca²⁺ and HCO₃⁻) inhibiting cement bicarbonation (eqs. 14-15).

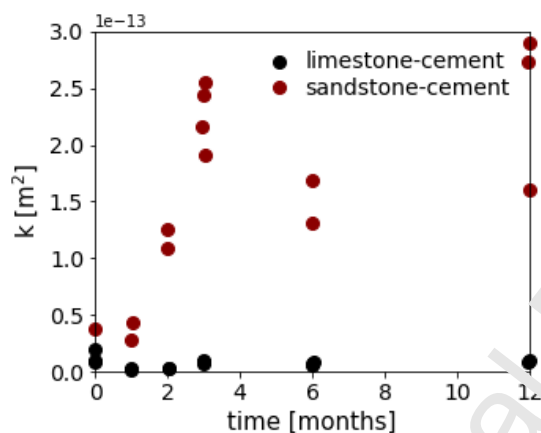


Figure 14. Evolution of cement-formation rock permeability upon flooding with carbonated brine under conditions representative of sandstone and limestone formations (pH= 5 and 50 °C). The permeability of the sandstone-cement system increased considerably with time and showed signs of degradation, whereas the cement embedded in the limestone samples showed no apparent alteration. Data from (Duguid et al., 2011).

The cement behaviour also depends on its initial conditions, e.g., cement with microcracks or with poor bonding (with the formation or casing) may respond differently to CO₂ compared to intact cement (Chavez Panduro et al., 2020; IEAGHG, 2012). Works of (Jung and Um, 2013) and (Um et al., 2017), among others, confirmed that (i) CO₂-saturated formation water damages the well cement much faster than dry or wet supercritical CO₂ and (ii) fractures, rock-cement, and steel-cement interfaces require more investigations. Several models (Kjøller et al., 2016b; Lecampion et al., 2013; Matteo and Scherer, 2012) were proposed to address and predict leakage rates through fractures or microannulus. Experimental evidence suggests also that cracks and microannulus may self-heal either by CaCO₃ precipitation (Chavez Panduro et al., 2020, 2017; Liteanu and Spiers, 2011), fines migration (Kjøller et al., 2016a), or cement swelling (Chavez Panduro et al., 2020). Besides the aqueous CO₂ concentration, the self-healing depends on other factors such as the confinement of the healed volume (Chavez Panduro et al., 2017), flow-rate (Luquot et al., 2013), residence time (Huerta et al., 2016), fracture geometry, stress state (Walsh et al., 2014a), and presence of corrosion products on the steel casing (William Carey et al., 2010; Wolterbeek et al., 2013). Apart from preventing self-healing, corrosion of the casing can itself jeopardize well integrity (IEAGHG, 2007, 2006). A review of natural and industrial analogues for CO₂

storage concluded that casing corrosion, caused by the incompatibility of construction materials with the injection fluids, represents the most frequent cause of failure of an injection well (IEAGHG, 2009a). Corrosion and pitting increased considerably after CO₂ was injected in a well in the K-12 B gas field in the Dutch sector of the North Sea (IEAGHG, 2006). British Petroleum (BP) also found that casing corrosion was more problematic than cement degradation in wells that are exposed to large temperature variations in the North Slope of Alaska and high CO₂ and H₂S content (IEAGHG, 2006). To avoid corrosion, chromium alloys are advised (e.g., 13 Cr steel showed good performance at CO₂ storage conditions (Hassani et al., 2014)), and an increase in the carbon content was also shown to increase the corrosion resistance (for the same chromium content, X20Cr13 < X46Cr13 (Pfennig et al., 2013)).

One of the advantages of storing CO₂ in depleted oil fields is the availability of wells. To ensure that the best wells are selected, the risk of leakage should be assessed. Reactive transport models can be used to predict CO₂ flow through the cement and the cement properties evolution upon reaction with the CO₂. These tools/models need to be calibrated with existing experimental evidence to carefully consider cement properties and the aqueous phase properties, such as ionic composition and pH (Gan et al., 2019). Different reactive transport models (Huet et al., 2010; Rabcus et al., 2012; Walsh et al., 2014b, 2014a; Wertz et al., 2013) that simulate reactions between the cement and CO₂/CO₂-saturated brines have been proposed. By history matching field or experimental results, porosity distribution and CO₂ diffusivity in the cement can be fit (Zhang and Bachu, 2011). Most reactive transport models consider a simplistic composition for the cement phase (portlandite and calcite) as thermodynamic data for the C-S-H phase is missing. Moreover, these mathematical tools rely on many adjustable parameters, making their applicability very case-specific. However, when CO₂ is planned to be injected into existing wells, any modelling attempts should include the mechanical and chemical characteristics of the in-situ conditions and not those of unaltered Portland cement as the cement may have already reacted with the formation brine and suffered compositional alterations (Scherer et al., 2011).

2.3. Injectivity

The permeability of the formation is a primary factor considered in screening potential sites for CO₂ storage. Formations with a permeability greater than 100 mD are usually regarded as preferable for hosting carbon dioxide (Antonsson et al., 2014); this is because a greater permeability allows higher CO₂ injection rates with lower pumping energy demand. The (generally) greater permeability of sandstones compared to carbonates may partially explain the preference for the former lithology in CCS projects. However, CO₂ mobility is not governed only by the formation permeability but also by its viscosity; since supercritical CO₂ has a low viscosity, its injection should not be restricted to high permeable formations (Kovscek, 2002). The fact that injectivity impairment has been reported at CO₂ storage sites with relatively high permeability (e.g., Ketzin (Zettlitz et al., 2010), Hontomín (de Dios et al., 2019), SACROC (Ghahfarokhi et al., 2016), In Salah (Aagaard et al., 2018; Eiken et al., 2011; Hansen et al., 2013), Snøhvit (Aagaard et al., 2018; Eiken et al., 2011; Hansen et al., 2013), Aquistore (Talman et al., 2020), etc.) shows that the ease of CO₂ injection is a function of many other factors besides permeability.

