Experimental and Numerical Investigations into CO₂ Interactions with Injection Well Infrastructure for CO₂ Storage

By

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A report submitted in partial fulfilment of the requirements for the MSc and/or the DIC.

September 2012
Declaration of Own Work

I declare that this thesis:

"Experimental and Numerical Investigations into CO₂ Interactions with Injection Well Infrastructure for CO₂ Storage"

is entirely my own work and that where any material could be construed as the work of others, it is fully cited and referenced, and/or with appropriate acknowledgement given.

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Abstract
Wellbore integrity is an essential requirement to ensure the success of a carbon capture and storage (CCS) project. Leakage of CO₂ from the well would not only severely impede the efficiency of CO₂ injection and storage but could also have dramatic impacts on the environment. Previous research has revealed that changes in bottomhole pressure and temperature conditions can lead to the formation of cracks and microannuli at cement/casing and cement/rock interfaces. It has been suggested that these interfaces may constitute preferential pathways for potential CO₂ leakage. This research investigates the sealing behaviour of a microannulus at the cement/casing interface under simulated pressure and temperature conditions and uses the findings to assess the overall integrity of CO₂ storage. Experimental investigations were conducted on a full-scale wellbore model incorporating a complete casing-cement-pseudorock assembly. A microannulus was artificially created and controlled at the cement/casing interface and subjected to continuous flow of pure CO₂. Evolution of permeability of the microannulus over time was recorded. The evolution follows an exponential decay with permeability reducing by a factor of 10 in two months as a result of carbonation reactions between CO₂ and cement. These results suggest that similar microannuli developing at the casing/cement interface and subjected to comparable flow conditions are self-healing with time. Numerical modelling work was carried out assuming a hypothetical stratified reservoir to assess the impact of such leakage pathways on the integrity of CO₂ storage. Experimental results were used as an input to implement a time-dependent microfracture permeability model in the numerical model. The influence of various parameters such as microfracture size, initial permeability and carbonation rate on the leaked volume of CO₂ was investigated. Volumes of CO₂ escaping through such leakage pathways were found to be relatively small in comparison with the total volumes of injected CO₂; this indicates the effectiveness of self-healing mechanisms. Carbonation rate was found to be the most influential parameter on total volume of CO₂ leaked.

Introduction
It is now widely accepted that recent human activities, through substantial emissions of greenhouse gases, are responsible for global warming and its associated impacts (IPCC, 2001). Emissions of greenhouse gases modify the composition of the atmosphere and alter the balance between incoming solar radiation and outgoing infrared radiation within the atmospheric system. Over sixty different gases contribute to the greenhouse effect, including carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O). Although CO₂ does not have the highest gas warming potential, it is largely dominant among human emissions, which makes it the major contributor to human-induced climate change (IPCC, 2007). Quantitative works on radiative forcing, a measure of the influence of different factors on the energy balance of the atmosphere, have confirmed this fact (IPCC, 2007). Numerous studies have pointed out that negative impacts of climate change on various sectors such as agriculture, health, economy and environment outweigh by far any positive ones (Cook, 2010). Reducing CO₂ emissions is considered as one of the most important challenges of the twenty-first century; unfortunately, the complexity of the problem is a major barrier to rapid progress and proposed solutions have given somewhat mixed results so far (UNFCCC, 2007).

In this context, carbon capture and storage (CCS) appears as a very promising solution and has been developing quickly for the past fifteen years. CCS is well adapted for targeting energy supply and industry sectors which account for 45% of total CO₂ emissions (IPCC, 2007). Three fundamental conditions need to be met by geological media to ensure the success of a CO₂ storage operation (Bachu and Bennion, 2009): accommodate the injected volume, accept the injected rate and impede any leakage of CO₂ from the storage zone. The last condition is of outmost importance since leakage of CO₂ could have detrimental impacts on the environment through contamination of shallow aquifers. A European Commission directive has recently underlined the importance of storage integrity and defined the requirements that CO₂ storage sites are required to fulfil (EC, 2009).

Wellbores can be seen as direct links between reservoirs and surface and hence provide preferential leakage pathways for stored CO₂. Figure 1 shows different leakage scenarios that could occur in a typical plugged wellbore configuration (Gasda et al., 2004). These are: a) between casing and cement, b) between cement plug and casing, c) through the pore spaces in cement, d) through casing, e) through fractures in cement and f) between cement and rock. As illustrated on the figure, the majority of these cases (c excluded) result from structural defects in the near-wellbore area. Defects could result from various mechanisms, occurring at various stages of well completion, operation and abandonment. Most of these mechanisms have been identified and described in literature. For example, a substantial mud filter cake that has not been removed prior to cementing...
could result in a weak and improper-sealing bond at the cement/formation interface (Bittleston and Guillot, 1991). The mud cake could also dehydrate and lead to the creation of an annulus at this interface (Bonett and Pafitis, 1996). Once the cement is set, changes in bottomhole conditions such as cyclic pressure and temperature variations can create sufficient stresses in casing, causing its displacement. This could eventually lead to cement debonding or failure, resulting in the formation of radial cracks and microannuli at cement/casing or cement/formation interfaces (Boukhelifa et al., 2004). Cement shrinkage may also be a cause of circumferential crack formation at these particular interfaces (Dusseault et al., 2000). High pressures encountered at reservoir depths will tend to propagate these cracks upwards and extend leakage pathways. During abandonment, inadequate plugging could potentially play an important role in well leakage although it has been suggested that most of the wells are now properly plugged (Bachu and Watson, 2006).

Although understanding these mechanisms is crucial when one wants to mitigate CO₂ leakage, understanding the further evolution of a pathway is equally important. This evolution is mainly dictated by three chemical phenomena: casing corrosion, cement carbonation and cement dissolution. Fluids encountered in CO₂ storage reservoirs are typically mixtures of CO₂ (from injection) and brine (from existing aquifer) and are indeed chemically reactive with both cement and casing.

Casing corrosion has been studied both in the laboratory (Carey et al., 2009) and using field data (Carey et al., 2006; Crow et al., 2009). Although laboratory experiments suggest significant erosion of cement by CO₂-brine mixtures, casing corrosion was found to be almost non-existent in CO₂ production wells after 30 years of exposure. Authors have suggested that the flux of CO₂ used during laboratory experiments was significantly higher than the fluxes at the field sites, thus explaining the discrepancy in results.

Interactions between cement and CO₂/brine mixtures have also been extensively investigated by researchers (Duguid and Scherer, 2010; Kutchko et al., 2007; Kutchko et al., 2008; Onan, 1984; Sauman, 1972). Two competing phenomena were identified: carbonation of cement and dissolution of cement. Carbonation is an exothermic reaction between CO₂ and aqueous hydroxide and cations to produce carbonates in liquid phase (Gervais et al., 2004). It results in alkalinisation of the pore water in the cement matrix due to the high alkalinity of carbonated cement matrix, pH of which is of the order of 12.5 (Lea, 1970). In this chain of reactions, the hydration of aqueous CO₂ to H₂CO₃ is slow and hence determines the duration of the entire process. The ionisation of H₂CO₃ to H⁺, HCO⁻ and CO₃⁻ is followed by exothermic dissolution of cementsitious phases (CaO₂)SiO₂ and (CaO)₂SiO₂. This results in the formation of unstable vaterite and aragonite, which ultimately converts into SiO₂, H₂O and amorphous CaCO₃. The fibrous morphology of these calcium silicate hydrate precipitates within the ultrafine pores of cement resulting in reduction in porosity. Simplified carbonation reactions are illustrated in equations 1.a and 1.b:

\[
\begin{align*}
CO_2 + H_2O & \rightarrow H_2CO_3 \\
Ca(OH)_2 + H_2CO_3 & \rightarrow CaCO_3 + 2H_2O
\end{align*}
\]

Researchers analysed the impact of dissolved CO₂ on the borehole cements (Robins and Milodowski, 1986) and concluded that CO₂ reacts with all hydration products of cement, including portlandite, to form alumina and silica gels in addition to the production of bicarbonate complexes. Simplified equations of the dissolution processes are illustrated in equations 2.a and 2.b:

\[
\begin{align*}
CaCO_3 + CO_2 + H_2O & \rightarrow Ca(HCO_3)_2(aq) \\
Ca_xSiO_{2+x} + xH_2O + 2xCO_2 & \rightarrow xCa(HCO_3)_2(aq) + SiO_2
\end{align*}
\]

