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De-risking CO₂ storage in chalk formations

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Abstract

Carbon Storage in geological media has been widely considered as a solution to reduce the environmental footprints of anthropogenic emissions of CO₂ from point sources. The depleted hydrocarbon chalk fields in the Danish North Sea are potential candidates for carbon storage¹, as they comprise a significant pore volume and many are approaching the final stages of production lifetime. To develop cost effective and insignificant risk CO₂ storage solutions in depleted hydrocarbon chalk reservoirs and to unlock the main storage potential in Denmark a new multidisciplinary CO₂ storage research program² has been initiated at the Danish Offshore Technology Centre. The scope for CO₂ storage in carbonate reservoirs is potentially large as carbonate reservoirs, including limestones, dolomites and chalks, comprise one of the most prevalent types of hydrocarbon reservoir worldwide. However, the geochemical response of chalk and other fractured and layered carbonate rocks to CO₂ and the geomechanical consequences must be investigated carefully to de-risk a storage scenario. The research program includes modelling studies covering a wide range of coupled simulations including chemical reactions and transport phenomena at different length scales. In addition, the program comprises laboratory experiments on core flooding at representative reservoir temperature and pressure condition, geochemical analyses and geomechanical tests under similar in-situ conditions. The chemical processes that can take place at the surface of the rock in contact with natural formation water and dissolved CO₂ can lead to potential reservoir changes, both upon injection and over time. CO₂ may be injected as supercritical CO₂ in which case solvation of the gas will take place upon contact with the formation water in place, or it may be injected in the form of carbonated water which will lead to a direct acidification of the water in contact with the rock. The geochemical reactions between carbonate rock and dissolved CO₂ depend on the thermodynamic conditions, the salinity of the water, pressure and can potentially affect significant dissolution of the rock matrix either in the near well bore areas or in the wider matrix, or in both. Other factors that can affect the safety and efficiency of storage are the presence of residual hydrocarbons, specific lithography and local water chemistry. The consequences of dissolution can be amongst other things general geomechanical weakening, subsidence, leakage, permeability changes and diversions of flow. The dissolution can take place rapidly upon injection or more slowly during storage. The studies included in the research program address both short and long-term responses of chalk to CO₂ injection considering the relevant physical and chemical processes as well as longer term storage test in the laboratory. The aim is to identify the conditions under which CO₂ storage in chalk is feasible and sufficiently safe. In this paper we first highlight the main challenges and enablers for CO₂ storage in chalk fields in terms of the storage mechanisms, injectivity, risk, containment, and longevity. Secondly, we describe the progress of the established research program towards determining if chalk is suitable as a long-term storage reservoir for CO₂.

Keywords: Chalk reservoir; mature fields; fractured reservoir; injectivity; dissolution; containment

1. Introduction

A decade ago, a series of projects in Denmark addressed the use of CO₂ for Enhanced Oil Recovery from Danish North Sea chalk reservoirs. However, due to the economic feasibility of acquiring and transporting CO₂ the idea was given up before field testing. Large infrastructure investments in pipelines, wells, and platforms for oil and gas extraction from chalk fields have been made in the Danish North Sea [1]. Some of these chalk fields have been water flooded for improved recovery and are now depleting. Large savings are potentially achievable if the same infrastructure were reused and given a second life, serving as transport and storage sites for CO₂. The uncertainties with respect to viability and risk in connection with CO₂ injection and storage in chalk are still large and will need to be constrained. Aspects related to the reactivity and mechanical behavior of chalk exposed to CO₂ and carbonated brine and other related uncertainties have been thoroughly discussed in [1]. Chalk is more susceptible to being affected by CO₂ induced acidity than sandstones and limestones and therefore a de-risking study to understand the safe operating windows for CO₂ storage in chalk is a prerequisite for implementation.

This paper describes a research program [2] consisting of a series of experimental and modelling studies designed to investigate the operating window for safe and lasting CO₂ storage in spent North Sea Chalk fields from all angles. The work will start with mapping the known data available in literature relating to the rock response of chalk and carbonates to CO₂ storage and describe the chosen research approaches to answer the most important questions relating to CO₂ storage in chalk. The main outstanding research questions relating to the suitability of chalk for CO₂ storage include the geomechanical alterations of the chalk in the presence of CO₂ (Project 1), the injectivity alterations potentially caused by CO₂ (Project 2), the effects of fluid-fluid contact and oil swelling (Project 2), density driven flow effects on the CO₂ dissolution in formation brine (Project 3), chalk dissolution kinetics (Project 4) and nature of pore-scale surface alterations caused by dissolution (Project 5). Five research projects addressing these questions will be described in the following. To study the long term and field wide effects of CO₂ storage the geomechanical and geochemical data for chalk will be incorporated into large-scale modelling tool which will help estimate the capacity for CO₂ storage in Danish chalk. The model will be used to estimate contributions of different trapping mechanisms for reservoir-scale CO₂ storage in chalk formations, recommending operational conditions that avoid jeopardizing reservoir integrity and ensure safe long-term storage.