2.3.1. Chemical effects

Among different chemical effects that impact CO₂ injectivity, salt precipitation is probably the most studied one. The injection of dry CO₂ can partially displace or vaporize the water in the near-wellbore area. Upon evaporation, the salt in the formation water precipitates occupying part of the pore space. If

the precipitate blocks the CO₂ flow, the pressure will eventually increase, hindering the injectivity. Despite its negative effects, salt precipitation can also increase the sealing capacity around the wellbore if this deposition occurs at the caprock interface (Bacci et al., 2013). Salt precipitation was identified as the main reason for the pressure drop increase and decreased injectivity at Snøhvit (Aagaard et al., 2018) and the Aquistore CO₂ storage site (Talman et al., 2020). At the latter site, with highly saline formation brine (330 g/L), downhole images confirmed the presence of halite, sylvite, evaporates, and CaCl₂-containing salts among the precipitates. The extent of injectivity impairment caused by salt precipitation depends on both the ionic strength (André et al., 2014) and the type of electrolytes (Peysson et al., 2011). The consequences of salt deposition are further aggravated by the re-imbibition of water in the pore space (Cui et al., 2018; Ott et al., 2011; Talman et al., 2020; Zhao and Cheng, 2017). The backflow of saline formation water brings additional salt, which once evaporated leads to more precipitation until completely clogging the pore space. This means that the salt precipitation phenomenon is not only affected by the brine salinity or the initial water saturation but also by characteristics of the porous medium such as relative permeabilities and wettability. Thus, different types of formations may be more or less prone to permeability reduction due to salt precipitation. Most existing experimental studies related to salt precipitation during CO₂ injection are performed on sandstone cores. Among 14 studies reviewed by (Miri and Hameiri, 2016), only two had targeted carbonate specimens, i.e., dolomite (Ott et al., 2013) and limestone (Bacci et al., 2013). Using a microfluidic setup, (He et al., 2019) showed the effect of wetting conditions on salt precipitation patterns. Water-wet surfaces were more likely to form precipitates in the pore throats, thereby blocking fluid flow. Hydrophobic surfaces showed precipitation within the pore space with no effect on the injectivity. Salt precipitation may also become more or less important depending on the geological peculiarities of the storage site. For instance, fractured reservoirs were shown to respond differently depending on the fracture type and the relevance of the flow conductivity through the matrix (Sokama-Neuyam et al., 2020). Fractured reservoirs where the flow is conducted by both matrix and fractures were much more susceptible to injectivity impairment compared to reservoirs with dominant fracture flow. Fracture aperture (Sokama-Neuyam et al., 2020) and the effective stresses normal to the fracture (Lima et al., 2020) were also shown to influence the injectivity impairment caused by salt precipitation. Salt precipitation does not only depend on the properties of the storage site or formation water but also the conditions of the CO₂. The presence of impurities in the gas stream may affect the solubility of water in the CO₂ and the overall drying rate. The injection of colder CO₂ may also decrease the risk of salt precipitation. The mere effect of temperature on the CO₂ properties was shown to contribute to the pressure developed within the reservoir more than any salt blockages (Zhao and Cheng, 2017); this depends, however, on the manner the amount of precipitated salt, porosity, and permeability reduction are related. Moderate changes in porosity can lead to large changes in permeability (Peysson et al., 2011) and existing studies show that a 5% decrease in porosity can reduce the permeability up to 80-90% (Bacci et al., 2013, 2011). In some cases, a reduction in the absolute permeability did not decrease the CO₂ effective permeability because the rock became more water-wet after CO₂ injection (Ott et al., 2011). Pore-scale visualizations (He et al., 2019) showed that the decrease in the porosity does not always affect permeability, but the place where salt accumulates dictates the extent and relevance of injectivity impairment. The presence of heterogeneities challenges the assessment of the spatial distribution of salt within the porous medium (André et al., 2014). Numerical models that include correlations or relationships linking changes in porosity and permeability (Bacci et al., 2013, 2011; Liu et al., 2013) (e.g., “the tube in series” approach (Verma and Pruess, 1988)) are usually used to predict the

extent of salt precipitation and its effect on injectivity (André et al., 2014; Bacci et al., 2013; Roels et al., 2014; Tang et al., 2018). However, these models are not always consistent with the experimental observations; a kinetic approach for the drying effect (Roels et al., 2014) or non-isothermal and thermo-hydro-mechanical-chemical models (Salimzadeh and Nick, 2019; Zhao and Cheng, 2017) may improve the prediction of the salt precipitation. Besides relying on numerical models, measurement of seismic waves velocity together with electrical resistivity were shown to be a possible monitoring tool for salt precipitation (Falcon-Suarez et al., 2020). If the occurrence of salt precipitation is identified promptly, mitigation measures could be adopted.

The migration of fines (either clays or solids released during dissolution) can also lead to injectivity issues (Xie et al., 2017). Experimental studies on sandstones (Sokama-Neuyam et al., 2017b, 2017a) have shown that a relatively small amount of fines was necessary to impair injectivity. Fines and salt precipitation can have a combined detrimental effect, e.g., if salt deposits shrink the pore throats, these can become blocked by fines more easily. A few studies (Khather et al., 2020; Yang et al., 2018) on chalk interpreted the decrease in the permeability at the outlet of the core during CO₂ injection as a sign of fines migration. The fact that permeability decreased only at the outlet does not mean that fines migrated only in that section of the core; at the inlet section, the negative effect of the fines could have been counteracted by mineral dissolution. This is consistent with the observation of (Khather et al., 2020) that showed that NMR T_2 distribution shifted to shorter relaxation times at both the inlet and outlet of a Middle Eastern composite chalk core subjected to carbonated water flooding.

