# Reactive Transport Modeling of Dissolved $CO_2$ in the Cap Rock Base During $CO_2$ Sequestration (Sleipner Site, North Sea)

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**Summary**

**During geological $CO_2$-sequestration, dissolved $CO_2$ will diffuse into the lower section of the cap rock where, depending on the cap rock mineralogy, it might trigger geochemical reactions affecting crucial parameters such as porosity and permeability (and therefore possibly the sealing capacity). This is modeled using reactive transport modeling for the $CO_2$-injection project at the Sleipner field (North Sea) where $CO_2$ is injected in the Utsira Sands underlying the Nordland Shale cap rock. Preliminary long term results suggest a slight decrease in porosity at the lower section of the cap rock. The sensitivity of the calculated porosity change with respect to the initial cap rock mineralogy (plagioclase composition) is discussed.**

## 1. Introduction

At the Sleipner field in the North Sea, $CO_2$ (here a byproduct of gas exploitation) is stored in an underground saline aquifer for the very first time. Seen as a way to dispose of $CO_2$ and avoid climate effects, this method has profound potential importance (SACS2, 2002). A group of energy companies together with scientific institutes and environmental authorities in Norway, Denmark, the Netherlands, France and the UK formed the Saline Aquifer $CO_2$ Storage (SACS) Project consortium which aims are to develop a consensus about the monitoring results and the validity of the available models and tools through an interdisciplinary approach. These projects are demonstrating that fossil fuel use for power generation with minimal $CO_2$ emission is possible and achievable as part of the EU strategy on the sustainable industrial development on the medium term (Torp & Gale, 2002).

This paper focuses on the geochemical aspects of the Sleipner injection project, with special attention to the long term integrity of the cap rock preventing upward migration of the injected $CO_2$. Geochemical reactions between dissolved $CO_2$ and the minerals present in the cap rock can lead to porosity changes inducing permeability changes in the cap rock. An increase in porosity can be deleterious since this would facilitate $CO_2$ transport through the cap rock. A decrease in porosity could further seal off the base of the cap rock and contribute to an improved sealing capacity. The effects of geochemical reactions which occur as a consequence of the diffusion of dissolved $CO_2$ in the cap rock at Sleipner are investigated using reactive geochemical transport modeling taking into account reaction kinetics.

## 2. The Sleipner site

The Sleipner area is located in the Norwegian part of the North Sea (Norwegian block 15/9). $CO_2$ is injected into the Mio-Pliocene Utsira Sand, a highly elongated sand reservoir with an area of some 26100 $km^2$ of which the depth ranges between 700 and 1000 m. The Utsira Sand is overlain by the sediments of the Nordland Group, which are reported to be mainly shales in their lower part (Figure 1). These sediments are expected to provide a seal for the Utsira Sands and to inhibit vertical $CO_2$ migration, with capillary leakage of $CO_2$ unlikely to occur. Repeat seismic surveys have clearly shown that the injected $CO_2$ moves, due to buoyancy effects, from the injection point and accumulates under the overlying cap rock (Torp & Gale, 2002).

At the base of the cap rock upward diffusion of dissolved $CO_2$ can take place leading to the acidification of the formation waters and interaction with the minerals present in the cap rock. Diffusion is a very slow process, even in cases where high concentration gradients are present, which means only the base of the cap rock will potentially be affected by geochemical interactions.

## 3. Geochemical modelling

Long term geochemical modeling (specifically over thousands of years) is still very much a research topic and involves large amounts of uncertainties at different levels (Gunter et al., 2000). However, it is the only tool available to assess the interaction between the $CO_2$ and the rock in the future.

The modeling is performed at 37°C, the Sleipner reservoir temperature, and at a pressure equal to the hydrostatic pressure of 10 MPa. According to Duan et al. (1992), under these conditions the fugacity of $CO_2$ is 52 atm. This $CO_2$ fugacity is introduced in the model. Two types of model calculations are performed. The first type, kinetic batch modeling, looks at the geochemical interactions between the dissolved $CO_2$ in the cap rock formation water and the minerals present in the cap rock only, without taking into account mechanisms of transport of reactive aqueous species. These model calculations are performed over a 15000 year period after injection. The second type of modeling couples the kinetic batch modeling to diffusional transport into the base of the cap rock. During this type of modeling, which is performed over 3000 years only because of calculation time constraints, all chemical parameters are identical to the kinetic batch modeling.

Changes in porosity of the cap rock are calculated using the molar volumes of the primary and secondary minerals.

***Mineralogical composition of the cap rock and formation water composition***

The mineralogy of the Nordland shale cap rock is conceptualized based on measurements of a selection of cutting samples taken at the base of the cap rock as described in Bøe and Zweigel (2001). The shales consist predominantly of quartz and clays such as illite, kaolinite and smectite as well as a small amount of feldspars (K-feldspar and plagioclase). Some modification of the observed mineralogy is necessary because the description of the observed minerals is not specific enough to allow it to be defined as mineral phases in the model: for the chlorite group minerals clinochlore is used as a proxy, the phase described as mica/illite is modeled as illite. Smectite is introduced as a Mg and Fe rich smectite variation. The observed plagioclase is modeled as albite only or a 50/50 mixture of albite and anorthite. Finally, the thermodynamic properties of chalcedony are selected instead of quartz, since chalcedony is generally the controlling $SiO_2$–phase in sedimentary rock at low temperatures. The mineralogical composition is listed in Table 1.