Carbonation and dissolution have opposite geomechanical effects on cement, the former increases mechanical strength and decreases permeability, the latter decreases mechanical strength and increases permeability (Chi et al., 2002; Claissse et al., 1999). Knowledge of the dominant mechanism in a given leakage scenario is thus of primary importance as it predicts the evolution of the sealing behaviour of the pathway. Early research focused on migration of CO₂ through cement matrix (case c. of Figure 1). Both laboratory (Kutchko et al., 2007; Kutchko et al., 2008) and field studies (Carey et al., 2006; Crow et al., 2009) have concluded that this process was essentially driven by diffusion and hence slow. Cement degradation due to CO₂ is controlled by the formation of a quasi-impermeable calcite layer at the contact between the CO₂-rich brine and cement. Such a layer forms a barrier to further cement degradation. Reported research has indicated that the casing/cement interface is more likely to be the primary path for potential CO₂ leakage compared to cement matrix (Bachu and Bennion, 2009; Carey et al., 2006; Crow et al., 2009). The sealing characteristics of a conductive microannulus have first been studied from a purely mechanical point of view in a full-scale annular geometry (Boukhelifa et al., 2004). Responses were shown to be heavily dependent on the type of cement used. Research that followed addressed the complex interaction between geochemistry and geome-

![Figure 1: Leakage pathways in the wellbore area.](Gasda et al., 2004)
chanics in a conductive pathway (Huerta et al., 2008). Changes in fracture apertures were investigated under large ranges of CO₂-brine flow rates and confining pressures. It was suggested that leaky wells may be self-sealing against fluxes of CO₂-rich fluids because of carbonation. Another research team (Bachu and Bennion, 2009) tested different leakage geometries at the cement/casing interface including perfect bonding, annular gap and presence of cracks. Results concluded that good Portland cement and good bonding are reliable barriers to the upward flow of CO₂-saturated brine. However, mechanical defects such as gaps in bonding or cracks lead to flow paths with significant effective permeability. In cases where the flow through such pathways is vigorous enough to wash the results of cement carbonation, further reactions are likely to lead to cement degradation and permeability enhancement.

The relatively small amount of available information on this important topic provides a strong motivation for further investigations. The aim of research presented in this paper was to investigate the sealing behaviour of a cement/casing interface subjected to CO₂ flux under simulated downhole conditions. The findings of this experimental work were implemented in a near-wellbore numerical model to evaluate the risk and assess the impact of leakage on CO₂ storage integrity.

Experimental Work

A full scale cement/casing/pseudo rock laboratory model was used to study the behaviour of casing expansion/contraction during changes in downhole temperature and pressure and to enable the formation and control of a microannulus at the cement/casing interface. The permeability evolution of this microannulus was measured for a continuous long term flow of CO₂. The following section describes the design, construction and testing procedure of the wellbore model in the laboratory.

Description of Experimental Setup. A general view of the experimental setup, including ancillary equipment is shown in Figure 2. The wellbore consists of an assembly of the following concentric elements: casing, cement sheath and outer ring. The stainless steel outer ring is intended to simulate the stiffness offered by the reservoir rock to the displacement applied at the wellbore. Assuming an isotropic formation and plane strain conditions for the stress distribution around the injection wellbore, the relationship between the dimensions of the outer steel ring and the shear modulus of the formation it simulates is given by the following equation:

$$G_s = \frac{G(r_2^2 - r_1^2)}{(1 - 2\nu)r_1^2 + r_2^2}$$

where $G_s$ is the shear modulus of simulated formation (in GPa), $G$ is the shear modulus of steel (70 GPa), $\nu$ is the Poisson’s ratio of steel (0.3), $r_1$ and $r_2$ are respectively the inner and outer radius of the stainless steel ring (in m). The present set of experiments were conducted using an outer stainless steel ring of 109.9 mm outside diameter and thickness of 3.76 mm which simulates a weak formation of shear modulus equal to 3.8 GPa. Strain gauges on the outer edge of the stainless steel ring measure the strain transferred from the wellbore casing and cement sheath.

![Figure 2: Schematic representation of the experimental setup.](image-url)

The Central Loading Mechanism (CLM) consists of four case-hardened shoes that can impart radial load onto the wellbore casing. The radial movement of the shoes is powered through the synchronised movement of four precision jacks controlled hydraulically (Figure 3.a). The cement mix used for the experimental work is based on the industry standard API RP10B Grade G consisting of 100 parts of cement to 44 parts of water. The cement was cast whilst the casing was subjected to a loading of 4.5 MPa (Figure 3.b) imparted through the CLM. Once the cement was set, the CLM was retracted, inducing tensile stress on the cement-casing interface and creating a microannulus. The cement was cured under water for 28 days.
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Figure 3: Components of wellbore experimental setup. a) Central loading mechanism (CLM) used to impart radial loading onto the wellbore casing, b) cement slurry poured between casing and outer stainless steel ring, c) fully instrumented wellbore experimental setup.

Once the cement was set, strain gauges were pasted in Wheatstone’s half bridge configuration on the outer steel ring and enclosed in an airtight enclosure (Figure 3.c). The enclosure consists of a top and bottom stainless steel platen of 10 mm-thick steel held in place by 8 high tensile steel bolts. A pressure maintainer pump is used to control the displacement of the casing through hydraulic jacks of the loading mechanism. In order to simulate reservoir conditions, the test setup is enclosed in a laboratory oven which acts both as temperature and safety enclosure. The pore fluid is supplied through two syringe pumps (Figure 2). Pure CO$_2$ is supplied through gas bottle sourced from BOC UK with a purity of 99.5%. For permeability measurements, brine (10% salinity) is supplied through the syringe pumps.

Experimental Procedure. The experimental programme consists of the following three stages: calibration of the loading mechanism, investigation of the stress-permeability behaviour of the microannulus using a Sleipner-type brine and conduction of permeability tests with CO$_2$ to simulate CO$_2$-well infrastructure interactions in shallow formations (Sleipner type).

System Calibration. Before assembling the wellbore-casing-cement model, calibration routines were carried out to investigate both uniformity in radial loading applied by the CLM and uniformity in radial deformation of the casing steel ring. Four strain gauges were pasted in Wheatstone’s Half Bridge configuration around the circumference of the casing at a uniform angular spacing of 90° between them. The schematic of the test setup for calibration routines is shown in Figure 4.a and the photograph is displayed in Figure 4.b. The calibration cycles were carried out by keeping the well casing stationary and rotating the CLM by 90° so that each time a different shoe of the CLM (indicated as L1, L2, L3 and L4) and a strain gauge (indicated as S1, S2, S3 and S4) face each other.
For each calibration configuration, pressure was increased incrementally and strain values were recorded. Figure 5.a depicts the strains recorded during the first calibration routine for four strain gauges. It can be observed from the figure that the stress applied was distributed uniformly along the steel casing. The divergent behaviour of strain 4 is attributed to experimental errors. Figure 5.b shows the average strain recorded during relative rotation of the CLM with respect to casing. It can be implied by observing Figure 5.b that the steel casing undergoes uniform deformation and the distortion in its cylindricity is minimal. The results from the calibration routines indicate that irrespective of the position of the CLM shoes, the average strain on the steel casing is consistent, which indicates that the hydraulic jacks were well-synchronized and stress applied by the CLM was uniform.

**Stress-Permeability Behaviour.** The initial permeability of the microannulus was evaluated using a non-reactive Sleipner-type brine without any loading on the CLM; a value of 300 mD* was recorded. Experimental studies were conducted to investigate the relationship between permeability of the microannulus and stress applied on the casing through the CLM. These sets of experiments were intended to characterise the stress-dependent permeability behaviour of the microannulus, thus providing a basis to control the size of the microannulus. Furthermore, these measurements of non-reactive mass transport through the microannulus would also serve as a benchmark for comparison with the reactive transport measurements of CO₂/C₂O₂ saturated brine (discussed later). Brine injection rate was maintained constant at 1 mL/min while pressure differential across the wellbore top and bottom section was measured. Results are shown in Figure 6. Permeability was observed to decrease log-linearly with the increase in stress on the microannulus.

