

Numerical Simulation of CO₂ Sequestration in Natural CO₂ Reservoirs on the Colorado Plateau

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Abstract

This paper outlines the proposed research and summarizes pre-project work that forms a basis for a new research program on CO₂ sequestration in saline aquifers. The pre-project work considers storage and disposal of CO₂ several kilometers beneath the surface in generic aquifers and demonstrates the use of reactive chemical transport modeling to simulate mineral sequestration of CO₂. The current research project applies these techniques to particular saline reservoirs on the Colorado Plateau. In a companion paper (Allis *et al*, these proceedings) the properties of the natural CO₂ reservoirs of the Colorado Plateau are described. This is joint work by Industrial Research Ltd., University of Utah and, Utah Geological Survey funded by DOE grant #DE-FC26-00NT4096.

Introduction

Groundwater (freshwater) reservoirs usually do not extend below 500 m depth, and will become increasingly utilized, as industrial and population development requires more fresh water. Maintaining unpolluted water supplies will be of great importance in the future, and disposal of waste gases into potable groundwater reservoirs is likely to be unacceptable. Where aquifers are saline, however, they may provide an ideal site for CO₂ sequestration. The Colorado Plateau and adjacent Rocky Mountain region is an obvious area for the application of such technology. This area contains over 10,000 MW of coal fired power stations emitting almost 100 million tonnes of CO₂ each year. Much of the region is underlain by saline aquifers that may be suitable for the sequestration of CO₂ from these power stations.

We have recently begun a three-year research project to investigate the storage potential of these aquifers. This paper outlines the proposed research and summarizes pre-project work that forms a basis for the current research program. In this we consider storage and disposal of CO₂ several kilometers beneath the surface in generic aquifers (Weir 1996a,b) and demonstrate the use of reactive chemical transport modeling to simulate mineral sequestration of CO₂ from aquifer fluids.

For many years it has been possible to compute the transport of heat and mass within the earth using simulators such as TOUGH2 and TETRAD. Reaction-path simulators are also well advanced, and used as a matter of course by geochemists to unravel the intricacies of the chemistry of fluids within the earth (e.g. Reed 1982). Recently there has been a growing interest in combining these disciplines to allow the modeling of reactive chemical transport in porous

media (e.g. Lichtner (1992), Steefel and Lasaga (1994), Friedly and Rubin (1992), White (1995) and Lichtner and Seth (1996). Our planned research will use the simulator CHEMTOUGH2 (White 1995) to model the eventual fate of CO₂ injected into a saline aquifer typical of those found on the Colorado Plateau. Such a simulator allows modeling of all the important sequestration mechanisms for CO₂, trapping, dissolution in aquifer fluid and mineral sequestration.

Objective

The three most important mechanisms for the sequestration of carbon dioxide in saline reservoirs are:

- 1) Trapping. In this case CO₂ is trapped as a gas or super-critical fluid beneath some low permeability barrier (caprock) within the earth. The obvious natural analogy to this is a natural gas reservoir. At 100 bars and 100 °C, storage density is about 300 kg/m³ of pore space.
- 2) Dissolution in the natural aquifer liquid (usually water or brine). As a first approximation the solubility of CO₂ is described by Henry's law at 100° C and 100 bars this gives a storage density of about 40 kg/m³ of pore space.
- 3) Mineral sequestration. The CO₂ rich aquifer fluid reacts with host rocks to form various carbonate minerals. Calcite for example contains approximately 1200 kg/m³ of CO₂.

Each of these mechanisms is likely dominant at a particular time in the processes of containment of CO₂ in saline aquifers. During the injection phase, CO₂ exists largely as a gas or supercritical fluid and the existence of a low permeability barrier to trap the CO₂ is the most important feature of the target formation. At later times the dissolution of the fluid CO₂ into the aquifer brine becomes the dominant mechanism for containment and storage. Finally the reaction of the CO₂ rich aquifer fluids with reservoir rocks can permanently trap CO₂ in a high-density mineral phase.

The aim of our research is to model the injection of CO₂ into a reservoir representative of those found on the Colorado Plateau, taking into account each of these mechanisms.

Approach

We will use the reactive transport code, CHEMTOUGH2 to model the injection of CO₂ into a saline aquifer based on the Farnam dome region of the Colorado Plateau. Such a simulator is capable of modeling all the important sequestration mechanisms described above. Model results, particularly those associated with mineral sequestration, will be verified with data taken from natural CO₂ reservoirs in the region. A number of core samples are available from these and will allow determination of rock alteration products.