2. Effect of CO₂ injection on chalk's mechanical properties (Project 1)

Chalk is composed of compacted carbonate ooze, raising concerns for storage safety and reservoir integrity. A major concern for large-scale CO₂ injection is the potential effect of acid CO₂ on the calcite mineral constituting the chalk frame causing mechanical weakening of the reservoir. The risk of unwanted reservoir damage due to chemical rock-fluid interaction needs identification and critical evaluation. However, if the Danish chalk reservoirs maintain stiffness and strength properties upon CO₂ injection, or if the risks of the negative consequences are manageable, are significant cost cuts in transporting and storing CO₂ anticipated.

The mixing of CO₂ and water forms a weak acid that may dissolve calcite minerals until meeting a new buffered equilibrium, leaving unresolved questions on how the CO₂ affects the chalk frame when mixed with saline formation waters. The stiffness and strength properties of a rock frame depend upon the constituting grains and how these are held together, i.e., the cementation degree commonly expressed as Biot's effective stress coefficient. Whether chemical dissolution plays a significant mechanical role depends on the quantity of dissolved calcite for a given injection scenario and whether calcite cement in contacts would diminish or even grow in area. Reducing the grain contact area is effectively a softening- and weakening mechanism, whereas the opposite is strengthening the frame, although it could potentially also clog pore throats. Some experiments with supercritical CO₂ injection in chalk have shown to cause worm holes near the injection point in some cases, but overall, no pore-blocking or significant mechanical weakening was found [3]. However, existing experimental data are too limited to address the risk of mechanical damage due to calcite dissolution from CO₂ injection as highly relevant near-well and/or far-field issues of long-term effects, micro-quartz, clays, and temperature are not yet fully addressed.

Corresponding to a high specific surface area, North Sea chalks commonly possess high porosity, but low permeability and consequently surface effects are much more important in chalks than in, e.g., sandstones. Calcite

minerals have charged surface sites, where differently charged ions can adsorb from the saturating water phase, causing a net electrostatic potential. When two charged calcite crystals are close, i.e., in a rock frame, these electrostatic potentials overlap, developing a disjoining pressure [4]. A disjoining pressure between crystals in a rock frame is effectively a softening mechanism, and a range of experiments on brine exposed chalks and calcite have shown significant softening and weakening due to changes in electrostatic potential. However, other experiments have shown a reduced creep rate when CO₂ is injected with brine during continuous creep experiments, consequently implying strengthening of the chalk frame [5]. Thus, there is a need to determine if there is an actual mechanical integrity risk toward the near-well and/or far-field reservoir chalk.

In order to increase our experimental knowledge and enable evaluation and quantification of the potential extent of CO₂-induced changes in mechanical chalk properties, we planned a series of mechanical triaxial tests with an injection of finite amounts of supercritical CO₂ into brine saturated reservoir chalk cores. The experiments have injection at different effective stress levels to resemble different reservoirs and depletion levels. We use both elastic wave velocity and stress-strain curves to determine stiffness and strength changes before, during, and after CO₂ injection/exposure. Subsequently, Biot's coefficient may be derived as a quantitative estimate of cementation degree. Pore fluid samples obtained at strategic stages in the triaxial tests are analyzed for changes in chemical composition. These data are combined with analysis for changes in chemical composition from a designated long-term flooding experiment with alternating brine and supercritical CO₂ injection to provide detailed knowledge for geochemical modelling and impact on mechanical properties. Before triaxial and long-term flooding experiments, chalk samples were analyzed using conventional core analysis, and by use of NMR-spectrometry, we seek to quantify CO₂-induced bulk changes in pore size distributions. Analysis of pore size distribution and geochemical composition is assisted by petrography (Backscattered electron micrography images) of polished thin sections from cross-sections of tested core samples.