Moreover, if CO₂ is stored in a depleted hydrocarbon field, the compatibility between the residual oil and supercritical CO₂ may also impact the flow in the near-wellbore area as already observed in several CO₂-EOR projects (Fu et al., 2018; Leontaritis and Ali Mansoori, 1988; Sarma, 2003) and coreflooding experiments (Khather et al., 2019; Zanganeh et al., 2012). The mixing of CO₂ with petroleum and hydrocarbon systems shows complex multiphase behaviour with up to 4 phases (VLLS) in equilibrium, including solid and heavy liquid phases such as asphaltenes. Shelton and Yarbrough (Shelton and Yarbrough, 1977) were, possibly, the first in publishing a detailed phase diagram for these systems (Figure 15). Barotropic phenomena with phase density inversion, which can be detrimental when appearing within oil reservoirs, have also been reported (Quiñones-Cisneros, 2004). The supercritical behaviour of CO₂ may also play a role in the solubility of heavy compounds. The asphaltene content is not necessarily governing the behaviour, and it is well known that low asphaltene crudes may show severe problems if the organics are destabilized (Sarma, 2003). Yonebayashi et al. (Yonebayashi et al., 2019) provided an overview of 28 cases of CO₂-induced asphaltenes precipitation in reservoir fluids with asphaltene contents down to 0.07%. Higher pressures are expected to be beneficial as they promote redissolution and disappearance of the solid material (Monger and Fu, 1987; Verdier et al., 2006); this can be explained by an increase in the solubility parameter of the oil-CO₂ mixture as the molar volume of the reservoir fluid decreases with increasing pressure, thereby causing the dissolution of organic solids such as paraffins, naphthenic acids, and asphaltenes. However, for CO₂-crude oil systems, the organic precipitates may not completely disappear. Specifically, crude oil from the South Arne field (Denmark) was shown to release a heavy phase fraction in the presence of CO₂ (Christensen and Pedersen, 2005). PVT experiments indicate that solid organic phases appear at CO₂ contents above approximately 20-30% (Deo and Parra, 2012). The CO₂ molar fraction triggering precipitation is affected by both temperature (Verdier et al., 2006) and crude oil composition (Deo and Parra, 2012), which can partially justify the

differences in the observed CO_2 fraction range required for the onset of solids precipitation reported in several publications (Afra et al., 2020; Monger and Fu, 1987; Srivastava and Huang, 1997).

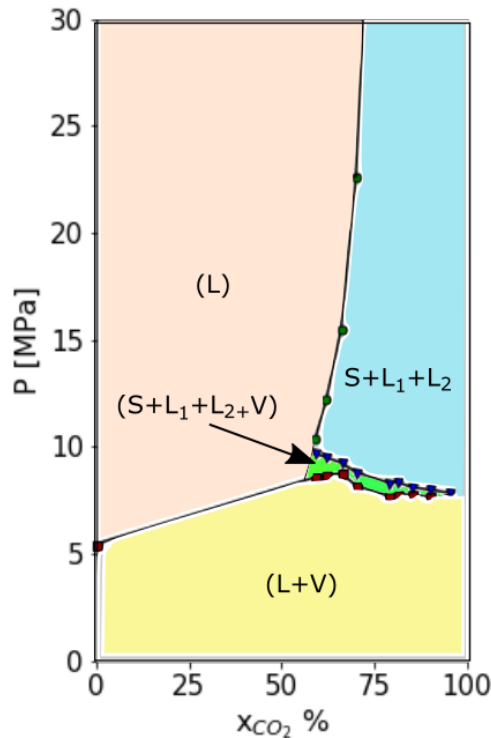


Figure 15. CO_2 -Crude oil phase diagram after (Shelton and Yarborough, 1977). Note the formation of solids phases at high CO_2 content.

The few experiments reported for tight chalk reservoirs in combination with CT scanning reveal that substantial deposition may take place when crude oil mixes with CO_2 (Okwen, 2006). The extent of deposition was shown to be affected by the presence of water (Monger and Fu, 1987), pore size (Qian et al., 2019), pore throat structure (Wang et al., 2020), and permeability (Srivastava and Huang, 1997) and less by the content of impurities in the CO_2 stream (Srivastava and Huang, 1997). However, these experiments are difficult to perform and, often, researchers have erroneously destabilized the oil by mixing it with CO_2 before injection in the core plug. In these cases, flocs may be large and the core entrance will serve as a regular filter leading to plugging by filter cake development without being representative of the reservoir behaviour. To observe the real behaviour, these experiments should be performed as a true gas injection on presaturated cores. The experiment may also be performed by premixing fluids to be stable but close to the onset and outside the phase transition region, load the live oil system onto the core, and then destabilize by final CO_2 injection or a change in pressure.

Although the phase behaviour issues for CO_2 -crude oil systems have been known for 40+ years, the research on this topic is surprisingly limited and scattered in the literature. In most cases, formation damage appears in the vicinity of production wells due to a large flux of organic solids and pressure drop caused by the increased income of particles and the potential high CO_2 content if under miscible conditions. In the Rangeley field operated by Chevron, CO_2 has been injected for many years and issues in production wells result in the need for replacement of submersible pumps as often as every two weeks. Besides the negative effect on the injectivity and the operational challenges, the deposition of

asphaltenes may also change the mineral wettability (Li et al., 2019) and therefore the relevance of the trapping mechanisms and CO₂ plume dynamics.