**Permeability Tests with CO₂.** After conducting permeability measurements with brine, experiments were carried out with CO₂ as pore fluid. Stress within CLM was kept constant and the evolution of microannulus permeability with time was investigated. To simulate leakage of pure CO₂ along the wellbore, pure and dry CO₂ was introduced at 2.5 MPa through the upstream end of the wellbore cell while maintaining the stress in CLM at 8 MPa. This stress induced an initial permeability of 5 mD (as anticipated from Figure 6). Flow rate and downstream pressure were measured at regular intervals; permeability was evaluated using Darcy’s law and is shown in Figure 7. It is observed that the permeability of the microannulus reduces with time and follows an exponential decay trend. Over a period of about two months, permeability of the microannulus was observed to be reduced by a factor of 10. The main reason for this decay is attributed to reactive transport of CO₂ resulting in the carbonation of cement and precipitation of calcite within the microannulus. These calcite depositions form a barrier to flow of

![Figure 5: Casing deformation.](image1)

a) At different gauges for a given CLM position and b) averaged at all gauges for different CLM positions.

![Figure 6: Stress-dependent permeability behaviour of microannulus.](image2)

(Test temperature: 45 °C).

*1 mD=9.869233e⁻¹⁶ m²
CO₂ and effectively reduce permeability of the microannulus. This work leads to the conclusion that permeability of leakage pathways developing at the cement/casing interface is considerably reduced with time under simulated conditions; cement carbonation seems to be dominant over dissolution in this case. It is suggested that such leakage pathways could eventually heal completely.

**Numerical Modelling**

The numerical modelling work focuses on the near-wellbore region, using the experimental results presented earlier to investigate cement/casing interface behaviour during and after CO₂ injection periods. It is also intended to develop an integrated modelling methodology to describe various physical processes (flow, thermal and mechanical loads) in the reservoir with an emphasis on near-wellbore processes. Experimental permeability results are utilised in this numerical model to ascertain the impact of CO₂ leakage.

In the development of an integrated modelling methodology, the complexity of the physics incorporated is increased systematically and tested with synthetic data. A fully coupled thermo-geomechanical model was implemented using COMSOL Multiphysics® for numerical prediction of wellbore stresses and displacements under subsurface reservoir conditions. Although, geomechanics and thermal loading can be described accurately using COMSOL, its flow module has limited capability to describe the fluid dynamics in the near-wellbore region.

To address this limitation and in view of the non-linear flow through the injection wellbore, it was decided to introduce T2Well, a specialised wellbore model developed by Lawrence Berkeley National Laboratories (LBNL), coupled with TOUGH2 to model the wellbore and near-wellbore pressure and saturation profiles. Detailed description of individual models is discussed in the subsequent sections. The modelling methodology is summarised in the workflow presented in Figure 8.

![Figure 7: Permeability evolution of microannulus subjected to reactive flow of CO₂.](image)

**Injection Modelling.** TOUGH2 is a general-purpose numerical simulation program for multi-dimensional fluid and heat flows of multiphase, multicomponent fluid mixtures in porous and fractured media (Pruess et al., 1999). The simulator supports several fluid property modules such as water, water/CO₂, water/air, water/brine/air and can be effectively applied to various sectors such as reservoir engineering, hydrology or environmental assessment. ECO2N is a fluid property module specifically
designed for applications to geological storage of CO₂ in saline aquifers and was hence used in this work. ECO2N gives a description of the thermodynamics and thermophysical properties of H₂O/CO₂/NaCl mixtures. Isothermal and non-isothermal flow processes can be modelled. The simulated fluid is represented as a three-phase mixture: a gas phase which includes water vapour and gaseous CO₂, a liquid phase containing water and dissolved CO₂ and a potential solid phase accounting for precipitated salt.

**Model Description.** As the injection scenario was not the primary focus of this thesis but is used to provide realistic data for further leakage modelling, synthetic data from a previous work (Pruess et al., 2002) was used as input in ECO2N. CO₂ is injected into a 50 m-thick saline aquifer with pre-injection conditions of 12 MPa pressure and 45°C temperature (corresponding to typical conditions at a depth of 1,000 m). The formation is entirely perforated and the injection rate was set to 30 kg/s, corresponding to an annual rate of one million tons of CO₂ injected. The simulation period extends for five years. The aquifer is considered infinite, a length of 100 km was chosen for this purpose. A logarithmic mesh was created to precisely capture flow behaviour in the near-wellbore region. The rock is homogeneous sandstone with a density of 2,600 kg/m³, a porosity of 0.12 and a permeability of 100 mD. The initial fluid present in the aquifer is brine with a salinity of 15% by weight and an initial gas saturation of zero. Relative permeability and capillary pressure are modelled using van Genuchten formulations (Genuchten, 1980; Pruess et al., 1999):

\[
\begin{align*}
    k_{rl} &= \sqrt{S_r^2}\left\{1 - \left[1 - \left(S^*\right)^{1/\lambda}\right]^{\lambda}\right\}^2 \\
    k_{rg} &= (1 - S_r)^2(1 - S^2) \\
    P_{cap} &= -P_0\left\{\left(S^*\right)^{1/\lambda} - 1\right\}^{1-\lambda} \\
    S^* &= (S_l - S_{tr})/(1 - S_{tr}) ; \quad S' = (S_l - S_{tr})/(1 - S_{tr} - S_{gr}).
\end{align*}
\]

where \(S_r\) and \(S_g\) are the irreducible liquid and gas saturations respectively and \(\lambda\) is a fitting parameter. \(P_c\) is the capillary pressure at maximum liquid saturation. Parameters for the permeability model were chosen so as to match published data (Bennion and Bachu, 2005) as closely as possible. This data (based on the Viking formation in Canada) is displayed in Figure 9 along with the curves obtained using van Genuchten model. Parameters \(\lambda = 0.55\), \(S_{tr} = 0.3\) and \(S_{gr} = 0.05\) were used. A value of 5.1e-5 Pa⁻¹ for \(1/P_o\) was chosen according to literature suggestions for analogous cases (Pruess et al., 2002). A good match was obtained. ECO2N specific options were left at default values and correspond to the following configurations: i) permeability does not depend on reduction of pore space due to precipitation of salt, ii) the model of water solubility in CO₂ is taken from published literature (Spycher and Pruess, 2005), iii) brine density varies with dissolved CO₂ concentration according to (Garcia, 2001) correlation for temperature dependence of molar volume of CO₂ iv) thermophysical properties are fully dependent on salinity and v) the correlation for brine enthalpy at saturated vapour pressure is after (Lorenz et al., 2000). Parameters pertaining to solver configurations such as time-stepping or convergence criteria were not investigated and left at default.

![Figure 9: Relative permeability curves used in simulation studies.](image-url)
Results and Analysis. Results from TOUGH2/ECO2N are shown in Figures 10 to 13. Figure 10 presents the evolution of pressure with time at different distances from the well i.e. 1 m, 10 m, 100 m and 1,000 m. Pressure evolutions all follow the same trend at these points, starting from the initial value of 12 MPa and increasing to reach a plateau at around 15-16 MPa. As expected, points situated closer to the well will undergo the pressure increase faster than those further away. The evolution of gas saturation at these points is presented in Figure 11. The same kind of trend is observed. When CO₂ starts migrating through the reservoir, it tends to dissolve in the formation brine. Once brine is saturated, CO₂ is in gaseous form and gas saturation increases. Starting from the initial value of zero, it eventually reaches a plateau value of approximately 0.66. The irreducible water saturation being equal to 0.3, this value of 0.66 indicates that approximately 4% of the pore space is occupied by solid salt. Since the reservoir is cylindrical and the injection rate is constant, time to reach a given point is proportional to the square of its distance to the well. Figure 12 shows the spatial distribution of pressure after 5 years of injection. The represented area extends up to 100 m from the well. The near-wellbore region is of interest for evaluating the effects of leakage through the microannulus. Two pressure gradients can be seen in this figure. The first one is a pressure increase as the distance from the well decreases; this corresponds to the effects of injection. The second one is a pressure increase as depth increases and corresponds to the effects of gravity. Figure 13 represents the spatial distribution of gas saturation. Again, both injection and gravitational effects have an impact on the distribution. Since injection of the gas is at the wellbore, gas saturation is higher in the vicinity of the well. The density variation between gaseous and dissolved phase would result in buoyancy driven flow and gas saturation decreases with increasing depth. Pressure and saturation distributions obtained after 5 years of injection were used as initial values for CO₂ leakage modelling.