3. De-risking CO₂ injection and storage in chalk (Project 2)

The goal of this research is to conduct a series of experiments to investigate the operating window for safe and lasting CO₂ injection and storage in spent North Sea Chalk fields. The work will be initiated by mapping known data from literature on the rock response of chalk and carbonates to CO₂ storage to assist with designing the most useful experimental series that the project time allows. The CO₂ injection experiments will be carried out in high pressure setups that allow for changing the temperature to investigate the effects of temperature on any chalk dissolution caused by lower or higher CaCO₃ solubility. In addition, the resilience of the rock material to CO₂ in the presence or absence of water will be investigated. The resulting pH in the reservoir water phase of injecting CO₂ will also vary and experiments need to investigate this. Following injection, the changes in the rock mechanics incurred by CO₂ storage will be investigated for different static "storage" periods in the experimental setup, including in a triaxial cell. All experiments will include monitoring of effluents, and CT-scanning of the core material before and after. As many experiments as time will allow will be carried out with additional ultrasonic acoustic measurements of P- and S- wave velocities and dynamic X-ray CT imaging. Additional measurements of CO₂ solubility in different brines will be carried out in batch experiments and surface studies of wettability effects and chemical-physical changes to the rock samples will be investigated in the DTU laboratories after flooding.

First contact miscibility of reservoir fluids with CO₂ at reservoir pressure leads to immediate swelling of the oil phase. Exact fluid composition can affect this miscibility, and the proposed experiments will be done using crude oils with previously determined composition. The solubility of CO₂ in the relevant reservoir brine also needs to be measured by solubility tests at the system pressures and temperatures. The wettability of the rock (which for calcite is inherently "water-wet") depends on the characteristics of the organic layers adsorbed and the interaction of these layers with bulk hydrocarbon fluid. When considering injecting CO₂ for storage the effect of CO₂ contact with the surface layers and potential wettability change is important. Surface infrared techniques are powerful for probing the types of molecular bonds present in the adsorbed species and therefore the chemical functionality. The injection of CO₂ may over time also lead to changes in the fluid composition as the gas is miscible with lighter constituents. The potential for fractionation of the reservoir fluid by CO₂ gas contact should be considered as it may cause the fluid to be rendered heavier with time. This effect may be investigated by CO₂ extraction of heavy components in crude oil as a function of pressure. The stability and leaking potential of the stored CO₂ could be affected by changes in fluid

composition. The consequences for flow assurance and hydrate formation in the production pipelines will also be considered.

The effect of CO₂ injection in both dry core and in presence of various amounts of water as well as dissolved CO₂ – in reservoir chalk (use of core flooding instruments and CT scanner) will be tested. The effect of changes in CO₂/brine and CO₂ mixture/brine concentration as well as brine composition on flow properties of chalk will be investigated in a series of dynamic core flood experiments at reservoir conditions (high pressure/temperature) for both homogeneous and fractured core plugs. Effluent collection will provide a basis for chemical analysis including changes in effluent brine composition, crude oil effects as well as particle size measurement for detection of fines production. To see changes in rock microstructure and monitor fracture flow, samples will be taken for imaging using high resolution μ -CT before and after core floods and CO₂ aging experiments. We aim for ~10 (CO₂ and carbonated water) flooding experiments to cover a wide range of conditions (e.g., Temperature 30 and 70 deg C, with various initial oil saturation, seawater/formation water) for CO₂ injection in chalk. We will use different flooding setups for flooding core samples with varied sizes (1cm to 15 cm long). The initial set-up for static CO₂ aging (storage) of reservoir chalk can be seen in Figure 1. The chalk dissolution is observable in the ion chromatography measurements.