Project Description

We have performed four simulations on generic aquifers. In the first three we consider trapping of CO₂ beneath a low permeability confining layer and in the fourth the reaction of a CO₂-rich aquifer fluid with a reservoir rock composed of plagioclase, k-feldspar and quartz.

In this section, we select a set of working parameters for our numerical simulations. These describe the structure and properties of the aquifers in which the CO₂ is injected, and also the injection rates to be used.

The first three simulations (referred to as scenarios A, B and C) consider injection into the reservoir shown in Figure 1. Initially the reservoir is assumed to have a temperature gradient of 30°C/km and be at hydrostatic pressure consistent with this temperature gradient. The density of CO₂ at reservoir pressure and temperature as a function of depth for temperature gradients of 25, 30 and 35 °C/km is shown in Figure 2. Density increases almost linearly with depth to about 1.5 – 2 km and below this it begins to plateau. Also below three kilometers it is usual for permeability to reduce significantly, due to compression of the rock. An injection depth between 1.5 and 3 km is likely to be the most economic. For this work we have used a depth of 3km, this gives an injection point temperature and pressure of about 100°C and 300 bars.

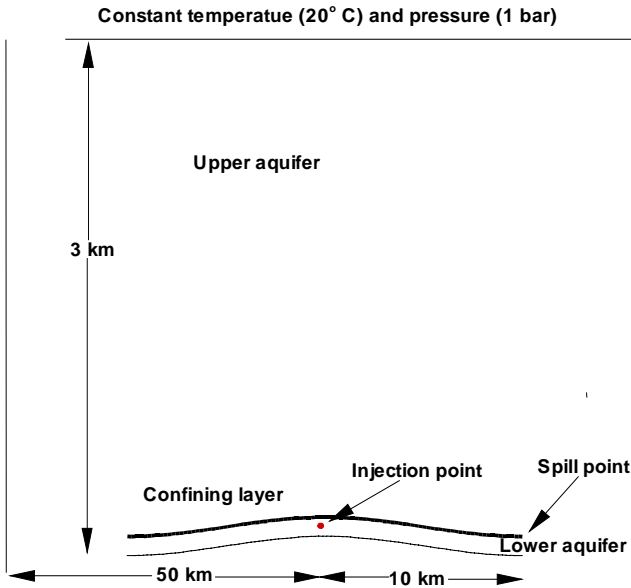


Figure 1: Generic reservoir used for simulations.

Case	Aquifer			
	Lower	Confining	Upper	Capillary Pressure
A	100 mD	0.01 mD	10 mD	No
B	100 mD	1 mD	10 mD	No
C	100 mD	1 mD	10 mD	Yes

Table 1: Permeability structures used in the first three scenarios.

We assume that a constant pressure boundary to the reservoir at a radius of 50 km. This is distant enough for the horizontal boundaries to have no affect on the calculation. Individual rock strata in petroleum reservoirs typically have gradients to the horizontal of about 1%. Thus, on a 50-km horizontal length scale, we might expect variations in height of up to 500 m. Our model consists of three such strata, which we have named the upper, confining and lower aquifers. The lower aquifer is at a depth of three kilometers and has a thickness of 100 m. The confining layer is immediately above this and has a thickness of 10 m, the upper aquifer extends

to the surface.

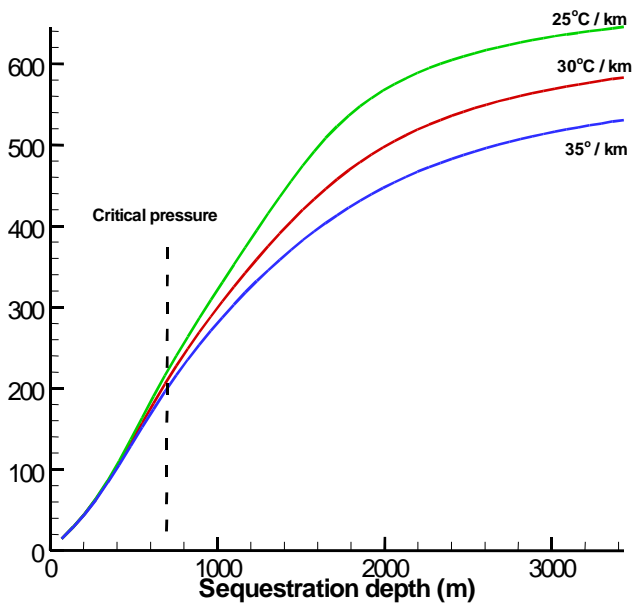


Figure 2: Density (kg/m^3) of CO_2 as a function of depth for different geothermal temperature gradients.