CO₂ Leakage Modelling. COMSOL Multiphysics is an engineering, design, and finite element analysis software environment for the modelling and simulation of physical phenomena (COMSOL website). Several modules such as AC/DC, heat transfer, pipe flow or structural mechanics are readily available. These modules are based on pre-defined equations which solve for the most common problem types. Different modules can be linked together to study coupled problems incorporating several physics-based models. COMSOL Multiphysics also allows the user to enter its own partial differential equations for more specific problems.

The vast majority of the work was conducted using the two-phase Darcy’s law subsurface flow module. A 2D-axisymmetric space dimension was chosen, the axis of symmetry being the centre of the wellbore. Flow modelling was performed using time-dependent analysis.
Figure 12: Pressure distribution after 5 years of injection. Pressure ranges from 15.9 MPa (dark blue) to 16.9 MPa (red).

Figure 13: Gas saturation distribution after 5 years of injection. Gas saturation ranges from 0 (dark blue) to 0.668 (red).
**Model Description.** The aim of this model is to investigate the influence of microannuli parameters, such as size and permeability on the volume of CO$_2$ that leaks out from the storage reservoir. The scenario of carbon dioxide leakage from a storage reservoir into a shallower aquifer is of particular interest because of potentially devastating environmental impacts; this scenario was hence modelled here. The geometry of the model consists of two sandstone formations interbedded by an impermeable shale layer (Figure 14). The lower sandstone formation is the one that has been used to store CO$_2$, its properties are exactly the same as those described in the injection modelling section (homogeneous, $\phi=0.12$, $k=100$ mD). The thickness of the formation is 50 m. The upper sandstone represents another aquifer that has not been used for CO$_2$ storage; it possesses the same rock properties and geometry as the lower one. The shale layer that separates both sandstone formations has a thickness of 10 m and is considered impermeable to flow. The wellbore was modelled as a cylinder of 73 mm in radius; casing and cement are annuli of thickness 3 mm and 22 mm respectively. Note that these figures correspond to a typical wellbore+casing diameter of 6” (15.2 cm). Diffusion through cement was modelled as a very low-permeability flow. A porosity of 0.1 and a permeability of 1 μD were assigned to cement, following data published in literature (Bachu and Bennion, 2009; Mellas et al., 2003). Casing was considered completely impermeable to flow. The microannulus develops along the casing/cement interface and extends from the lower formation upwards. It is expected that CO$_2$ will enter into the microannulus through perforations, propagate upwards and leak into the top formation. The size of the microannulus is not a fixed parameter but rather a variable whose effect was investigated in the current work. The lower sandstone formation is perforated along its entire thickness (as mentioned in the injection scenario). It is assumed that the wellbore is plugged in this zone. Relative permeability data from the Viking formation (Bennion and Bachu, 2005) was used once again in the model. Initial pressure and gas saturation distributions correspond to those obtained from injection modelling after 5 years of injection. Pressure and saturation values have been imported from TOUGH/ECO2N and set as initial conditions for the lower formation leakage modelling in COMSOL Multiphysics. The top formation was assumed not to have been affected by injected CO$_2$, its initial pressure and gas saturations are respectively set to 12 MPa and 0. A no-flux boundary condition has been imposed on all boundaries but the far-field one, situated at 100 m from the well. It is assumed that pressure and gas saturation will not be affected by leakage at such a distance and have thus been set as constant and equal to initial values at this boundary.

To achieve the modelling objectives, three parameters representing size and permeability have been chosen for the investigations: $L^*$, $k_0$ and $c$. The first one is a normalized microannulus size. The microannulus interacts with the rest of the reservoir through its area of contact with permeable media. In two dimensions, it corresponds to the perimeter of the fracture that is not in contact with the casing. This “effective” perimeter is normalised by the thickness of the caprock which was chosen arbitrarily in the modelling scenario. The normalised perimeter of the microannulus is denoted by $L^*$. The experimental perme-
ability curve was used as a starting point to implement a time-dependent fracture permeability in the model. The curve can be approximated by an exponential decay of the following form:

\[ k = k_0 e^{-ct} \]  

(5)

where \( k_0 \) is the initial permeability of the fracture at \( t = 0 \) and \( c \) represents carbonation rate. COMSOL Multiphysics does not allow the user to enter a continuously changing time-dependent property. For this reason, permeability is approximated by a step curve using steps of three days. Since experiments were run for 45 days, values are only available for that period of time. To evaluate the long-term impact of leakage on the reservoir, the last permeability value is extended until reaching a simulation time of one year. This approach overestimates leakage volumes of CO\(_2\) by overestimating permeability of the fracture and is intended to serve as a “worst case scenario”. An example of a permeability curve used during simulation along with the associated experimental curve is shown in Figure 15.

The influence of \( L^* \), \( k_0 \) and \( c \) on the leaking volume of CO\(_2\) was investigated by running several simulations. Base case values were chosen as follows: \( L^* = 2 \), \( k_0 = 300 \text{ mD} \) and \( c = 0.04 \). The value of \( k_0 \) corresponds to the initial permeability of the microannulus measured during experiments when no wellbore loading was applied. The value of \( c \) comes from the trendline parameters of the experimental curve (Figure 7 or 15). The volume of CO\(_2\) that leaks out into the shallower aquifer is denoted by \( V_l \).

Although pressure drops in the storage reservoir as a result of leakage are of interest as those can be directly observed from downhole measurements at a real CO\(_2\) storage site, the simulated pressure differential values were found to be too small to be conclusive and a clear relationship between pressure drop and microannulus parameters could not be established. Therefore, these results are not presented here.

Results and Analysis. Results from microfracture modelling in COMSOL Multiphysics for the first 45 days of leakage are shown in Figures 16-19. Figure 16 presents final gas saturation distributions in top and lower formations using base case microannulus parameters. A closer view of the lower wellbore area of top formation makes the leakage clearly visible: a volume of CO\(_2\) is seen to have migrated from the storage reservoir into the shallower aquifer. The volume of CO\(_2\) is uniformly distributed as expected: the rock is homogeneous and CO\(_2\) therefore propagates uniformly through it. The path followed by leaking CO\(_2\) flow corresponds to what is anticipated: CO\(_2\) follows the shortest possible leakage pathway; it first enters into the microannulus through perforations and then migrates upwards, bypasses the caprock and enters into the shallower formation at the lowermost point by diffusing through cement. The gas saturation distribution in the lower formation is also changing to accommodate new flow conditions (termination of injection and presence of a leakage pathway).

Figure 17 shows the evolution of \( V_l \) with time for different values of \( L^* \). Parameters \( k_0 = 300 \text{ mD} \) and \( c = 0.04 \) were fixed, permeabilities at any given time were therefore the same for all values of \( L^* \). The general trend is the same in all three
cases: a first rapid increase followed by a gradual flattening as simulation time increases. The permeability of the microannulus is a time-dependent parameter that decreases with time in accordance with observed experimental results. According to Darcy’s law, permeability has a direct influence on the rate of leakage through the microannulus; rate being the derivative of volume with respect to time. A decrease in permeability makes the slopes of the curves flatten with time. It can be also noted that, at any given time, $V_l$ increases with increasing $L^*$. A higher $L^*$ implies a higher effective perimeter (as defined previously). In three dimensions, it corresponds to a higher contact area of the microannulus and hence results in a higher leakage rate as predicted by Darcy’s law. The slopes of the curves are different at early times but very similar after 45 days, which indicates that the leakage rate is almost the same in all three cases at this time. This suggests that the dimensions of the microannulus do not have significant influence on the leakage rate when permeability reduces.

The influence of the initial permeability $k_0$ on the evolution of $V_l$ with time is presented in Figure 18 (parameters $L^*=2$ and $c=0.04$ are fixed). This influence is clearly illustrated by the initial slopes of the curves, a higher permeability leading to a higher leakage rate. Since parameter $c$ is fixed, permeability ranking is maintained throughout the simulation and slopes are influenced accordingly: at any given point, the leakage rate (and therefore the slope) corresponding to a higher $k_0$ will be higher. The increase in $V_l$ as a function of initial permeability (illustrated by the spacing between curves) appears to be almost linear, especially at later times.