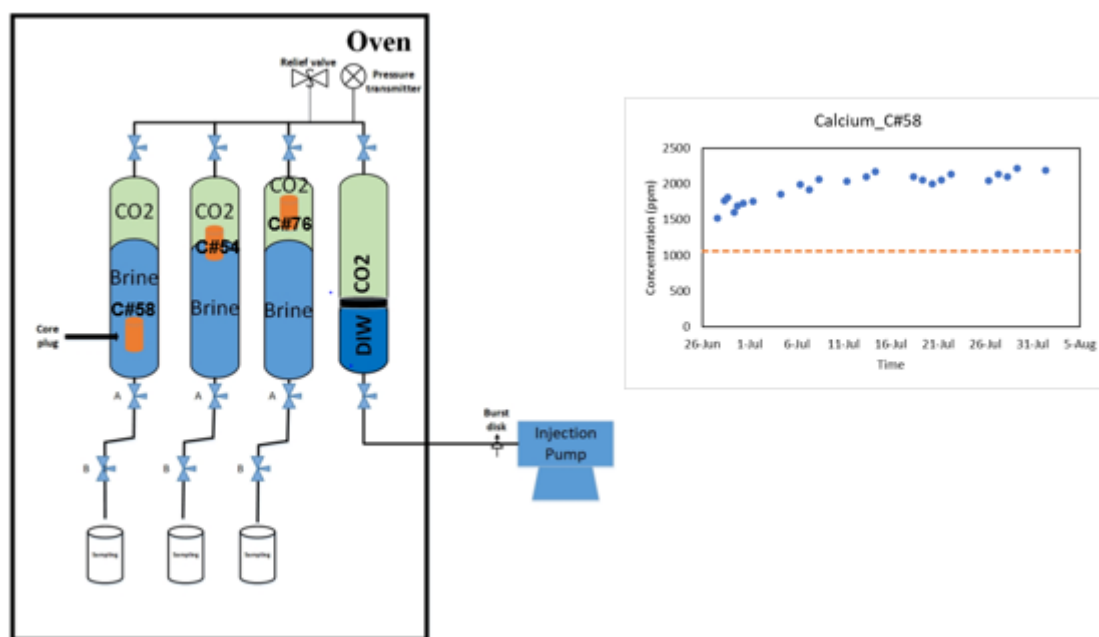


Figure 1. Set-up for static contact experiments with chalk core plugs and Brine in contact with, semi-contact and pure CO₂ under reservoir conditions. The Calcium ions indicate the chalk dissolution caused by carbonated brine.

This study combines water and CO₂ core flooding experiments on both homogeneous and heterogeneous chalk samples at varying pressure states with simultaneous acquisition of ultrasonic acoustic measurements of P- and S-wave velocities and X-ray CT imaging. Ultrasonic acoustic measurements will be conducted using transducers optimized for shear wave production, placed in an axial direction along the core. Alteration in P- and S- wave velocity will be monitored for various saturation conditions. These can be used for field scale seismic interpretations.

In addition, triaxial tests will be run on some of the core plugs after flooding with CO₂ or carbonated water to quantify any alteration in the chalk's mechanical properties. This will enable us to assess the impact on compaction, injectivity and wellbore stability. We will study the dynamic of fluid flow in chalk under variable pressure and temperature to establish a base chalk failure model for flooded samples. Using the stress-strain relation and velocity measurements, static and dynamic elastic moduli of rock, the stiffness, failure, and permeability change can be estimated. This data will be used as input for numerical modelling. The mechanical properties of the chalk (bulk

modulus, Young's modulus, ...) will be also correlated to the developed electrical charges on the calcite surface at different ionic composition.

Numerical simulation of CO₂/carbonated water flooding at core scale will let us predict the flow and sweep behavior of the studied methods for chalk matrix and fractures. The focus is to investigate the efficiency of both CO₂ and carbonated water flooding at different reservoir conditions at the small-scale. The small-scale model will also be used to perform history matching of the core flooding data (from previous tasks) and to obtain the upscaled parameters (e.g., relative permeabilities) and seismic responses. Advanced numerical platforms will be utilized to enlighten the short- and long-term responses of chalk formations to CO₂ by investigating: 1) Role of fractures on enhancing/hindering CO₂ movement in reservoirs under thermal and chemical stresses, and 2) The transport and trapping mechanisms contributing to two-phase flow in chalk formations.

4. Accelerated CO₂ dissolution as a game changer in fractured chalk (Project 3)

The dissolution of CO₂ increases the density of the formation brine. For the supercritical CO₂ overlaying a brine-saturated chalk formation, the high-density fluid on top of a lower-density brine creates an unstable system that leads to the downward flow of CO₂-saturated brine in the form of fingers. In a low permeable chalk system with permeabilities often lower than 1 milli Darcy ($0.00110^{-12} \text{ m}^2$), the density-driven dissolution of the overlaying CO₂ is expected to be slow occurring over centuries. However, the naturally fractured system in the Danish North Sea chalk reservoirs provides a large interfacial area between the injected supercritical CO₂ that invades the high permeable fractures and the formation brine that is kept in the low-permeable water-wet chalk matrix, shown schematically in Figure 2.