High pressure and relatively low temperatures are also suitable conditions for the development of CO₂ hydrates (Gauteplass et al., 2020), which can also block the fluid paths. This could be especially relevant in several cases, i.e., oil reservoirs that have been cooled down by waterflooding, injection of cold CO₂, injection of CO₂ into (low pressure) depleted reservoirs which could lead to Joule-Thomson cooling effect (Hoteit et al., 2019; Li and Laloui, 2016).

Lastly, the reactivity of carbonates in presence of CO₂ and water can also affect injectivity. The existing data on the evolution of the petrophysical characteristics of chalk upon injection of CO₂-saturated water are mixed. Whereas some studies (Alam et al., 2014; Høgnesen and Johansen, 2007; Olsen, 2011, 2010; Soong et al., 2016) observed no significant changes in porosity/permeability of chalk (Figure 16), others (Olsen, 2009, 2007) report dissolution features with increased permeability close to the inlet.

Dissolution events at the inlet can enhance injectivity by increasing the permeability; however, if this is too vigorous and leads to wormholes penetrating larger lengths, it may endanger the storage integrity by inducing weakening. X-ray CT scan imaging revealed the development of wormholes in limestone (Seyyedi et al., 2020), dolomite (Khather et al., 2019), and chalk (Khather et al., 2020) samples exposed to CO₂-saturated brine. Although numerical simulations using high-resolution digital models of North Sea chalk discarded the development of wormholes with CO₂ solutions and advocated for a homogeneous dissolution near the wellbore, the increase in the porosity at the inlet can still induce mechanical effects or mobilize contaminants (Yang et al., 2017). Additionally, by upscaling the changes in permeability caused by rock/CO₂-saturated water interactions observed at the core-scale, (Wang et al., 2019) showed that at the field-scale, because of a higher pH buffering capacity, mineral reactions will have a lower impact on the overall permeability evolution. For this reason, porosity-permeability relationships obtained from coreflooding experiments should not be directly implemented in large-scale models.

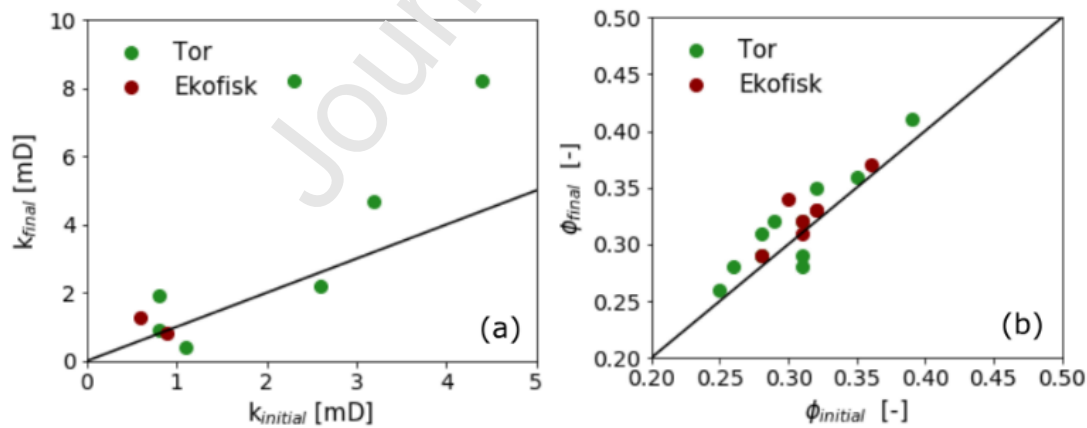


Figure 16. Effect of supercritical CO₂ injection on the (a) permeability and (b) porosity of chalk samples. The measurements were performed before (initial value) and after the experiments (final value). Data from (Alam et al., 2014).

2.3.2. Biological effects

Despite their extreme conditions, hydrocarbon reservoirs and saline aquifers host a variety of microorganisms. The injection of new electron acceptors or donors may induce changes in the microbial community, impacting both the injectivity and storage mechanisms (Kirk, 2011; Kirk et al., 2016; West et al., 2011). Biofilm formation caused by CO₂ injection is stimulated in the presence of minerals (Kirk et al., 2016). These biofilms can alter formation properties (e.g., porosity, permeability, wettability) (Jahanbani Veshareh and Ayatollahi, 2020; Morais et al., 2020; Mu and Moreau, 2015) and also impact the geochemistry (e.g., carbonate precipitation) by changing the pH and redox conditions (West et al., 2011; Zambare et al., 2020). Thus, although biofilm may decrease injectivity, it also benefits the CO₂ trapping by promoting precipitation and mineralization (Kasina et al., 2017; Kirk, 2011).

Several experimental studies (Kirk et al., 2016; Mayumi et al., 2013; Peet et al., 2015) showed that some bacteria strains can grow and adapt at high CO₂ partial pressures, whereas other communities may decrease (Yang et al., 2016). The bacterial growth and its effects at a storage site will be highly dependent on the availability of organic matter. Therefore, it may have different relevance for hydrocarbon fields compared to saline aquifers.