Table 1 Mineralogical composition of the Nordland Shale used for the modeling, the molar volumes, specific surface areas and kinetic rate parameters at 37°C.

| | % | Amount present (mole/l water) | Molar volume ($10^{-6}$ $m^3$/mole) | Specific surface area ($m^2$/g) | Kinetic rate (log) (37°C) (mole $m^{-2}$ $s^{-1}$) | Source of kinetic rate data |
|---|---|---|---|---|---|---|
| Albite | 6.2 | 12.8 | 100.25 | 6.95E-02 | -8.44 | Blum and Stillings (1995) |
| Anorthite | 6.2 | 12.8 | 100.75 | 6.59E-02 | -4.93 | Blum and Stillings (1995) |
| Calcite | 1.0 | 5.6 | 36.93 | 6.71E-02 | -6.35 | Azaroual et al. (2003) |
| Chalcedony (Quartz) | 21.5 | 196.0 | 22.69 | 6.86E-02 | -11.73 | Azaroual et al. (2003) |
| Clinochlore-7A | 4.1 | 4.0 | 20.98 | 1.13E-01 | -11.63 | Nagy (1995) |
| Dawsonite | 0* | 0* | 59.3 | 8.49E-02 | -6.86 | Intermediate calcite/dolomite |
| Dolomite-dis | 0* | 0* | 64.39 | 6.35E-02 | -7.38 | Azaroual et al. (2003) |
| Gypsum | 0* | 0* | 74.69 | 7.89E-02 | -7.38 | Azaroual et al. (2003) |
| Illite | 24.7 | 35.21 | 59.89 | 4.68E-01 | -13.08 | Knauss and Wolery (1989) |
| Kaolinite | 18.0 | 38.15 | 99.52 | 1.16E+00 | -12.54 | Nagy (1995) |
| K-feldspar | 2.1 | 4.08 | 108.87 | 7.11E-02 | -8.79 | Blum and Stillings (1995) |
| Pyrite | 2.8 | 12.90 | 23.94 | 3.63E-02 | -3.72 | Wiersma and Rimstidt (1984) |
| Siderite | 1.6 | 8.60 | 29.37 | 4.61E-02 | -7.38 | Azaroual et al. (2003) |
| Smectite-high-Fe-Mg | 8.8 | 11.93 | 140.71 | 1.04E+00 | -13.25 | set to muscovite rate (Nagy, 1995) |
| Magnesite | 0* | 0* | 28.02 | 6.04E-02 | -7.38 | Azaroual et al. (2003) |

*Introduced as secondary mineral in the model

The composition of the formation water in the cap rock was at the time this research is performed unknown. Therefore the composition of water samples resulting from experiments on the Utsira sands (Czernichowski-Lauriol et al., 1999) was equilibrated with the cap rock mineralogy and used as initial cap rock formation water (Gaus et al., 2002).

***Rate equation for the water-rock interactions***

The kinetic rate law for the dissolution and precipitation of the minerals is simplified from Lasaga (1984):

$$rate_m = A_m k(T)_m \left( a_{H+} \right)^n \left[ 1 - \left( \frac{Q_m}{K_m} \right) \right] \quad (1)$$

where *m* is the mineral index, *rate* is the dissolution/precipitation rate (positive values indicate dissolution, negative values precipitation), A is the reactive surface per kg water, *k(T)* is the temperature dependent rate constant, $a_{H+}$ is the proton activity, *n* is the order of the reaction (0<n<1), *K* is the equilibrium constant for the mineral water reaction written for the destruction of one mole of mineral and *Q* is the corresponding ion activity product. For all minerals it is assumed that the precipitation rate equals the dissolution rate.

The dependency of the rate constant on the temperature is defined as:

$$k(T) = k_{25} \exp\left[ \frac{-E_a}{R} \left( \frac{1}{T} - \frac{1}{298.15} \right) \right] \quad (2)$$

where $E_a$ is the activation energy (in J/mol), $k_{25}$ is the rate constant at 25°C (in mol/m$^2$s), *R* is the gas constant (8.31 J /mol K) and T is the absolute gas temperature.

The previous rate law is applied to all the minerals present in the cap rock apart from pyrite, for which a rate law based on Wiersma and Rimstidt (1984) is used:

$$rate = k(T) \frac{A \left( Fe^{3+} \right)}{m_{solution}} \quad (3)$$

with $m_{solution}$ the mass of the solution in kg. This law better reflects the redox determined behavior of pyrite.

***Rate expression data***

The kinetic rate parameters for the selected minerals are taken from the literature and recalculated for 37°C (Table 1). Literature data for the value of the power (n) in the rate expression are fragmentary and sometimes inconsistent. Therefore a uniform average value for *n* of 0.5 is selected for all minerals. With respect to dawsonite no kinetic rates are published to our knowledge. An intermediate rate between dolomite and calcite is selected.

Since no direct measurements of BET-surfaces are available for the Nordland Shale, the geometric surface area is used for the calculation of the specific reactive surface area assuming that mineral grains are spherical. For all the minerals, apart from the clays, an average grain diameter of 0.0033 cm is assumed. For the clays an average grain diameter of 0.0002 cm is taken which is the coarsest clay size since also the silt fraction contains some “clay” minerals (Kemp et al., 2001).