While the injection of carbon dioxide is ongoing, a large mass transfer area will be maintained between the CO₂ and formation brine, which will increase the dissolution rate by diffusion. Moreover, in the vertical planar fractures, the CO₂-saturated brine acts as a single finger that can start a convective flux in the chalk matrix. The convective mixing can enhance the dissolution rate, so it is possible to fully saturate the formation brine with CO₂ during the reservoir's lifetime, i.e., two to three decades. To prove this hypothesis, we will conduct an experiment, in which the curved area of a brine-saturated chalk cylindrical core is exposed to a fixed volume of high-pressure CO₂ at a constant temperature. The novelty of this experiment is that it provides a larger mass transfer compared with the previous experiments that overlay CO₂ phase on top of the porous medium [6–10]. Therefore, this experiment can be performed faster and thus in low permeable cores representative of a real field. The design of the experimental setup is shown in Figure 3. The experimental results are used to validate a numerical model that solves the reactive transport of the single-phase CO₂-brine mixture in a two-dimensional cylindrical domain. The density and viscosity of the fluid are concentration-dependent, which leads to a nonlinear system of equations. We linearize the system of equations and discretize them with a cell-centered finite volume scheme [9]. The dissolution of chalk is modelled sequentially in PhreeqcRM [12]. The preliminary results show that the large interfacial area provided by the fracture networks can lead to faster uptake of the injected CO₂ by dissolution in the water-saturated chalk matrix. The dissolution is still controlled by diffusion in low permeable chalk formations. However, at slightly higher permeabilities, e.g., 50 milli Darcy (0.0510^{-12} m^2), the advective flow becomes dominant and accelerates the dissolution of CO₂. In a fractured system with a fracture density of 1 fracture per meter, the dissolution rate is large enough to saturate the formation water with CO₂ within two to three decades of injection time [11].

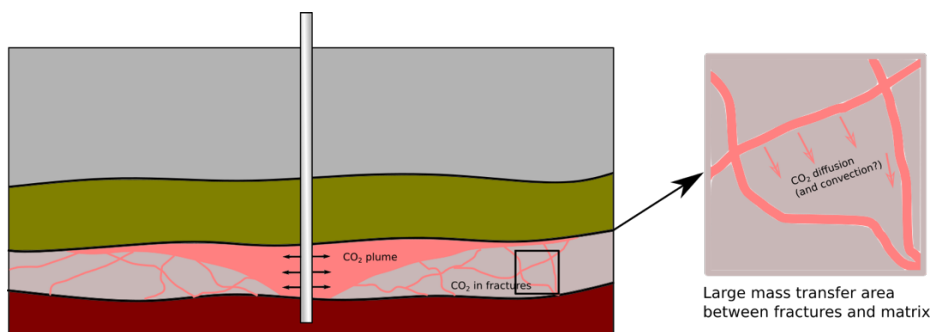


Figure 2. A schematic of the flow of CO₂ in a fractured system and the formation of large interfacial area between the gaseous CO₂ and the water-saturated matrix

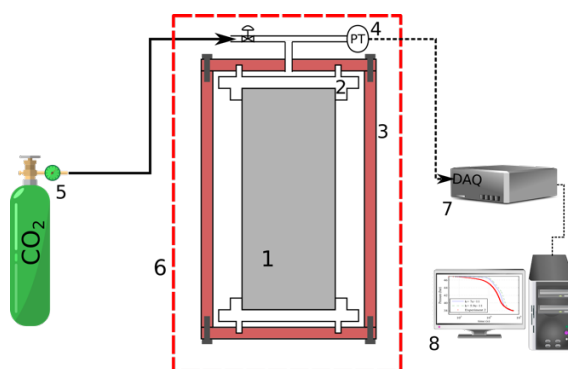


Figure 3. A schematic of the experimental set up: 1- water-saturated chalk core; 2- Teflon caps; 3- Core holder; 4- Pressure transducer; 5- CO₂ vessel; 6- oven; 7- data acquisition (DAQ) system; 8- data recording

5. Chalk reactivity during CO₂ injection in depleted reservoirs (Project 4)

Apart from the high offshore implementation cost and the low chalk permeability, a significant challenge for CO₂ storage in chalk is the higher reactivity of chalk with CO₂ compared to other types of rock (incl. carbonate). Chalk dissolution is expected to be the primary reaction. However, it is a fast reaction, and quantifying its kinetics is experimentally challenging. The apparent reaction rate depends on the reactive surface area, the density of the active sites, and the organic coating on the mineral surface. As observed in previous chalk studies, the complex dependence results in different chalk dissolution behaviors. Meanwhile, it is crucial to correctly estimate the extent of the chalk reaction in different scenarios (near the wellbore, far into the reservoir, in the presence of varying fluid phases) because the reaction has severe implications for injectivity, formation stability, and the development of leakage path.