Figure 19 represents the influence of $c$ on the evolution of $V_l$ with time (parameters $L^*=2.5$ and $k_0=300$ mD are fixed). A higher $c$ implies a faster decrease of permeability with time and therefore a lower $V_l$. The initial slopes are equal since a value of 300 mD was used in all three cases during the first time step. A higher $c$ also implies a faster decrease in leakage rate; therefore, slopes flatten out more rapidly for high values of $c$. In other words, accelerated carbonation rate would result in reduction in leakage sooner in comparison with moderate or low carbonation rates. From a chemical kinetics perspective, a high reaction rate results from either a high molar concentration of the reactants or favourable pressure and temperature conditions. These results suggest that deeper formations or thick-cemented reservoirs (or a combination of these) could result in higher carbonation rates.

As mentioned before, simulations were extended up to one year to assess the long-term impact of different leakage scenarios on $V_l$. For each case, constant values of permeability equal to those at 45 days were used for the duration of the simulation. Results are represented in Table 1, the value of $V_l$ after one year is shown for different values of $L^*$, $k_0$ and $c$.

<table>
<thead>
<tr>
<th>$L^*$</th>
<th>$V_l$ after one year of leakage [m$^3$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>c=0.04</td>
<td>k$_0$=100 mD</td>
</tr>
<tr>
<td></td>
<td>k$_0$=300 mD</td>
</tr>
<tr>
<td></td>
<td>k$_0$=500 mD</td>
</tr>
</tbody>
</table>

The effects of increasing $L^*$, increasing $k_0$ and decreasing $c$ on increasing leakage volume $V_l$ of CO$_2$ are once again illustrated here. Parameter $c$ seems to have the strongest influence on $V_l$ after one year; a 50% change from the base case induces a difference of 93 m$^3$ in $V_l$ compared to 13 m$^3$ and less than 26 m$^3$ for $L^*$ and $k_0$, respectively.

**Discussion**

It is now strongly suggested that human beings are responsible for global warming by emitting substantial amounts of CO$_2$ and other greenhouse gases. Different measures have been proposed to reduce these emissions: fostering the use of natural gas, switching power generation from fossil fuels to nuclear fission or making use of renewable forms of energy. In the short-term, carbon capture and storage can be seen as a relatively quick and viable solution for reducing CO$_2$ emissions and mitigating environmental impacts. The main challenge of CCS consists in storing CO$_2$ in a safe manner over geological periods of time. If this condition is not satisfied, CO$_2$ could eventually find its way back into the atmosphere, producing an effect opposite to that intended: the entire concept of CCS would then be discredited. A European Commission directive has strongly underlined the need to: minimise CO$_2$ leakage as much as possible, quantify any potential leakage rates and volumes and make use of available remediation techniques. In that context, leakage pathways in CO$_2$ storage reservoirs constitute a key issue and need to be investigated with greater attention. Wellbores provide natural connections between underground reservoirs and surface: these have been pointed out as offering preferential pathways for leaking CO$_2$. Literature suggests that, in a typical abandoned well that has been plugged, microannuli developing at the cement/casing interface provide a primary path for potential CO$_2$ leakage compared to the cement matrix. A poor cementing job or changes in bottomhole conditions, such as cyclic pressure and temperature variations for example, can lead to the formation of such microannuli. Flow of CO$_2$ at the casing/cement interface is reactive and involves two chemical phenomena: carbonation and dissolution of cement. The dominance of one of these phenomena over the other dictates the evolution of the flow conductivity of the pathway: carbonation tends to "heal" the fracture whereas...
Figure 16: Gas saturation distributions after 45 days of leakage. Upper and lower formations are represented on the left. The left boundary corresponds to the well. A closer view of the lower wellbore area of the upper formation shows presence of CO₂. Gas saturation ranges from 0 (dark blue) to 0.667 (red).

Figure 17: Influence of normalised microfracture size on leaked volume of CO₂.
Experimental and Numerical Investigations into CO$_2$ Interactions with Injection Well Infrastructure for CO$_2$ Storage

Figure 18: Influence of initial microfracture permeability on leaked volume of CO$_2$.

Figure 19: Influence of carbonation rate on leaked volume of CO$_2$. 
dissolution aggravates it. The evolution of these pathways is highly debated in literature and therefore provides motivation for further investigations.

Experiments have been conducted on a full-scale wellbore model, a microannulus was artificially created at the casing/cement interface and subjected to reactive flow of CO₂. The evolution of permeability of the microannulus was investigated. Experiments have shown that permeability clearly reduces with time, supporting the idea that microannuli are self-healing under certain subsurface conditions. These first findings are positive from an environmental perspective as they minimise possible impacts of CO₂ leakage. However, further research is needed to test the model under different conditions.

A numerical model was built to quantify the volumes of CO₂ escaping through a typical microannulus at the casing/cement interface. Quantifying these volumes is useful both to address the European Commission directive and to measure the extent of possible environmental impact. Knowing the volume of CO₂ that leaks into a groundwater reservoir, for example, is essential to assess water toxicity and possible consequences on the environment. The impacts of microfracture parameters such as size and permeability were investigated in a numerical model. Volumes escaping from the reservoir were found to be relatively small in general, under assumed configuration. The numerical model is flexible and can serve in future research with different input parameters. Experimental results can also be used for implementation into various time-dependent microfracture permeability flow models or can serve as a basis for further experimental investigations.

Conclusions and Suggestions for Further Work

1. Experimental work strongly suggests that microannuli subjected to flow of CO₂ at the cement/casing interface are self-healing with time under certain physical and geometrical conditions.
2. The permeability evolution of such microannuli follows an exponential decay with time.
3. Carbonation dominates over dissolution under tested flow rate and pressure conditions.
4. Numerical models show that volumes escaping through such leakage pathways are relatively small compared to total volumes of stored CO₂.
5. The parameter which has the greatest impact on leakage volume was found to be carbonation rate, followed by initial permeability and size of the microannulus.
6. Future experimental work will need to investigate microfracture permeability behaviour at different temperatures and CO₂ pressures. Numerical work should focus on building a coupled flow/structural mechanics model. The influence of stress on size and permeability of the microannuli could then be studied.

Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>2D</td>
<td>Two-dimensional</td>
<td></td>
</tr>
<tr>
<td>AC/DC</td>
<td>Alternating/Direct current</td>
<td></td>
</tr>
<tr>
<td>c</td>
<td>Carbonation rate</td>
<td></td>
</tr>
<tr>
<td>Ca(HCO₃)₂</td>
<td>Calcium bicarbonate</td>
<td></td>
</tr>
<tr>
<td>Ca(OH)₂</td>
<td>Calcium hydroxide</td>
<td></td>
</tr>
<tr>
<td>CaCO₃</td>
<td>Calcite</td>
<td></td>
</tr>
<tr>
<td>Ca₅SiO₂₋ₓ</td>
<td>Calcium silicate hydrate</td>
<td></td>
</tr>
<tr>
<td>CCS</td>
<td>Carbon capture and storage</td>
<td></td>
</tr>
<tr>
<td>CH₄</td>
<td>Methane</td>
<td></td>
</tr>
<tr>
<td>CLM</td>
<td>Central loading mechanism</td>
<td></td>
</tr>
<tr>
<td>CO₂</td>
<td>Carbon dioxide</td>
<td></td>
</tr>
<tr>
<td>CO₃⁻</td>
<td>Carbonate</td>
<td></td>
</tr>
<tr>
<td>G</td>
<td>Shear modulus [GPa]</td>
<td></td>
</tr>
<tr>
<td>Gₜ</td>
<td>Effective stiffness [GPa]</td>
<td></td>
</tr>
<tr>
<td>H⁺</td>
<td>Hydron</td>
<td></td>
</tr>
<tr>
<td>H₂CO₃</td>
<td>Carbonic acid</td>
<td></td>
</tr>
<tr>
<td>H₂O</td>
<td>Water</td>
<td></td>
</tr>
<tr>
<td>HCO⁻³</td>
<td>Hydrogen carbonate</td>
<td></td>
</tr>
<tr>
<td>k</td>
<td>Permeability [mD]</td>
<td></td>
</tr>
<tr>
<td>k₀</td>
<td>Initial microfracture permeability [mD]</td>
<td></td>
</tr>
<tr>
<td>kₑ</td>
<td>Relative permeability of gas phase</td>
<td></td>
</tr>
<tr>
<td>kᵥ</td>
<td>Relative permeability of liquid phase</td>
<td></td>
</tr>
<tr>
<td>L</td>
<td>Normalized microfracture size</td>
<td></td>
</tr>
<tr>
<td>LBNL</td>
<td>Lawrence Berkeley National Laboratory</td>
<td></td>
</tr>
<tr>
<td>N₂O</td>
<td>Nitrous oxide</td>
<td></td>
</tr>
<tr>
<td>NaCl</td>
<td>Sodium chloride</td>
<td></td>
</tr>
<tr>
<td>Pₖcap</td>
<td>Capillary pressure [Pa]</td>
<td></td>
</tr>
<tr>
<td>pH</td>
<td>Potential hydrogen</td>
<td></td>
</tr>
<tr>
<td>P₀</td>
<td>Capillary pressure at maximum liquid saturation [Pa]</td>
<td></td>
</tr>
<tr>
<td>r₁</td>
<td>Inner radius [m]</td>
<td></td>
</tr>
<tr>
<td>r₂</td>
<td>Outer radius [m]</td>
<td></td>
</tr>
<tr>
<td>Sₙ</td>
<td>Saturation of gas phase</td>
<td></td>
</tr>
<tr>
<td>Sᵣᵣ</td>
<td>Residual saturation of gas phase</td>
<td></td>
</tr>
<tr>
<td>SiO₂</td>
<td>Silica</td>
<td></td>
</tr>
<tr>
<td>Sᵣ</td>
<td>Saturation of liquid phase</td>
<td></td>
</tr>
<tr>
<td>Sᵣᵣ</td>
<td>Residual saturation of liquid phase</td>
<td></td>
</tr>
<tr>
<td>V₁</td>
<td>Volume of CO₂ leaking into the shallower aquifer</td>
<td></td>
</tr>
<tr>
<td>λ</td>
<td>van Genuchten parameter</td>
<td></td>
</tr>
<tr>
<td>ν</td>
<td>Poisson’s ratio</td>
<td></td>
</tr>
<tr>
<td>ϕ</td>
<td>Porosity</td>
<td></td>
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</table>
References
Onan, D. D., 1984, Effects of Supercritical Carbon Dioxide on Well Cements: SPE, no. 12593.
UNFCCC, 2007, Compilation and synthesis of fourth national communications.
### Appendix A: Literature Review

<table>
<thead>
<tr>
<th>Paper n°</th>
<th>Year</th>
<th>Title</th>
<th>Authors</th>
<th>Contribution</th>
</tr>
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<tbody>
<tr>
<td>SPE 87195</td>
<td>2004</td>
<td>&quot;Evaluation of Cement Systems for Oil and Gas-Well Zonal Isolation in a Full Scale Annular Geometry&quot;</td>
<td>Boukhelifa, L., Moroni, N., James, S.G., Le Roy-Delage, S., Thiercelin, M.J., Lemaire, G.</td>
<td>First paper to describe a full-scale laboratory experiment with annular geometry. Pioneering experimental methods that will be used extensively afterwards by other research groups. First paper to investigate rigorously the sealing characteristics of a conductive micro-annulus and compare it with a numerical model.</td>
</tr>
<tr>
<td>SPE 113375</td>
<td>2008</td>
<td>&quot;Cement Core Experiments with a Conductive Leakage Pathway, Under Confining Stress and Alteration of Cement's Mechanical Properties via a Reactive Fluid, as an Analog for CO₂ Leakage Scenario&quot;</td>
<td>Huerta, N. J., Bryant, S.L., Conrad, L.</td>
<td>First paper to explicitly address the interaction between geochemistry and geomechanics in a conductive pathway.</td>
</tr>
<tr>
<td>International Journal of Greenhouse Gas Control I</td>
<td>2009</td>
<td>&quot;Experimental assessment of brine and/or CO₂ leakage through well cements at reservoir conditions&quot;</td>
<td>Bachu, S., Bennion, D.B.</td>
<td>First paper to study and compare the behaviour of various conductive pathways.</td>
</tr>
</tbody>
</table>
SPE 903 (1965)
Control and Prevention of Inter-Zonal Flow
Authors: Bearden, W.G., Spurlock, J.W., Howard, G.C.
Contribution: First paper to address the problem of leakage through casing-cement interface.
Objective: To investigate factors affecting communication at the casing-cement bond and methods which aid in preventing it.
Methodology: Used different experimental setups to investigate the effects of perforation, curing pressure and deformable seals on inter-zonal flow. Performed field tests to evaluate the efficiency of proposed seal rings.
Conclusions:
A pressure differential of approximately 1000 psi is sufficient to cause communication past a well-aged, neat Portland cement sheath. This communication occurs at the casing-cement bond. Perforation does not seriously affect the pressure required to leak past the casing. Under ideal cementing conditions, communication at the casing-cement bond can be prevented up to pressures sufficient to cause the cement to fail or the casing to collapse, by attaching to the casing a seal ring of deformable rubber backed up on one side with a fixed flange or casing collar and on the other side with a removable flange.
Comments: Relatively basic yet pioneering investigations.
SPE 64733 (2000)
Why Oilwells Leak: Cement Behavior and Long-Term Consequences
Authors: Dusseault, M.B., Gray, M.N., Nawrocki, P.A.
Contribution: Not much, as it uses some results from previous literature. Some interesting experiments on cement shrinkage.
Objective: To clarify the mechanisms involved in old leaking wells.
Methodology: Analysed the mechanisms of cement shrinkage as well as the behaviours of cement strength, cement rigidity and bond at reservoir conditions. Results are inspired by literature review as well as lab experiments.
Conclusions:
Various mechanisms, but mainly cement shrinkage, lead to a drop in radial stress and can cause formation of circumferential fractures.
Differences between lateral stress gradients and pressure gradients provide forces for vertical growth.
The fracture will tend to become gas filled as gas slowly diffuses into it, increasing the driving force.
Eventually the fracture will rise, and gas will enter shallow strata or leak at the surface.
Comments: This paper focuses mainly on the causes of well leakage, not on the leakage itself.
SPE 87195 (2004)
Evaluation of Cement Systems for Oil and Gas-Well Zonal Isolation in a Full Scale Annular Geometry
Authors: Boukhelifa et al.
Contribution: First paper to describe a full-scale laboratory experiment with annular geometry. Pioneering experimental methods that will be used extensively afterwards by other research groups. First paper to investigate rigorously the sealing characteristics of a conductive micro-annulus and compare it with a numerical model.
Objective: To evaluate mechanical responses of cement under wellbore conditions. To compare experimental results with numerical predictions
Methodology: Performed large scale laboratory tests using an annular geometry. The apparatus consists of: a central core that can be expanded and retracted to mimic the expansions and contractions of the inner casing string of a well, outer metal rings that simulate formation stiffness. Tested seven different cements (classified as neat, expanding or flexible) to determine the effects of cement mechanical properties, loading parameters and boundary conditions on cement cracking and permeability to air. Compared results with an analytical stress-analysis model.
Conclusions:
Cement expansion depends on boundary conditions.
Expanding systems generate initial compressive stress in the cement sheath and reduce the risk of tensile failure.
Expanding and flexible systems behave best in loading stages and are the only ones able to close a micro-annulus by releasing energy stored during expansion.
Predictions by the numerical model are in close agreement with experiments in large-scale annular geometry.
The lifetime of a well may be greatly extended by selection of the most appropriate cement.
Comments: Very good paper that sets a basis for further work on this topic.
SPE 90450 (2004)
Characterizing Casing-Cement-Formation Interactions under Stress Conditions: Impact on Long-Term Isolation

Authors: Mueller et al.

Contribution: Not much as it relied mainly on previous literature.

Objective: To describe the cement evaluation and wellbore-modelling methodologies specifically developed to predict the magnitude of tensile or compressive forces created by changing wellbore or reservoir conditions.

Methodology: Described wellbore stress modelling as well as Young's modulus and tensile strength determination using methods from published literature. Presented a case example to illustrate the methods.