Interaction with the minerals is generally expected to occur only at selective sites at the surface and the difference between total surface area and reactive surface area can be between 1 and 3 orders of magnitude. Hence, a scaling factor of 0.001 is used for all minerals to account for the difference between geometric surface area and the specific reactive surface area. The surface area for precipitating minerals is very difficult to estimate. Here it is assumed that for all precipitating minerals the reactive surface area during precipitation is 10000 m$^2$/kg water.

Because many parameters in the rate expression are insufficiently known or were not estimated for very long time scales, the length of time over which reactions take place is relative rather than absolute.

***Sensitivity analysis***

An analysis was carried out with respect to the selection of the secondary minerals based on equilibrium batch modeling. Secondary minerals are those minerals formed as a result of geochemical reactions and which are not present in the initial cap rock mineralogy. A secondary mineral assemblage was assumed acceptable when the resulting solution after being brought in equilibrium with $CO_2$ was not supersaturated with respect to minerals which would normally precipitate under the current conditions. The selected secondary minerals were disordered dolomite, dawsonite, magnesite, and gypsum.

Although the mineralogical analysis of the cap rock cuttings does not specify the exact composition of the plagioclase, this is likely to have an important impact on the modeled results. To assess this, one run is performed where all plagioclase consists of albite (Run 1), while for a second run a 50/50 ratio between albite and anorthite is defined (Run 2).

***Diffusion modeling***

Diffusion into the cap rock can be described by Fick's law for diffusion in sediments (Domenico & Schwartz, 1990):

$$J = -D'_d \; grad(C) \qquad (4)$$

with $J$ the diffusional flux (moles/$L^2$T); $D'_d$ the effective diffusion coefficient ($L^2$/T) and $C$ the concentration (moles/$L^3$).

The diffusion coefficient of $CO_2$ in water only is 1.8 $10^{-9}$ $m^2$/s (20°C). When taking into account the porous medium with an estimated porosity of 5%, this value decreases according to the approach described in Helfferich (1966) to a range of 4.5 $10^{-11}$ $m^2$/s down to 1.18 $10^{-12}$ $m^2$/s. For the simulations the highest value of this range (4.5 $10^{-11}$ $m^2$/s) is used.

**4. Results**

The code used for the modeling is PHREEQC (V2.6) (Parkhurst and Appelo, 1999). The Davies approach is used for the aqueous speciation calculation. For this the LLNL.dat database was adapted. For the diffusion modeling the lower 10 m section of the cap rock is modeled as 10 cells of 1 m each.

*Dominant reactions*

The results for the kinetic batch modeling are shown in Figure 2 for the case where all plagioclase is present as albite (run 1) and in Figure 3 for the case where plagioclase is present as a 50/50 mixture of albite and anorthite (run 2).

With an infinite source of $CO_2$ with a fugacity of 52 atm present the concentration of dissolved $CO_2$ in the formation water stabilizes at around 1.14 moles/l. An initial pH decrease just after contact of the formation water with the $CO_2$ occurs and the pH drops to a value of 3.0 during the first 4 days. This triggers the dissolution of calcite which stabilizes the pH at 4.5 after approximately 9 years. This fast initial reaction of the geochemical system to the $CO_2$ injection is thus caused by buffering of the carbonate system. The amount of calcite dissolved is minor when compared to the amount which precipitates in subsequent reactions.

When *all plagioclase is present as albite*, the dominant reaction in the system over the 15000 year time period is the dissolution of albite with the subsequent formation of kaolinite and calcite using the limited amount of $Ca^{2+}$ initially present in the formation water:

$$\underset{\text{albite}}{2NaAlSi_3O_8} + CO_2 + 2H_2O + Ca^{2+} \rightarrow \underset{\text{chalcedony}}{4SiO_2} + \underset{\text{calcite}}{CaCO_3} + \underset{\text{kaolinite}}{Al_2Si_2O_5(OH)_4} \qquad (5)$$

According to this reaction $CO_2$ becomes permanently sequestered as calcite. After approximately 10000 years, this reaction slows down due to the decreasing $Ca^{2+}$ concentration in the formation water and the formation of dawsonite, which does not require $Ca^{2+}$, starts to occur:

$$\underset{\text{albite}}{NaAlSi_3O_8} + CO_2 + H_2O \rightarrow \underset{\text{chalcedony}}{3\,SiO_2} + \underset{\text{dawsonite}}{NaAlCO_3(OH)_2} \qquad (6)$$

Both albite alteration reactions are extremely slow and over the 15000 year period very little carbonates have formed. Overall the system is very little reactive when in contact with $CO_2$ and the porosity is hardly affected by dissolution and precipitation reactions (Figure 2 (D)). Solubility trapping ($CO_2$ trapped in the solution as dissolved $CO_2$ or as various complexes such as $NaHCO_3$, $CaHCO_3^+$, etc) is the main $CO_2$ sequestration process with dissolved $CO_2$ being the bulk of this amount.