Methanogens, a group of microorganisms that use CO₂ or HCO₃⁻ as their electron acceptor and H₂ as their electron donor, are more likely to increase their activity upon CO₂ injection in (depleted) hydrocarbon reservoirs where fermentators create a source of H₂ production. Several studies (Golding et al., 2013; Ma et al., 2018; Mayumi et al., 2013; Tanaka et al., 2014) suggest that high CO₂ partial pressures can accelerate methanogenesis and transform the crude oil/coal into methane. If the H₂ source for methanogens activity is unlimited, the biofilm can reach thicknesses up to 240 µm (Droste et al., 1990) and occupy up to 80% of the pore volume (Ebigbo et al., 2013). Shelton et al. (Shelton et al., 2018) investigated the effect of CO₂ on the microbiome of a hydrocarbon reservoir (Olla Oil Field in LaSalle Parish, Louisiana, USA) microbiome 40 years after CO₂ injection. By extracting and comparing DNA from water samples obtained from sectors affected by CO₂ injection to others that had not been exposed to CO₂, they did not see a significant difference in the microbiome. However, given the decay rate of $[1e^{-3}-1e^{-2}] \text{ hr}^{-1}$ for active microorganisms and $[1e^{-5}-1e^{-3}] \text{ hr}^{-1}$ for inactive microorganisms, any potential change in the microbiome during CO₂ injection could have been diminished over 40 years. (Ma et al., 2019) studied the microbiome of soil samples collected from CO₂ leakage points close to an injection site in Gaoqing County, Shandong Province, China. They showed that an external CO₂ disturbance can significantly impact the structure of a microbial community and its interactions.

The CO₂ geological storage can also increase the activity of sulfate-reducing microorganisms (SRM). This was the case at the onshore Ketzin storage site where precipitation of iron sulfide in the near-wellbore area affected the injectivity (Zettlitzer et al., 2010). In (depleted) hydrocarbon fields, SRM can cause reservoir souring (Jahanbani Veshareh and Nick, 2019), which may induce corrosion processes and threaten storage safety (Morozova et al., 2011). The ammonium content in the produced water of a Danish reservoir with souring issues (Halfdan oil field) (Vigneron et al., 2017) shows that the activity of SRM is most likely phosphorus limited. The dissolution of chalk during CO₂ injection may release phosphate, which could increase the growth rate of SRM. DTI (Danish Technological Institute, 2011) enriched formation water samples with PO₄³⁻ to study its effect on SRM but did not observe any increase in the number of cells due to the phosphate addition. This observation should be treated with caution as the limited cell growth may be explained by the lack of an organic matter source in their experimental system. In a (depleted) hydrocarbon reservoir with access to organic matter, the increased phosphate

levels may increase methanogen and SRM activity significantly, leading to bioclogging and loss of injectivity.

2.3.3. *Mechanical effects*

The CO₂ conditions and injection procedure can lead to (geo)mechanical issues within the wells and reservoir with a detrimental effect on the injectivity (Torsæter and Cerasi, 2018). Excessive effective stress can reach the failure strength near the borehole wall in uncased sections and cause wellbore collapse. If CO₂ is injected intermittently, the cycles of pressure increase followed by well shut-in can cause fatigue with negative implications for the borehole stability (Cerasi and Walle, 2016). This will lead to a narrow injected pressure range required to avoid wellbore failure. Moreover, as observed in coal reservoirs, mechanical fatigue is also caused by cyclic saturation, which has a greater effect on the pore structure and mechanical properties (e.g., uniaxial compressive strength (UCS) and elastic modulus) of the rocks compared to sustained saturation (Su et al., 2020). Thus, an intermittent injection may increase the potential risk of shear and compaction deformations that become critical near the borehole and faults with influences in injectivity due to the change in pore structures.

Because of their low permeability, chalk formations may also be exposed to hydraulic fracturing when injecting high-pressure fluid (Fu et al., 2017; Papanastasiou et al., 2016; Salimzadeh et al., 2019). Fractures developed vertically may increase the risk of leakage by allowing vertical migration of the CO₂ whereas horizontal fracture propagation can improve injectivity. Although several studies suggest that CO₂ injection is more likely to have a positive effect on the injectivity (Kim et al., 2018) and result in horizontal fractures (Papanastasiou et al., 2016), simulations using digital models of chalk showed the development of fractures perpendicular to the flow direction (Yang et al., 2018). The fracturing can be further enhanced by temperature differences between the formation and injected CO₂ (Vilarrasa et al., 2017) or by changes in the CO₂ phase caused by pressure reductions (Zhou et al., 2017). The temperature difference between the colder injected CO₂ compared to the warm reservoir could also result in stress variations that can damage the well, threatening its integrity by potentially extending pre-existing defects creating leakage pathways from the storage reservoir to the overlying strata (Roy et al., 2018). Moreover, repeated thermal loading is another potential source of mechanical fatigue failure in wellbores influencing the injectivity.

The potential risks (e.g., leakage path) and benefits (increased injectivity) associated with these fractures could be assessed through numerical modelling. The development and the extent of these mechanical effects are highly dependent on the mechanical properties of the chalk exposed to CO₂. As previously discussed in section 2.2.1, unconsolidated rocks such as high porosity chalk can suffer from excessive deformation and weakening upon injection of CO₂ leading to strength reduction in the shear, transitional, and compaction regimes. However, studies conducted on reservoir samples point out a comparable behaviour of chalk saturated with water and CO₂-containing water.

Experience gained from demonstration projects, e.g., Snøhvit, In Salah, shows that geological features such as compartmentalization and connectivity strongly affect pressure development. These features are most likely less known for saline aquifers that are (geologically) poorly characterized but not expected to be an issue for depleted oil fields, especially the waterflooded ones.

2.4. Monitoring

The subsurface storage of CO₂ needs to be monitored to ensure that the injected CO₂ stays within the reservoir and that there is no breach of the top seal or leaks via faults and wells. By monitoring the movement of the CO₂ plume, it is possible to update dynamic models and take early measures. On top of this, there are legal requirements. The total mass balance needs to be monitored (injected equals the actual stored amount) and even small amounts of CO₂ leaked to the surface/groundwater must be quantified as stated in the emissions trading system of CO₂ quotas (Wartmann et al., 2009). Furthermore, there are other legislations such as those towards marine life.