Conclusion:
Stress modelling of a cemented wellbore can be used to predict the magnitude and location of the maximum stress imposed on a cemented annulus.
Wellbore stress modelling can be used as a design tool to determine the tensile and compressive strength requirement of the cement sheath.

Comments: This paper does not present any new material but offers a relatively good summary of existing modelling methods.
Analysis and performance of oil well cement with 30 years of CO₂ exposure from the SACROC Unit, West Texas, USA

**Authors:** Carey et al.

**Contribution:** First paper to present an *in-situ* experimental approach by studying data from the field.

**Objective:** To evaluate the integrity of Portland-cement based wellbore systems in CO₂-sequestrations environments

**Methodology:** Investigated a core sample including casing, cement and shale caprock obtained from a 30-year old CO₂-flooding operation. Analysed core sample mineralogy and chemistry using X-ray diffraction, X-ray fluorescence and scanning electron microscopy as well as permeability and porosity. Conducted computer modelling of cement degradation using methods published in literature.

**Conclusions:**
- Portland-cement retained its capacity to prevent significant transport of fluid through the cement matrix.
- Observations show evidence that CO₂ migrated along the casing–cement and shale–cement interfaces for some period of time.
- The integrity of these interfaces appears to be the most critical issue in wellbore performance for CO₂ sequestration.
- The key variables appear to be the initial width and connectivity of the interfaces in addition to the pressure gradient driving flow from the reservoir.
- Numerical calculations are consistent with these results.

**Comments:** A good paper that studies the problem using a completely different source.
SPE 113375 (2008)
Cement Core Experiments with a Conductive Leakage Pathway, Under Confining Stress and Alteration of Cement's Mechanical Properties via a Reactive Fluid, as an Analog for CO₂ Leakage Scenario

Authors: Huerta, N. J., Bryant, S.L., Conrad, L.

Contribution: First paper to explicitly address the interaction between geochemistry and geomechanics in a conductive pathway.

Objective: To investigate the interaction between fluid flow in a conductive pathway, the effect of confining stress on the magnitude of the pathway aperture, the increase in pressure drop as fluid flows through the pathway and the behaviour of pathway cement subject to variable confining stress and fluid pressure. To test the hypothesis that the altered cement will behave plastically and deform under stress so as to plug the leakage path.

Methodology: Performed preliminary theoretical calculations to design apparatus. The apparatus consists of a positive displacement pump, a Hassler cell to simulate confining stress, a pressure transducer and a data acquisition module. Created artificial fractures in cores using various methods. Performed acidic brine flow tests over large ranges of flow rates and confining pressures. Investigated changes in fracture apertures.

Conclusion:
Change in aperture directly depends on change in confining stress
When a fracture core is reacted extensively with low pH brine, substantial plastic deformation occurs as the core is brought to the initial value of confining stress.
It is possible that "leaky-wells" will be self-sealing against fluxes of CO₂-rich fluids.

Comments: Although the objectives of the paper can be confusing at first sight, these will prove to be at the very heart of the problem and lead to some interesting results.
Experimental assessment of brine and/or CO₂ leakage through well cements at reservoir conditions
Authors: Bachu, S., Bennion, D.B.
Contribution: First paper to study and compare the behaviour of various conductive pathways.
Objective: To better understand the potential processes involved in well leakage in the presence of CO₂.
Methodology: Carried out two sets of experiments: in the first set, the permeability of cement to CO₂-saturated brine was measured for three different cases of brine salinity. In the second set, the permeability of the cement-casing pair was measured for the cases of perfect bond between the two, an annular gap between the two and an annular gap plus cracks in the cement. Ethane, rather than CO₂, was used in this set of experiments in order to avoid geochemical effects. The apparatus consists of a cement sample, a confining sleeve that simulates the overburden stress present in the reservoir and a steel coreholder that simulates reservoir pressure. Pumps, pressure transducers and flow meters are also used.
Conclusion:
Good Portland cement and good bonding with casing and the surrounding rock will likely constitute a good and reliable barrier to the upward flow of CO₂ and/or CO₂-saturated brine.
The presence of mechanical defects such as gaps in bonding or cracks leads to flow paths with significant effective permeability.
The external and internal interfaces of cements (between cement and casing and rock, and along cracks) constitute the main flow pathways for fluids leakage along wells, including CO₂ and CO₂-saturated brine. In cases where the flow of CO₂-saturated brine through such mechanically produced pathways is vigorous enough to wash away the results of cement carbonation, geochemical reactions between the acidic brine and cement will likely lead to further cement degradation and permeability enhancement.
In the case of CO₂ storage, it is essential to ensure zonal isolation through good cementing and bonding, particularly through the caprock immediately above the storage unit.
Comments: This paper presents a very clear and rigorous experimental procedure.
A solution against well cement degradation under CO₂ geological storage environment

Authors: Barlet-Gouédard et al.

Contribution: Presents a new remediation technique for cement degradation.

Objective: To compare a new cement with better CO₂ resistance with conventional cement. To use experimental procedure and methodology simulating the interaction of set cement with injected, supercritical CO₂ under downhole conditions

Methodology: Performed geomechanical modelling and geochemical experiments to compare behaviours of Portland cement and new cement (SCRC). Used a mix of literature and personal results. Evaluated the risk of micro-annulus generation and cement degradation for both cements.

Conclusion:
Portland cement does not fulfil the requirements for well integrity, since micro-annulus generation is predicted by the model. Only the addition of an expanding agent to the SCRC ensures cement integrity and avoids the generation of a micro-annulus. In an environment with CO₂ in pure water, Portland cement shows strong degradation and is therefore not a good candidate for cementing new wells for CO₂ storage. The new cement (SCRC) exhibits a more reliable behaviour under exposure to wet supercritical CO₂ and CO₂ dissolved in pure water. It may therefore be suitable for cementing new CO₂ wells.

Comments: This paper does not address the main issue specifically but presents an interesting remediation technique. The problem is that Portland cement has been used in most of the old wells suitable for CO₂ storage.
Energy Procedia I (2009) 71-78

Wellbore Flow Model for Carbon Dioxide and Brine

Authors: Pan et al.

Contribution: First paper to describe a wellbore flow simulator specifically designed for CO₂-brine mixtures.

Objective: To describe a wellbore flow simulator developed to model non-isothermal open-well flow dynamics of CO₂-brine mixtures and quantify the impacts of leakage of CO₂ and brine through wellbores.

Methodology: Solved mass and thermal energy balance equations numerically by a finite difference scheme with wellbore heat transmission handled semi-analytically. Demonstrated the capabilities of the wellbore flow model using an example problem.

Conclusion:

A well-bore flow simulator that models two-phase CO₂-brine mixtures for use in GCS leakage studies has been successfully developed.

This simulation capability is intended to be used for quantifying potential leakage up wells using pressures and CO₂ saturations at depth calculated by reservoir simulation.

Although the test problem is based on flow up an open borehole, the approach can be used for flow in an annulus region by suitable modifications of roughness coefficients and geometric parameters.

Comments: An interesting paper, one of the few that presents an analytical approach to the problem.
Wellbore integrity and CO2-brine flow along the casing-cement micro-annulus
Authors: Carey et al.
Contribution: First paper to incorporate the effect of steel corrosion in the experiments.
Objective: To investigate the casing-cement micro-annulus through supercritical CO2 + brine core-flood experiments.
Conclusion:
A high flux of CO2-brine at the casing-cement interface produced significant steel corrosion and deposition of FeCO3 during a two week experiment under in situ sequestration conditions.
The cement reactions were limited to slow diffusion of CO2 into the matrix without significant erosion of cement by the CO2-brine mixture.
Despite the high flux, some of the grooved channels in the casing were partially filled with FeCO3 corrosion products, suggesting that in this case flow could eventually be limited by a self-sealing carbonate deposition.
The absence of such corrosion at the field sites suggests that the flux of CO2 producing carbonated wellbore cement at the field sites is relatively small or perhaps relatively dry.
Comments: A good paper that presents a clear experimental procedure and some novel findings.
Energy Procedia I (2009) 3561-3569
Wellbore integrity analysis of a natural CO\textsubscript{2} producer

Authors: Crow et al.