When *plagioclase is present as albite and anorthite*, anorthite proves to be more reactive than albite and the alteration of anorthite dominates the feldspar reactions leading to the formation of calcite and kaolinite:

$$\underset{\text{anorthite}}{CaAl_2(SiO_4)_2} + CO_2 + 2H_2O \rightarrow \underset{\text{calcite}}{CaCO_3} + \underset{\text{kaolinite}}{Al_2Si_2O_5(OH)_4} \quad (7)$$

This reaction is completed after approximately 5000 years when all anorthite has disappeared. Also here the alteration of albite occurs according to reaction (6) at the same rate. The minor amount of dawsonite formed is unstable with the saturation index of dawsonite dipping slightly below 0 between 1200 and 12500 years. Minor K-feldspar alteration (in the order of 0.01 moles) is also predicted. The mineralogical transformation due to the anorthite alteration leads to an absolute porosity reduction of 2.8%. Mineral trapping is the dominant process and approximately 12 moles of $CO_2$ is trapped as carbonates in a rock volume in contact with 1 l of formation water.

Apart from the formation of kaolinite, the reactivity of the clays (illite, smectite, etc) is low. This is caused by their extremely slow dissolution rates. It is likely that over longer time-scales the reactivity of the clays would become more important.

*Can one use the thermodynamic equilibrium hypothesis as an alternative to kinetic modeling?*

Kinetic calculations necessary to provide predictions over thousands of years are extremely time consuming. In order to reduce calculation times one could therefore be tempted to discard the reaction kinetics and assume that after 15000 years the system will be close to or at the equilibrium state. However, when mineralogical changes as a consequence of $CO_2$ interaction after 15000 years are compared with these occurring assuming equilibrium between $CO_2$ and the cap rock large differences are observed (Figure 3). At the equilibrium state large changes in the clay minerals are calculated with dissolution of smectites and illite and precipitation of chalcedony, kaolinite and K-feldspar as well as large amounts of carbonates (although calcite disappears). When incorporating kinetics, after 15000 years the system is still far from the equilibrium state and using the equilibrium hypothesis would overestimate dramatically the geochemical impact of $CO_2$-injection and might erroneously predict major porosity changes.

*Diffusion of $CO_2$ in the base of the cap rock – reactive transport*

The penetration profile of the dissolved $CO_2$ into the cap rock, the major mineralogical changes as well as the porosity changes profile after 1000, 2000 and 3000 years is shown in Figure 5 and Figure 6 for run 1 and run 2 respectively.

When plagioclase is present as albite only (run 1) the impact of geochemical reactions on the base of the cap rock is minor. Figure 5 (A) shows that the diffusion of dissolved $CO_2$ occurs almost at the same rate as if no geochemical interaction would take place. Some alteration of albite is observed in the lowest 4 m of the cap rock. No significant porosity changes are observed in the area where geochemical interaction occurs. The section of the cap rock which is affected by elevated $CO_2$ concentrations approximates 6 m after 3000 years.

The presence of anorthite (run 2 – Figure 6) slows down considerably the diffusion of dissolved $CO_2$ in the cap rock. After 3000 years only the lower meter of the cap rock has elevated dissolved $CO_2$ concentrations. The dissolution of anorthite and the subsequent precipitation of calcite and kaolinite causes a porosity decrease in the lower 2 m of the cap rock with a maximum of 2.8 % in the first meter. The maximum porosity decrease depends on the initial amount of anorthite present since all anorthite is removed after approximately 2500 years in the first meter of the cap rock.

## 5. Conclusions

Evaluating the long term geochemical behavior of the injected $CO_2$ and its possible impact on the cap rock integrity is a crucial aspect of geological $CO_2$ sequestration safety assessment. Long term diffusion modeling of $CO_2$ into the Nordland Shale cap rock at Sleipner shows that:

- Due to low diffusion velocities the geochemical impact is limited to the lower section (first meters) of the cap rock even after thousands of years.

- Based on the initial mineralogy of the cap rock plagioclase alteration is identified as the most important reaction involving the dissolved $CO_2$, leading to the precipitation of clays (kaolinite) and carbonates (calcite and dawsonite).

- A minor porosity decrease (up to a few percentages) can occur in the lowest meters of the cap rock depending on the initial composition of the feldspars present. It is possible that this minor porosity reduction locally improves the cap rock sealing capacity.

These conclusions are based on the assumption that the cap rock is a homogeneous medium and that diffusion is the only mechanism in the cap rock causing the displacement of dissolved $CO_2$. Because many parameters in the rate expression cannot be known accurately and because the rate parameters were not estimated for very long time scales, the kinetic calculation has to be regarded as relative in time rather than absolute. It is however clearly illustrated that the use of the thermodynamic equilibrium approach instead of including kinetics would definitely lead to an erroneous assessment of the impact of geochemical reactions on the caprock.

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Figure 1. Overview of the Sleipner area with $CO_2$-injection (SACS2, 2002).