Several technologies are commonly used to monitor the movement and potential leakage of CO₂ (Figure 17). The most commonly used is still time-lapse seismic (4D seismic), which is based on the comparison of seismic data recorded over the same area at different times. 4D seismic gives a spatial coverage of the subsurface for both the reservoir and the overburden and potential leakage through faults (Johnston, 2013; Yilmaz, 2003). There is a major change in seismic velocity, and therefore in the magnitude of the signal, as brine is substituted by brine with even a low CO₂ saturation. However, when the CO₂ saturation of the brine continues to increase the corresponding signal change is much smaller. 4D seismic is therefore very good at identifying leakage but less useful in quantifying it (Furre et al., 2017).

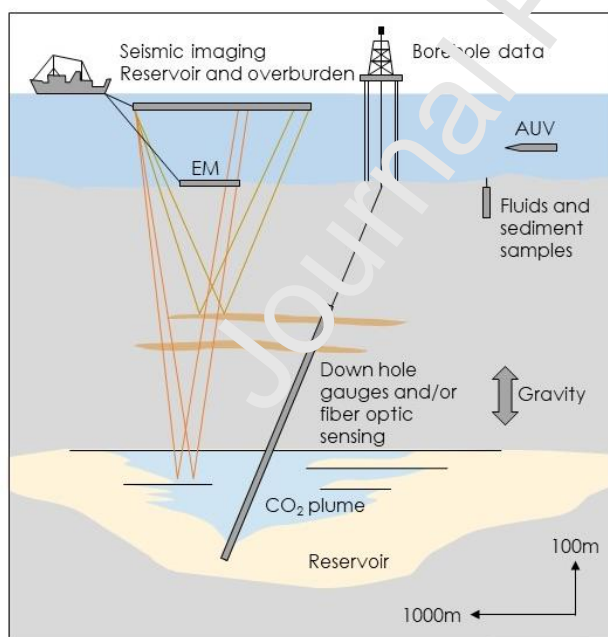


Figure 17. Overview of some monitoring techniques for offshore CO₂ storage. Adapted from (Ringrose, 2020).

Seismic technology has been used extensively for several areas such as Sleipner, Norway (Arts et al., 2004), Weyburn field in Canada (White, 2009), Ketzin, Germany (Schilling et al., 2009), Decatur field, United States (Finley, 2014), Otway and Naylor fields, Australia (Pevzner et al., 2011; Urosevic et al.,

2011, 2010), and New Zealand (Bannister et al., 2009). Close to the wells, data can also be collected using vertical seismic profile (VSP) either using conventional geophones or using Distributed Acoustic Sensing (DAS) that uses fibre optics cables in the well. The latter has the advantage of being employable in normally inaccessible wells and it can furthermore give measurements along the entire well (Grandi et al., 2017). While there is no direct experience of using 4D seismic for monitoring CO₂ within the Danish North Sea, there is extensive experience in utilizing 4D seismic for oil and gas production and optimization. 4D seismic has and is still being used with great success in fields such as South Arne (Lüthje et al., 2013), Dan (Zaske et al., 2013), Halfdan (Calvert et al., 2014), and Valdemar (Calvert et al., 2018).

When brine in the reservoir is replaced with lighter CO₂, the gravitational field changes and similar to 4D seismic this gives knowledge about the movement of the CO₂ plume. This has a poorer resolution but comes at a lower cost compared to 4D seismic. Also, since saturation is directly related to density, gravity monitoring can help to quantify changes (Furre et al., 2017). Reservoir-scale time-lapse gravity has also been used extensively for monitoring many fields (Alnes et al., 2008) but can also be applied in wells to give a high level of resolution within the area close to the well (Jvindhagen et al., 2005).

Another geophysical technique, especially useful in combination with seismic and gravitational measurements, is controlled-source electromagnetic (CSEM) surveying where the electromagnetic field is recorded (Tveit et al., 2018). The presence of free gas in the subsurface can potentially be seen as a local change in the recorded signal (Berndt et al., 2020). To limit the amount of noise in the signal, methods such as surface-to-borehole CSEM, where the signal is recorded in the wells, are currently being investigated (Park et al., 2013).

The seepage of CO₂ through faults, wells, or directly through the overburden can be surveyed using tools such as autonomous underwater vehicles or fluid and sediment samples to look for biological and chemical changes. In general, these methods face the problem of covering large areas and of knowing exactly where the CO₂ is coming from. One technique to overcome this is to add chemical tracers to the CO₂ even though this has not been done on a large-scale before (Berndt et al., 2020). Since seepage can occur directly through the wells, several measuring techniques are used, e.g., measuring pressure and temperature changes, directly in the wells. Moreover, corrosion sensors and cement bond logging are used to specifically assess and monitor well integrity (Zhang and Akemu, 2010).

Since reservoir chalk is highly compressible, monitoring subsurface deformation will be required during CO₂ injection (Schutjens et al., 2019). This type of monitoring has already been in place in the Danish North Sea for over four decades. Direct strain measurements are carried out using formation subsidence monitoring tool that consists of repetitive measurements over time of the distance between radioactive bullets implanted every 5 to 10 m in the formation along the wells. A historical dataset of the seafloor subsidence is also available in many fields from the North Sea and includes several decades of measurements (Angus et al., 2015; Hatchell et al., 2007; Hettema et al., 2002; Kristiansen and Plischke, 2010). The GPS systems are present in platforms and can be re-deployed to monitor seafloor displacement after the onset of CO₂ injection. Permanent sensors installed on the seabed can also be used to measure vertical and horizontal displacement of the seafloor. Therefore, monitoring potential reservoir deformation during CO₂ injection supported by already-built infrastructures and the existing dataset on past deformation can be instrumental for reservoir management during the lifetime of the platforms/infrastructures.