The permeabilities chosen for these aquifers are selected by considering typical cases from the petroleum and geothermal industries. Generally, useful horizontal permeabilities are found to be of the order of 100 mD, although local variations can be very great. At the other extreme, permeabilities of the order of 0.01 mD are associated with cap rocks or confining layers. To allow some exploration of different regimes, we have selected two values of 0.01 mD and 1 mD for the (isotropic) permeability of the confining aquifer. The permeability in the upper aquifer is taken as 10 mD in both the horizontal and vertical directions, and the permeability of the lower aquifer (where injection occurs) is isotropic and has a value of 100 mD. These permeabilities are summarised in Table 1. Corey (1977) relative permeability functions are used, with residual gas and liquid saturations of 0.05 and 0.3 respectively. Porosity is taken to be 0.1 in the upper and lower reservoirs, and 0.04 in the confining layer.

The variation of capillary pressure as a function of saturation for scenario C is taken from Vavra *et al.* (1992) and is discussed in more detail in Weir *et al.* (1996b).

The last major parameter to consider is the injection rate of CO_2 . There are several ways in which this might be determined, but again we can draw on experience from the petroleum industry. Hendriks and Blok (1994) consider injection of CO_2 into depleted natural gas reservoirs, and suggest that a rate of 10 kg/s per well is appropriate. Our experience with liquid reinjection in geothermal fields suggests a rather higher figure, perhaps as high as 100 kg/s per well might be practical. 100 kg/s is also typical of the waste CO_2 from a 1000 MW thermal power station, and for this reason we have adopted this figure.

The fourth simulation reported here (scenario D) was chosen to illustrate the modeling of the third storage mechanism, mineral sequestration. The physical conditions for this simulation actually represent a degassing magmatic intrusion and are somewhat different than those found at depth on the Colorado Plateau. The scenario has been included in this paper to illustrate the capabilities of CHEMTOUGH2 for modeling complex flow and chemical situations.

We have again adopted a cylindrical symmetry for the model. The cylinder has a radius of two kilometers, and extends from the surface to two kilometers depth (Figure 3).

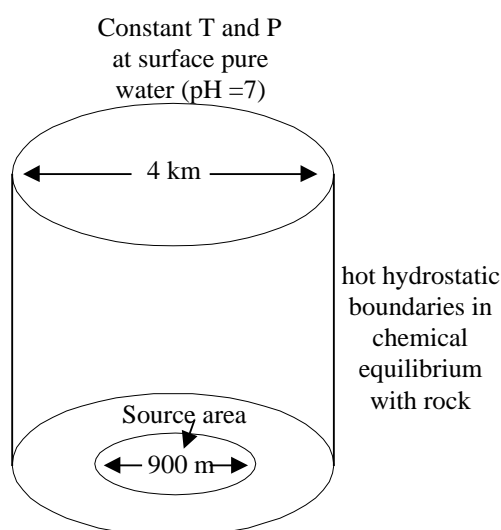


Figure 3: Layout and boundary conditions for model.

At the surface we assume water containing no dissolved solids or gases, a pH of 7, a constant temperature of 20°C and a pressure of one bar. On the vertical boundary of the model we assume hydrostatic pressures and temperatures consistent with a temperature gradient of 40°C/km. Fluid on the boundaries is in equilibrium with the rocks which are assumed to make up the unaltered reservoir. No fluid or heat flow is possible across the base of the model except over a central circular region of radius 450 meters at the base where CO₂ and high temperature water enter the modeled region from the degassing intrusion. The temperature in this region is about 300°C.

The upper 200 meters of the reservoir forms a partial cap and has a permeability of 0.01 mD. The rest of the reservoir has a permeability of 1.0 mD and porosity throughout is 0.1.

Modeling the transport of reactive chemicals is a computer intensive activity, and requires that a balance be struck between chemical complexity and calculation time. For this initial model, we adopted a simplified subset of reservoir component species, including H₂O, H⁺, Cl⁻, HCO₃⁻, SiO₂, Al⁺⁺⁺, Ca⁺⁺, K⁺ and Na⁺. These fluid components allow the modeling of reactions between the CO₂ and the most common rock-forming minerals (albite, anorthite, K-feldspar and quartz). Twenty-six of the most prevalent secondary aqueous species and four alteration minerals are also considered (Table 3).