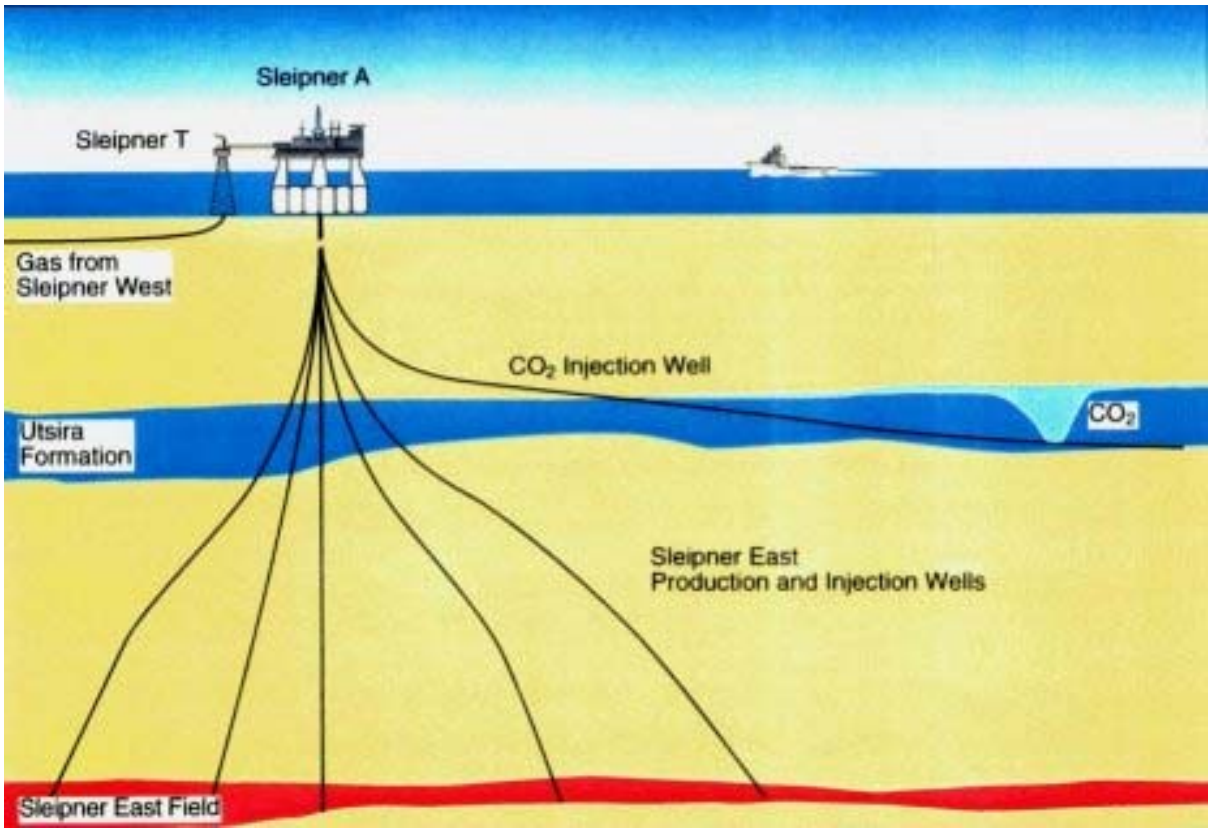

Figure 2. Kinetic batch modelling of the interaction between dissolved $CO_2$ and the cap rock (case 1: all plagioclase is present as albite). A) evolution of the most relevant species in the solution. B) dissolving and precipitating minerals of the dominant reaction (reactions (5) and (6)). C) geochemical $CO_2$ sequestration. D) porosity change.