Currently, development into greater use of machine learning in interpreting data is considered as one of the most promising emerging technologies for CO₂ monitoring (Personal Communication with Eliasson and Jordan, 2020), as well as greater development into technologies such as DAS (Grindei et al., 2019). Other methods exist but these have their main use within on-shore CO₂ storage and are, therefore, not discussed here.

2.5. CO₂ storage in saline aquifers vs. depleted hydrocarbon fields.

Although many sites may be suitable for CO₂ storage, the diverse physical and chemical conditions at these geological spots will eventually dictate how the CO₂ is stored. Accordingly, the trapping mechanisms and the challenges encountered in the storage in saline aquifers will be different than those in depleted oil fields or CO₂-EOR projects.

The injection of CO₂ in saline aquifers may be limited to one or only a few wells due to the lack of pre-existing infrastructure and the high costs associated with the drilling operations (especially in offshore locations). This can considerably restrict reservoir management strategies to optimize the storage efficiency. Because of the high salinity of the formation water of these aquifers and (expected) water-wet behaviour, the formation of salt deposits is an expected threat to the injectivity. In the same way, the high salinity decreases also the CO₂ diffusion and solubility in the solution, which may eventually inhibit the density-driven convective mixing; this may not only retard but also reduce the overall solution trapping. On the other hand, the great amount of water and water-wet conditions may be favorable for capillary trapping. Thus, in saline aquifers, a significant fraction of the injected CO₂ is expected to be stored as a residual disconnected phase. At longer timescales, the aqueous chemistry, dictated also by the mineral phases composing the host rock, may also lead to suitable conditions for mineralization.

Depleted oil fields, despite bearing lower storage capacity compared to saline aquifers, have in place infrastructure that could be reused for CO₂ injection (provided that this infrastructure is or can be easily retrofitted to be compatible with the CO₂ stream). Moreover, these sites are well characterized from previous O&G operations, which will ease the upscaling of CO₂ storage from core to field scale. Nonetheless, the injection of CO₂ in depleted oil fields may incur several operational challenges. If the reservoir has initially a very low pressure (relevant to depleted gas fields), the CO₂ should be injected in a gas phase to avoid a great temperature drop caused by CO₂ expansion (Joule-Thomson effect) with detrimental consequences for the wellbore integrity. Once the reservoir pressure increases, a switch to liquid CO₂ injection would be required. Because of the lower ionic strength of the formation water of depleted oil fields, salt precipitation is not, apriori, expected to pose the same risk as in saline aquifers. In this case, chemical effects such as asphaltene precipitation and biological effects will most likely take over and represent a threat when CO₂ is injected in depleted oil fields; the importance of this threat may be proportional to the residual oil saturations and its properties. The lower ionic strength of the formation water can also be positive for the overall solution trapping by promoting the CO₂ diffusion, solubility, and convective mixing; nonetheless, the very low permeability of many chalk fields may act towards minimizing the development of convective fingers. Additionally, the availability of many wells can be strategically used to inject the CO₂ at places with formation water undersaturated in CO₂ so to maximize solution trapping; yet, higher number of well penetrations may also increase the risk of CO₂ leakage. The presence of a residual hydrocarbon phase may also be beneficial for the trapping as a

fraction of the injected CO_2 may diffuse and dissolve in the crude oil. Additionally, in this three-phase system (i.e., residual oil, water, and CO_2) under oil/mixed wet conditions, the CO_2 may be the intermediate-wetting phase and trapped in smaller pores or pore corners with reduced mobility. However, when the remaining oil saturation is not enough to assist in the capillary trapping of the CO_2 more emphasis will be placed on the structural trapping. Fortunately, opposite to many saline aquifers, depleted oil fields are known to have an effective caprock seal that, apriori, should be suitable to offer capillary resistance to CO_2 flow. An additional positive effect of the residual oil coatings is that they may diminish the carbonated water weakening of the chalk and its effects on the reservoir geomechanical behaviour.

At the moment, most CO_2 storage initiatives are run along with EOR projects. CO_2 storage activities + EOR predominate as the profit obtained by the additional oil recovery can compensate for the cost related to the storage. While the trapping mechanisms in CO_2 -EOR projects will be similar to those in depleted oil fields, the higher hydrocarbon saturation (compared to depleted fields) present in the reservoir will pose a higher threat not only to the injectivity but also the operations overall (e.g., in the production lines). If the reservoirs have been previously waterflooded and cooled down, the injection of CO_2 may also lead to hydrates formation. Despite the operational challenges, the production of hydrocarbons while injecting CO_2 will increase the available space for CO_2 storage.

3. Final remarks and implications for CCS deployment in chalk

CCS has recently been publicly embraced in Denmark, where several climate and energy institutions emphasize that capturing and storing CO_2 is essential for the mid-term achievement of the emission reduction targets and for becoming carbon neutral in the long term. Re-using the O&G fields for CO_2 storage entails several advantages (e.g., existing infrastructure and subsurface data, more social acceptance for offshore locations) which can conciliate Denmark's ambition of materializing CCS starting in 2025. However, the lack of CCS/ CO_2 -EOR projects implemented in chalk, the predominant lithology of the Danish hydrocarbon fields, may hamper the implementation of CCS in this type of formation. Although the existing experimental investigations on CO_2 -EOR in chalk represent a valuable source of information, CO_2 storage requires additional specific studies to reveal long-term behaviour/aspects. The main conclusions and suggestions to ensure safe CO_2 storage in chalk formations are summarized hereafter:

1. When depleted oil fields are considered for storage, it is usually assumed that the caprock will have analogous sealing properties for the fluid to be stored, e.g., CO_2 , as it initially had for the in-situ hydrocarbons. Yet, CO_2 may interact both physically and chemically with the caprock, which could impair its sealing properties. Specifically for the Danish chalk fields, the limited cored caprock material leaves the interactions between CO_2 and the overlying shales unexplored. However, the experimental results obtained recently on shales from the same geological formation are promising as they show that these can seal a large column of CO_2 .
2. The density-difference (between saturated and non-saturated CO_2 formation) driven convection is expected to be less relevant in chalk formations given their low permeability. However, the fairly low salinity of the depleted oil fields compared to saline aquifers will lead to greater CO_2 solubility and diffusion. The absence of convective patterns may be beneficial in carbonate formations as fingering may result in non-homogeneous dissolution leading to wormholes.