Simplifying assumptions adopted in the treatment are:

- chemical equilibrium is maintained;
- the reacting minerals exert no direct redox control on the system; and
- reservoir permeability is isotropic (fracture flow is ignored).

Each of these assumptions, of course, represents a significant departure from natural hydrothermal systems. We believe the assumption of chemical equilibrium to be justified in the hot area of the reservoir, this assumption is certainly invalid in the cool areas, for example, the pH in the cool surface waters is much higher than normally found, but is correct for water in equilibrium with the assumed rock assemblage at 20°C. The assumption of chemical equilibrium will almost certainly be incorrect for CO₂ sequestration simulations where water rock interactions will need to be treated as kinetic rather than equilibrium reactions. (See White and Mroczek (1998) for an example of modeling water rock reactions kinetically.)

Name	Composition	Weight %
Albite	NaAlSi ₃ O ₈	40
Anorthite	CaAl ₂ Si ₂ O ₈	40
K-Feldspar	KAlSi ₆ O ₈	6
Quartz	SiO ₂	14

Table2: Initial rock composition

Name	Composition
Calcite	CaCO ₃
Kaolinite	Al ₂ Si ₂ O ₅ (OH) ₄
Muscovite	KAl ₂ (AlSi ₃ O ₁₀)(OH) ₂
Pyrophyllite	Al ₂ Si ₄ O ₁₀ (OH) ₂

Table 3: Rock alteration products included

Results

Reservoir Sequestration

Scenarios A-C began with an initial hydrostatic steady state pressure profile corresponding to the geophysical temperature gradient at 30° C/km. At the top boundary, 'atmospheric' conditions of 20°C, 1 bar were adopted. At the lower boundary the temperature was fixed at 113°C. Only heat flow (no mass flow) was permitted across this boundary. In the final steady state, the pressure at the lower boundary was about 324 bars. The steady state was achieved by running the model to a simulation time of 10¹³ seconds, by which time only small changes due to heat conduction were occurring. These are of small amplitude and have an associated timescale much longer than the 5000 year period we are modeling, and so can be safely neglected.

In all of the simulations, 100 kg/s of CO₂ was injected into the lower aquifer for a period of 10 years. Injection was assumed to be at a point located at a depth of 50 m below the confining layer. The simulations were continued to a total time of 5000 years in order to examine the consequences of the CO₂ injection. In particular, we are interested in the amount of CO₂ (if any) which might escape to the atmosphere, and also the distribution of CO₂ remaining in the aquifers.

Figure 4 shows the development of a single-phase bubble of CO₂ gas about the injection point. While this figure shows the results for scenario A, there is little difference between scenarios (A-C) during the early stages of the simulation (while gas is being injected). The iso-surface shown encloses the gas bubble and the color shading indicates gas saturation. This figure illustrates that a much larger 2-phase region surrounds the single-phase gas region.

The bubble has reached the bottom of the confining layer (double white lines) after 3.9 years, after which time it begins to spread laterally. In scenario B there is a greater loss of gas through the confining layer than in scenario A or C. After injection ceases the bubble collapses rapidly. In scenarios A and B much of the gas rises due to buoyancy forces forming a two-phase zone in the upper aquifer with no CO₂ in the gas phase trapped in the lower aquifer. In scenario C a significant amount of gas remains trapped beneath the confining structure. Figure 5 shows this two-phase region for scenario B after about 30 years. As can be seen, the gas has almost reached the surface, and after 100 years this two-phase region reaches to the surface and gas escapes to the atmosphere.

Neither of the simulations of scenario A or B indicates that a static single-phase gas zone forms under the confining layer after injection has ceased. This is because capillarity has been ignored in these models. In particular, the vapor entry pressure (Corey, 1977) is therefore zero, by definition.