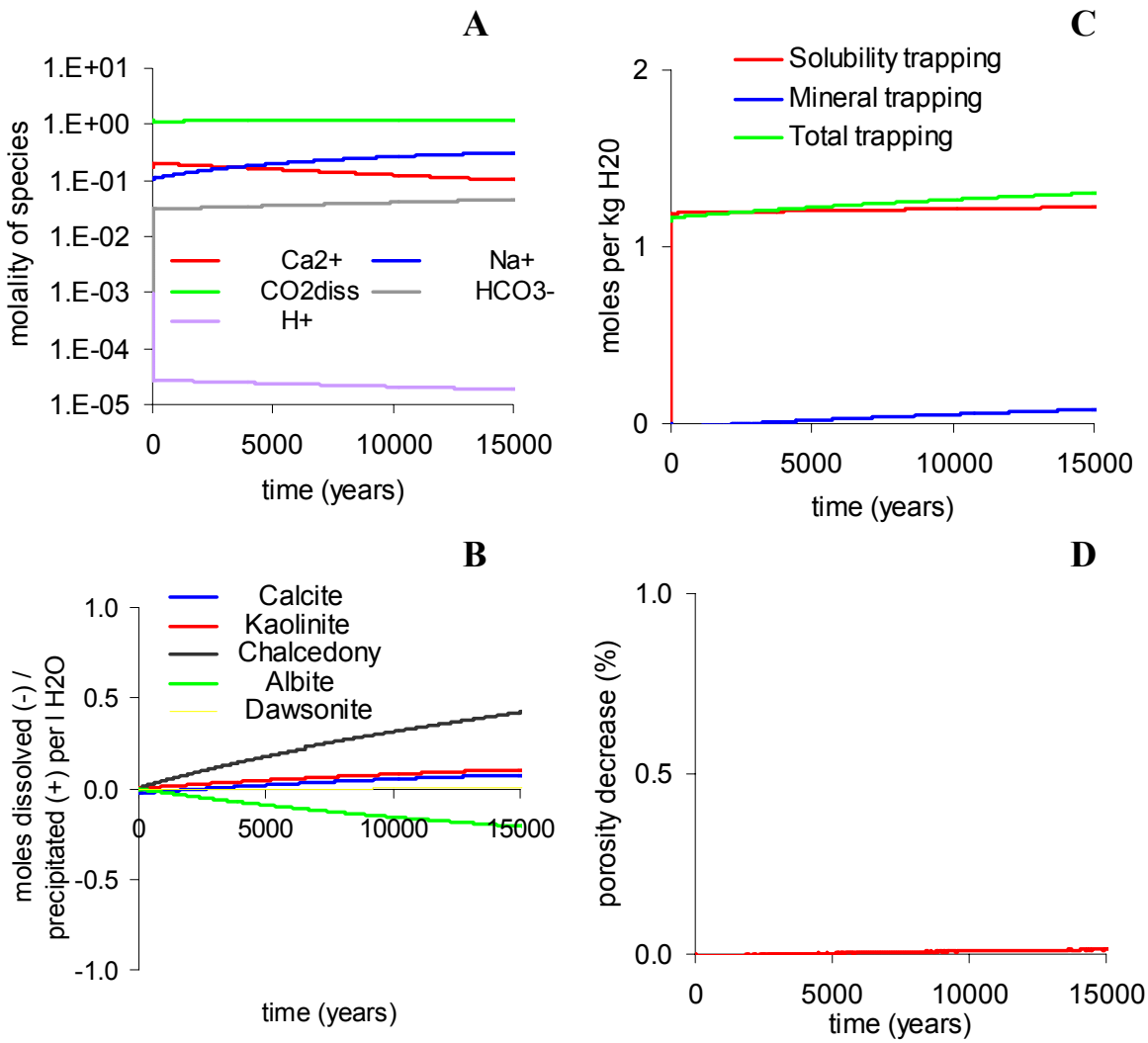

Figure 3. Kinetic batch modeling of the interaction between dissolved $CO_2$ and the cap rock (case 2: plagioclase is present as 50/50% albite/anorthite). A) evolution of the most relevant species in the solution. B) dissolving and precipitating minerals of the dominant reaction (reaction (7)). C) geochemical $CO_2$ sequestration. D) porosity change.

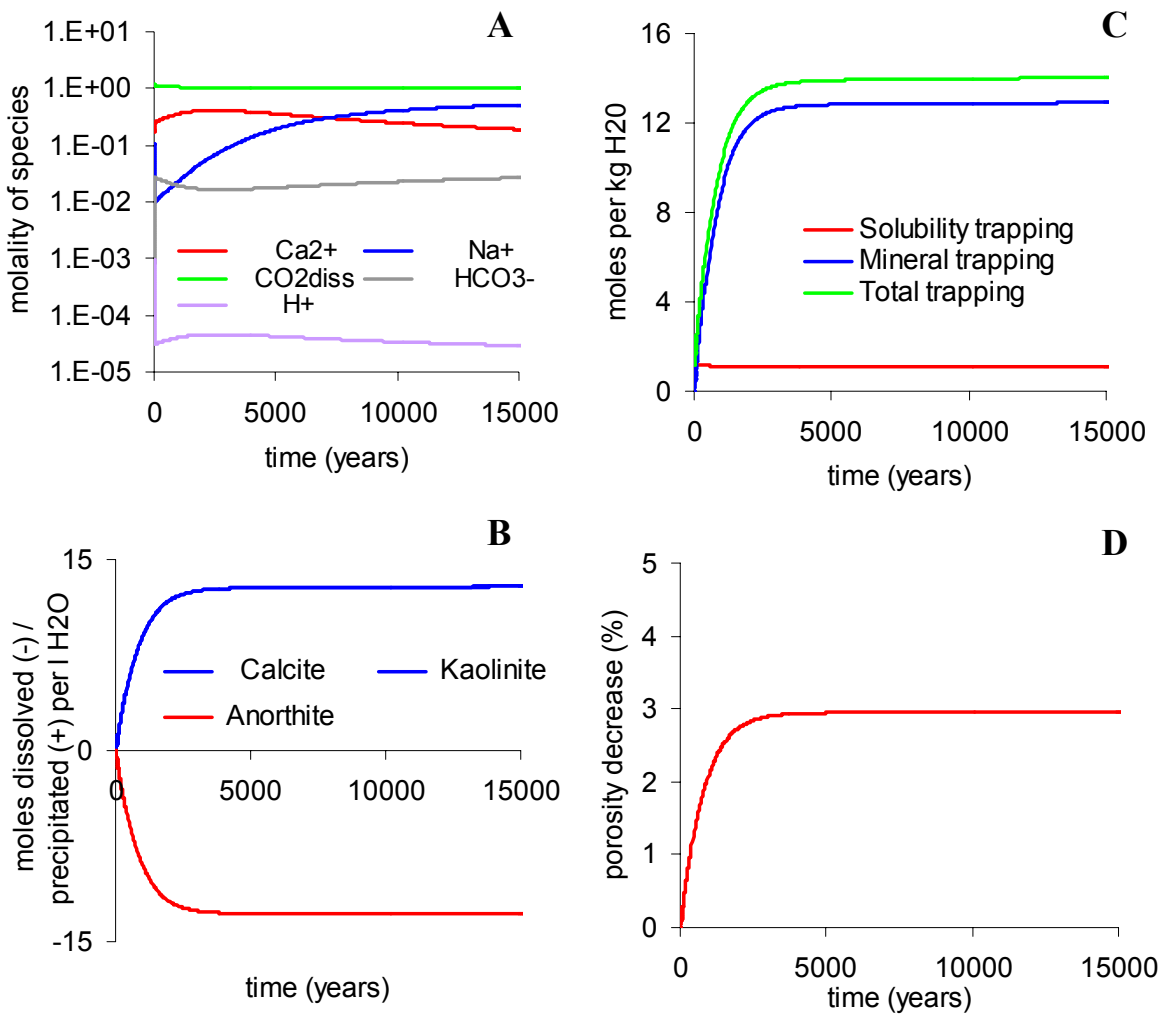

Figure 4. Comparison of the mineralogical changes in the cap rock at the Sleipner site due to $CO_2$ injection after 15000 years of kinetic calculation (run 2) and once the equilibrium state is reached.