3. Experimental evidence suggests that the capillary-trapped CO₂ could reach around 40% of the pore volume. However, the potential for capillary trapping depends strongly on the wettability conditions and initial water saturation. This means that different chalk fields may have a different capillary trapping potential depending on their formation water and residual oil properties. Given their greater water saturation, waterflooded fields are expected to have a greater capillary trapping potential. However, the available pore space for CO₂ will also be lower. On the other hand, depending on the temperature, pressure, and residual oil saturation (and its properties), CO₂ may also dissolve and become trapped in the remaining oil.
4. The overall potential for mineral trapping of chalk is expected to be lower compared to that of sandstones because of the lower availability of feldspars to react with the CO₂ and form secondary phases. In a relatively clean carbonate formation, the dynamic dissolution/precipitation episodes are expected to keep an overall neutral balance of newly precipitated CaCO₃. Moreover, an increasing number of studies point out that calcite reactivity cannot be extrapolated to chalk. This means that the kinetics of calcite dissolution/precipitations cannot be used to describe the behaviour of chalk. Besides this, the geochemical reaction kinetics in depleted oil fields will be most likely affected by the presence of oil coating the mineral surface. The dissolution/precipitation reactions in chalk formations are not only important to assess mineral or solution trapping but they also have an impact on the porosity, permeability, and mechanical properties; these factors will eventually affect the migration of the CO₂ plume and the integrity of the storage site.
5. The effect of CO₂ on the mechanical properties of chalk depends greatly on the experimental conditions and characteristics of the sample (e.g., texture, grain size). Although the evidence is mixed, the studies on reservoir samples showed that CO₂ does not weaken the chalk. The chemical effects (e.g., dissolution) leading to weakening become more relevant at non-equilibrium conditions. At the field scale, these conditions will most likely occur near the wellbore, therefore, chalk weakening may become especially relevant for borehole stability and injectivity. However, simultaneous effects near the wellbore, e.g., drying and water vaporization due to CO₂ injection, may impact positively the strength of the chalk.
6. To benefit from the number of wells in depleted O&G fields, the selection of CO₂ injectors can be made based on their remaining “sacrificial lifetime” (time left before abandonment considering the corrosion rates in presence of CO₂); when this is reached, the well could be abandoned by co-injection of CO₂ and chemicals to shift the environment towards alkaline conditions, promoting precipitation. Nonetheless, the availability of wells can only be seen as an opportunity once an integrity study has been carried out, guaranteeing the compatibility of the wells with the CO₂ injection conditions. Although cement degradation has been frequently associated with a great risk for well failure, thus leakage, several studies have shown that Portland cement not only is suitable for CO₂ applications but is also less affected when embedded in carbonate formations. Moreover, (pre-) existing microannulus or fractures have the potential to self-heal in the presence of carbonated water. However, less is known about the interactions at the cement/caprock interface or the influence of corrosion products on the behaviour (or the healing) at the casing/cement interface.
7. Injectivity issues are commonly reported at CO₂ storage sites. These issues may be even more relevant for low permeability chalk fields. Several synergistic chemical (e.g., salt precipitation, rock

dissolution, fines migration, asphaltene deposition), mechanical (chalk weakening, compaction), and biological (e.g, bioclogging) phenomena are expected to impact the injectivity. Both their individual and combined effects need to be assessed so that the injectivity can be predicted and solutions and mitigation measures are proposed beforehand. The loss of injectivity is more detrimental for dedicated CO₂ storage sites that have few wells and may require interrupting the injection until the permeability is restored; for depleted oil fields, the injection could be switched to a different well.

8. Although 4D seismic is a reliable tool for monitoring several storage sites, the cost can challenge the economical feasibility of CCS projects. The further development of controlled-source electromagnetic surveying and distributed acoustic sensing together with the increasing use of machine learning in designing the monitoring and interpreting the data are all expected to decrease the costs. Moreover, some of the existing wells in the depleted oil fields could also be strategically used for monitoring purposes.
9. Performing CO₂ injection instead of waterflooding during the late-life operation of chalk oil reservoirs offers significant potential for increasing storage capacity while benefiting from the existing infrastructure (e.g., wells and surface facilities) and profound knowledge of the subsurface character and behaviour. This would also allow de-risking CO₂ storage in chalk formations in a more controlled and safe manner.

Some of the above conclusions, e.g., chalk reactivity, mechanical behaviour, and importance of trapping mechanisms are not only relevant for existing oil and gas chalk fields but also to other tight limestone reservoirs (e.g., Al Shaheen, Qatar) and underground chalk aquifers. Given the worldwide large chalk formations with high porosity and thick sealing units, their potential storage capacity should not be disregarded. Future research is encouraged to expand the possibilities for CO₂ storage to other “less conventional” lithologies such as chalk.

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Competing interests

There are no conflicts of interest to declare.

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Highlights

- A growing number of countries are contemplating CO₂ storage to reduce CO₂ emissions
- Existing research largely leans towards CO₂ storage in highly permeable formations
- Global deployment of CO₂ storage can benefit by diversifying the lithology options
- Porous chalk formations can represent reliable CO₂ storage sites.