It is well known that the reactivity of chalk deviates from its chemical counterpart—calcite crystal. We hypothesize that the chemical reactivity of calcite and chalk are indeed similar. However, three factors may lead to significantly different “apparent” dissolution rates:

1. Density of truncated surface crystallographic sites
2. Evolution of reactive surface area
3. Organic coating on the mineral surface.

In this study, we combine a pH-sensitive fluorescent optode with mathematical modeling to investigate the deteriorating effect of anthropogenic CO₂ on North Sea chalk. We will use a pH-sensitive molecule and an optical probe to monitor mineral dissolution progress in real-time. The collected data will be processed using a novel surface renewal model that quantifies the intrinsic relation between surface morphology, surface speciation, and mineral dissolution rate. Our apparatus will allow the delineation of aforementioned factors from the intrinsic reactivity of chalk and therefore provide more easy-to-use kinetic parameters and rate-laws in analyzing the CO₂-chalk reactions under different scenarios during CO₂ storage in depleted chalk reservoirs.

6. In-situ observation of chalk dissolution during CO₂ injection (Project 5)

The main objective of this project is to observe, at reservoir conditions (pressure, temperature and fluid composition), the extent of dissolution of real chalk reservoir core during injection of CO₂. We will combine in-situ CT 3D imaging with modelling at different scales – geochemical calculations and plume migration. A new in-situ tri-axial cell has been designed to be able to CT scan a one-inch chalk core sample during injection of CO₂, with the possibility of applying mechanical pressure along the length of the core. We want to observe the dissolution of the core during injection. The CO₂ injection can be adjusted so that we simulate various scenarios: i) continuous CO₂ injection, ii) alternate between CO₂ and artificial sea water injection and iii) using CO₂ with various degree of impurities. The pressure of injected fluids will also be varied, so that various dissolution mechanisms can be eventually observed (fingering, front dissolution).

The time step in the in-situ CT scanning experiment is dependent on how many projection images are taken during one scan. With the help of in-house reconstruction software [11], the number of projections needed for a superior quality 3D image will be reduced. We plan to achieve one scan in 25-30 minutes. If the dissolution is significant, the applied mechanical pressure can trigger the collapse of the core. Our time step during injection will be too large to capture such a collapse, but we will have before and after collapse images to be able to quantify when the integrity of the core is compromised during dissolution.

We will perform before and after an experiment a detailed characterization of the surfaces of the core, using scanning electron microscope (SEM), X-ray photoelectron spectroscopy (XPS) and atomic force microscopy coupled with infrared spectroscopy (AFM-NanoIR). This will allow to link dissolution of the chalk sample with its surface composition, a key parameter for surface reactivity of chalk [12]. This project will therefore provide insight on the risk of dissolution and collapse of chalk during CO₂ injection. The use of reservoir chalk core and reservoir conditions means that experimental conditions will be as close as possible to reality. Finally, our goal is to provide a safe operation procedure in terms of injection pressure to minimize dissolution effect.

7. Discussion and conclusion

The studies included in the research program address both short- and long-term responses of chalk to CO₂ injection considering the relevant physical and chemical processes as well as longer term storage tests in the laboratory. The main question to be addressed is under which conditions CO₂ storage in chalk is feasible and sufficiently safe. In this paper we have highlighted some of the main challenges and enablers for CO₂ storage in chalk fields in terms of the storage mechanisms, injectivity, risk, containment and longevity. Secondly, we describe the progress of the established research program towards determining if chalk is suitable as a long-term storage reservoir for CO₂. The challenges with CCUS in chalk relate to the dissolution effects of chalk in the presence of water acidified by CO₂, and the extent of dissolution in different scenarios. The effects on rock chemistry, geomechanical stability, flow properties, kinetics and injectivity must be understood to de-risk a CO₂ storage program for the mature Danish North Sea oil fields which are mostly located in the cretaceous. This combined research program [2] and the Bifrost project [13-17] seek to answer these questions about the suitability of chalk and provide safe operating conditions for CO₂ storage in chalk by combining state of the art experimental approaches with advanced modelling.

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