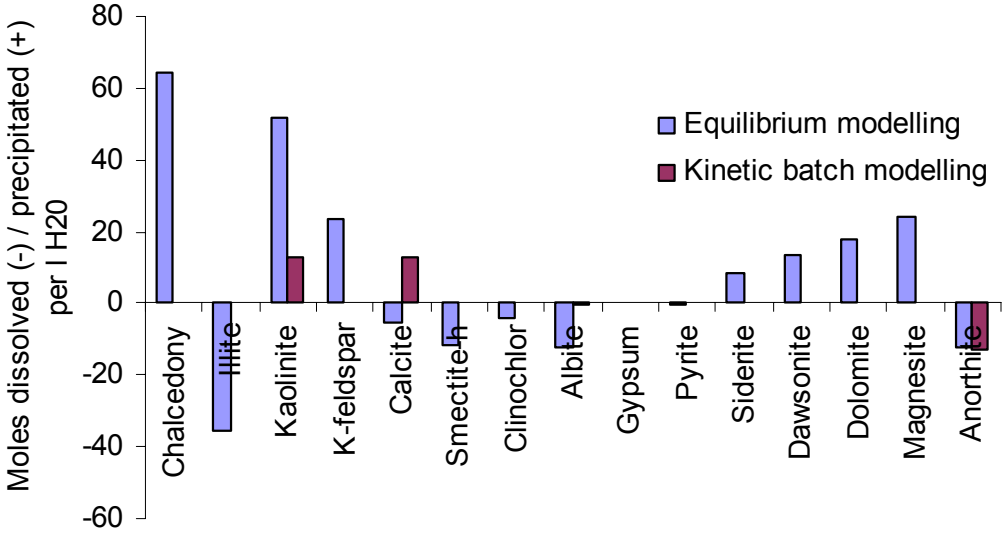

Figure 5. Profiles of impact of dissolved $CO_2$ in the cap rock after 1000, 2000 and 3000 years (all plagioclase present as albite). A) diffusion of dissolved $CO_2$ into the cap rock. B) dissolution and precipitation of minerals (reaction (5) and (6)). C) porosity changes.