Contribution: First paper to analyse field data from a CO\textsubscript{2} producer.

Objective: To investigate the wellbore integrity of a 30-year old well from a natural CO\textsubscript{2} production reservoir

Methodology: Cut sidewall cores through the casing to recover casing, cement and formation samples. Collected and analysed fluid samples from outside the casing. Analysed cement samples for permeability, porosity, capillary pressure properties, formation factor and Young’s Modulus. Determined cement core mineralogy by X-ray diffraction. Run an Ultrasonic Imaging Tool (USIT) to provide an indication of the cement quality and its bond to the casing. Conducted a Vertical Interference Test (VIT) to assess the extent of hydraulic communication along the exterior of the well casing. Developed a numerical model and compared simulated results with experimental findings.

Conclusion:
The barrier system in this well provides hydraulic isolation across the caprock.
Carbonation of cement has been observed in varying degrees in cores from this well. Carbonation of cement increased permeability, porosity and had a mixed effect on capillary properties. It decreased the compressive strength but the cement still provides an effective barrier. These changes, while not improvements in cement properties, do not result in a significant loss of cement resistance to CO\textsubscript{2} migration.
The casing is in very good condition, consistent with good cement coverage and limited circulation of reservoir fluids along the casing-cement interface.
Cement interfaces with casing and formation appear to be tight and do not have significant calcium carbonate deposition
The VIT data analysis indicates the permeability of the barrier system including the cement interfaces to be 1-10 millidarcies, which is approximately 3 to 4 orders of magnitude greater than the cement core permeability of 1 microdarcy that was taken from the same interval.
The cement interfaces with casing and/or formation are the primary path for potential CO\textsubscript{2} migration compared to the cement matrix; however, these interfaces appear to provide sufficient flow restriction between formations in this well.

Comments: Good paper that investigates a very reliable source of information: data from the field.
The influence of confining stress and chemical alteration on conductive pathways within wellbore cement

Authors: Huerta et al.

Contribution: Not much, as it is quite similar to a previous paper by same authors.

Objective: To determine if cement with a conductive pathway could form a self-sealing system when subject to the coupled effect of confining stress and alteration of the mechanical properties along fracture surfaces by reaction with an acidic fluid. To compare bench-scale behaviour with the expected field conditions and use these insights to improve our understanding of well leakage risk.

Methodology: Created fractures in cylindrical cores using the Brazilian method. Conducted flow experiments through fracture to infer its effective aperture as a function of confining pressure. Degraded disassembled fracture faces with hydrochloric acid to simulate exposure to CO$_2$-saturated brine along this conductive pathway and conducted further flow experiments. The apparatus consists of a Hassler core flow cell, two pressure transducers, a data acquisition system and a positive displacement pump.

Conclusion:
Cyclic loading of naturally fractured cement cores shows a decrease of aperture size with increased confining stress, hysteresis in loading / unloading cycle, and strain-hardening Exposing cement fracture faces to weak acid alters the chemical and mechanical properties of the faces. Depending on the extent of reaction, the mechanical weakening permits a much more rapid closure of aperture as confining stress increases.
Degradation of cement by CO$_2$-rich fluids coupled with decreasing reservoir fluid pressure could render leaky wellbores self-sealing.

Comments: Although the paper is written in a more understandable fashion, results are very similar to those from previous experiments by same authors.
SPE 126666 (2009)
Computation Studies of Two-Phase Cement- CO₂-Brine Interaction in Wellbore Environments
Authors: Carey, J.W., Lichtner, P.C.
Contribution: Not much, as it is mainly based on previous literature.
Objective: To summarize recent progress in numerical simulations of cement-brine- CO₂ interactions with respect to migration of CO₂ outside of casing.
Methodology: Used numerical model to simulate flow and reaction of supercritical CO₂ through cement as well as carbonation of wellbore cement due to CO₂-bearing caprock.
Conclusion:
Supercritical CO₂ will not flow through good quality cement due to the capillary properties of cement. Leakage of CO₂ is confined to wellbore interfaces and carbonation of cement occurs by diffusion of CO₂ into the cement from the interface. Carbonation by diffusion creates reaction fronts that are distinct from the uniform carbonation pattern generated by flow of CO₂ through cement.
Comments: A purely numerical approach is used here which makes the paper a little less interesting and detached from any possible experimental agreement.
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**CO₂ Underground Storage and Wellbore Integrity**

**Authors:** Gaurina-Medimurec, N., Pasic, B., Simon, K.

**Contribution:** Not much, as everything has already been described in previous works.

**Objective:** To investigate geochemical alteration of hydrated Portland cement due to supercritical CO₂ injection. To present new cement with better CO₂ resistance.

**Methodology:** Reviewed findings from literature.

**Conclusion:**
The injection wells as well as any well penetrating through the cap rock have to maintain sufficient integrity over a long time period.
The chemical interactions between injected CO₂ and existing set cements could potentially lead to leakage because of cement degradation.
The integrity of casing-cement and cement-formation interfaces appears to be the most important issue in the performance of wellbore systems in a CO₂ sequestration reservoir.
Reducing the amount of Portland cement by incorporating pozzolanic materials, reducing porosity/permeability, and adding reactive supplementary materials to reduce the Ca(OH)₂, as well as changing the C-S-H composition to a more CO₂-resistive one could reduce detrimental effects of carbonation on mechanical integrity and loss of zonal isolation.

**Comments:** A relatively low-level paper that is useful to get familiar with the topic.
Dynamic alteration along a fractured cement/cement interface: implications for long term leakage risk along a well with an annulus defect

Authors: Huerta et al.

Contribution: Not much, the paper is quite similar to previous works by same authors but presents a slightly more in-depth approach.

Objective: To determine the conditions under which leakage of CO$_2$-saturated brine through a fracture in cement becomes either self-sealing or self-reinforcing.

Methodology: Fractured cement samples using the Brazilian method. Reassembled core halves with an axial offset of about a millimeter. Used a caulk down the mating sides of the fractured halves to prevent fluid from moving outside the fracture and to keep the core halves in place. Conducted flow experiments using a Hassler cell assembly, pressure transducers to monitor inlet pressure and confining pressure, a positive displacement constant flow rate pump, a graduated cylinder and a digital pH probe to measure incremental volume and acid concentration of effluent fluid. Estimated fracture aperture from the measured relationship between flow rate and pressure drop using an equation published in literature. Evaluated the relationship between hydraulic aperture and confining stress by performing tests before and after acid injection, at different flow rates and different confining pressures.

Conclusion: These experiments support the conjecture that leakage paths for CO$_2$-saturated brine may be self-sealing or at least self-limiting. Both experiments showed a permanent reduction in effective hydraulic aperture. The influence of confining stress on the aperture of a leakage pathway is significantly less than in analogous experiments in which chemistry was decoupled from confining stress. The load-supporting asperities on the fracture faces appeared to undergo no reaction, so minimal change in the mechanical behaviour of the system occurred

Comments: The methods are similar to previous works by same authors but a more rigorous approach to the problem leads to some novel results.
Stability of a leakage pathway in a cemented annulus

Authors: Deremble et al.

Contribution: First paper to present an explicit criterion for the self-sealing or self-reinforcing characteristic of a leakage pathway.

Objective: To examine theoretically and numerically the evolution of a CO₂ leak through a pre-existing leak path in the cement sheath.

Methodology: Detailed the mechanism of opening and clogging of the leak path, qualitatively and using numerical results. The numerical model of the flow takes into account the particular physics and geometry of the problem. Investigated the governing equations and presented a dimensional analysis in order to identify the driving mechanisms and define the dimensionless groups that rule the process. Used this analysis to predict the domain of stability of the defect by extracting a general criterion corresponding to the clogging conditions and to represent the domain of stability of the leakage pathway in the cemented annulus.

Conclusion:
The behaviour of the system is highly dependent on the effective width of the defect. The data on this particular value is sparse and quite uncertain, which may jeopardize the validity of the prediction.
The characteristic time of the healing process may be prohibitive, depending on the initial parameter of the system.
There is no stability over time for a leak path in a cemented annulus. As soon as it appears, the pathway will either be enlarged or clogged by the reactive leak, but never remain with the same resistance to the flow.

Comments: Good paper that uses analytical as well as numerical methods to derive some new and interesting results.