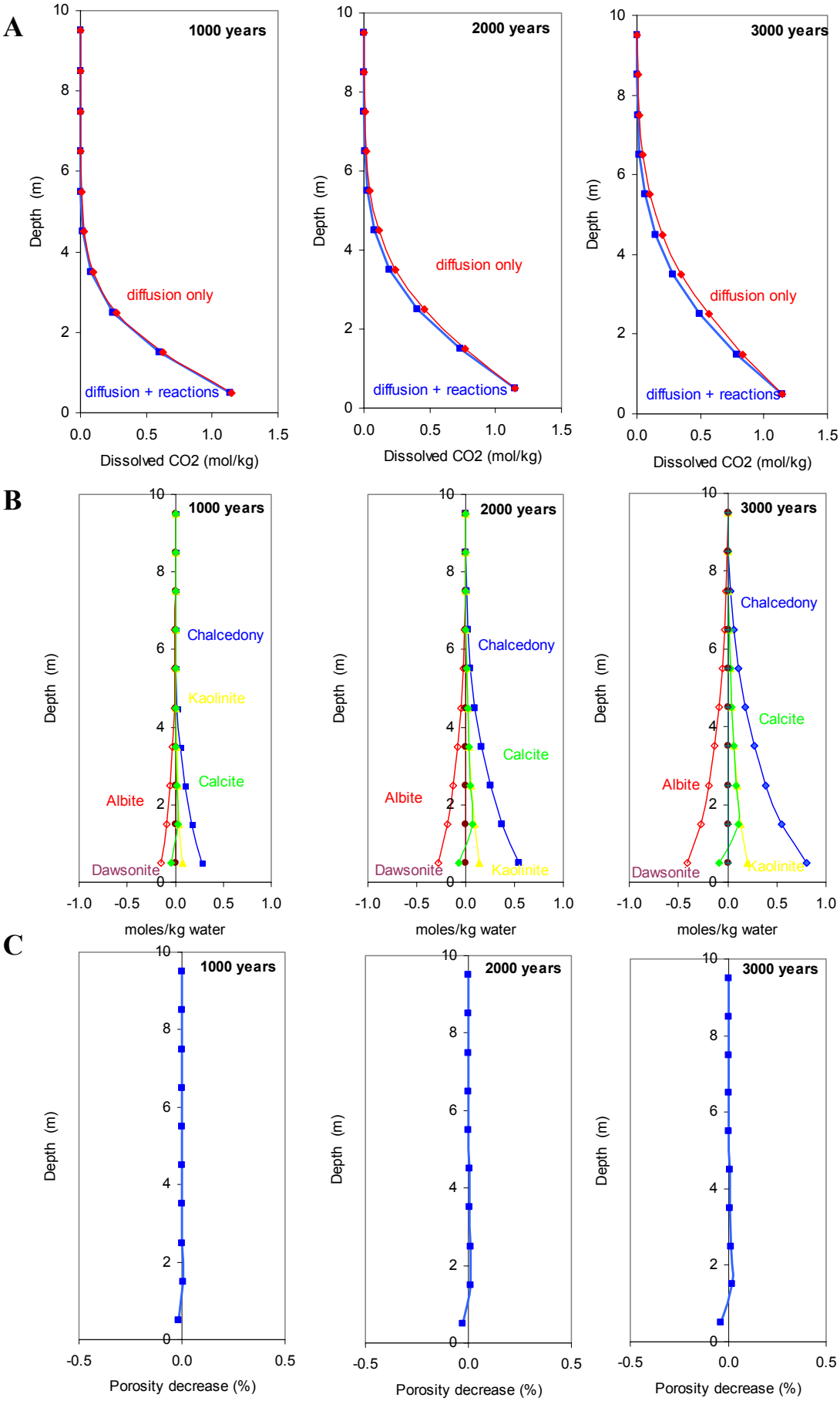
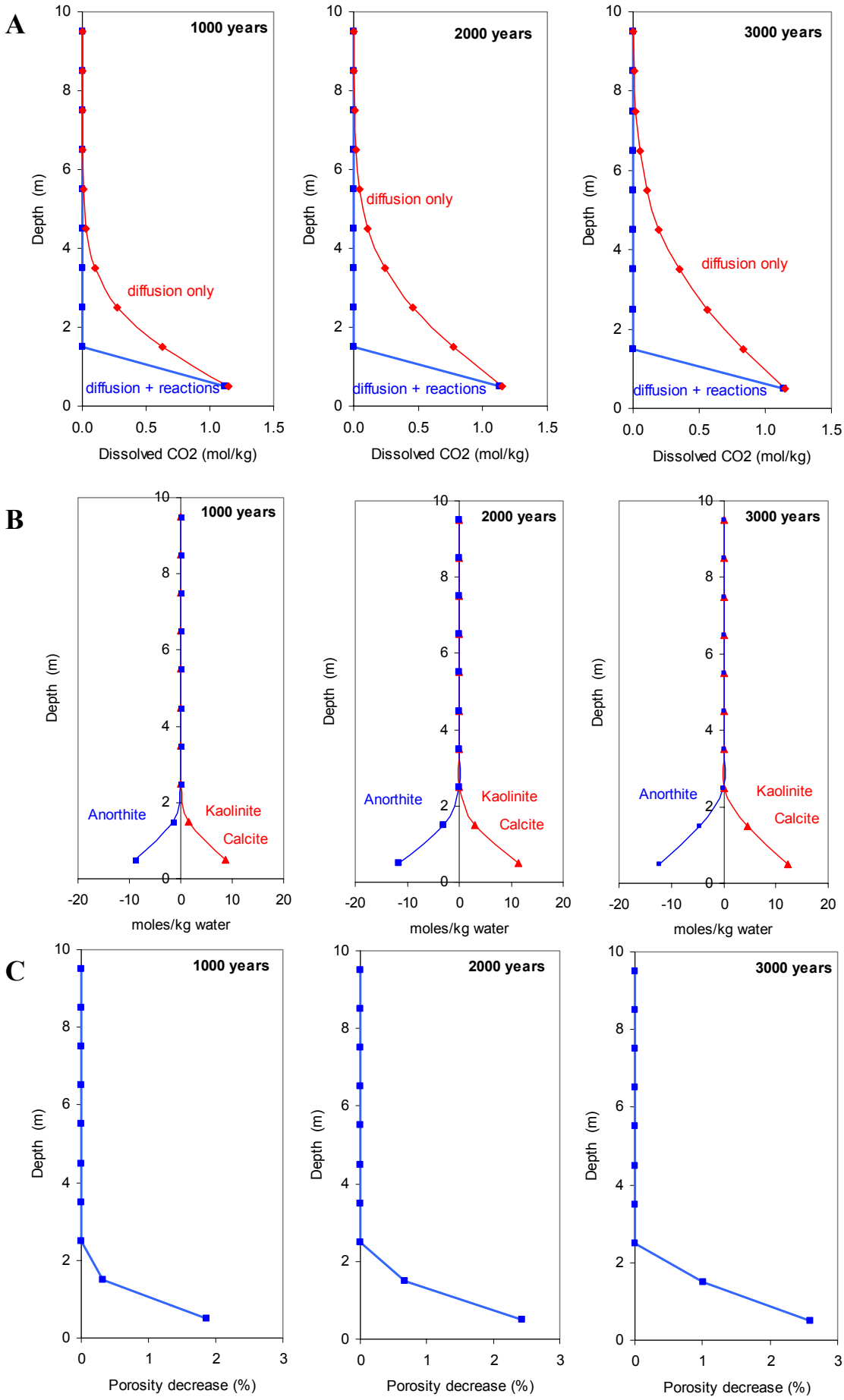
Figure 6. Profiles of impact of dissolved $CO_2$ in the cap rock after 1000, 2000 and 3000 years (plagioclase present as 50/50% albite/anorthite). A) diffusion of dissolved $CO_2$ into the cap rock. B) dissolution and precipitation of minerals (reaction 7). C) porosity changes.