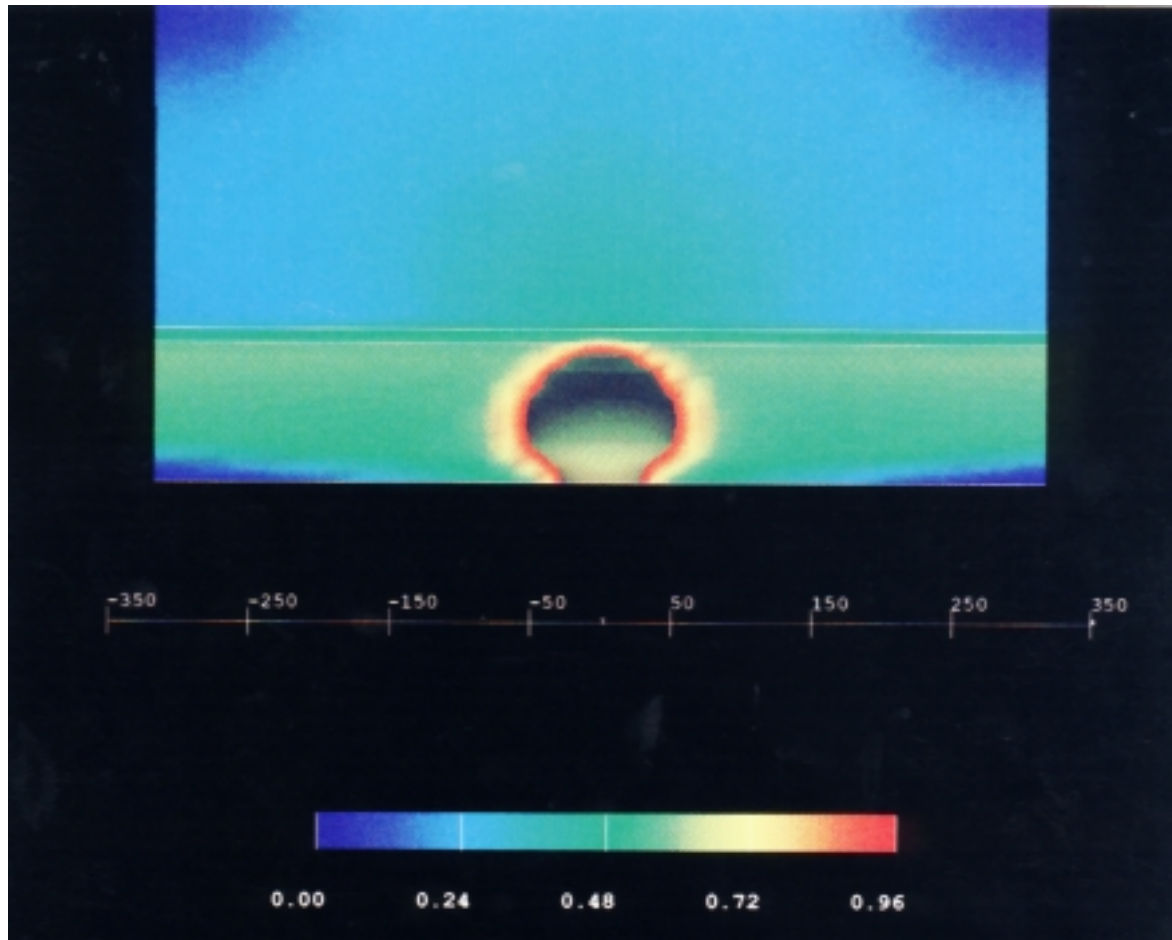


Figure. 4. Isosurface plot of 0.96 gas saturation, after 4 years for scenario A. The parallel white lines indicate the confining layer. The horizontal distance scale (in meters) is given below the plot.

The evolution of the two-phase region can be explained by gas moving upwards buoyantly, and becoming dissolved in unsaturated liquid. This causes the isosurface to contract at its horizontal extremities, and extend vertically upwards in the central region. The liquid saturated in CO₂ is slightly denser than unsaturated fluid and as the two-phase region rises the gas is 'washed' from it and carried back to the lower aquifer.

The time-dependence of the spatial distribution of the total mass of injected carbon dioxide is shown in Figures 6, 7 and 8. The three curves in each figure represent the percentage of gas injected to that time in the lower and upper aquifers, and escaping to the atmosphere for each scenario.

Figure 8 shows that no gas escapes to the atmosphere for scenario A or C but does in scenario B. The percentage of injected gas stored in the lower aquifer initially decreases with time for all scenarios, due to escape of gas up through the confining layer in the vapor phase. However, after several thousand years, the percentage stored in the lower aquifer begins to increase as the denser liquid containing the carbon dioxide begins to flow back down through the confining layer. About 12% of the injected gas escapes to the atmosphere for scenario B. At the end of injection roughly equal amounts are stored in the upper and lower aquifers for scenarios A and B. In scenario C 90% of the gas is contained in the lower aquifer throughout the simulation.

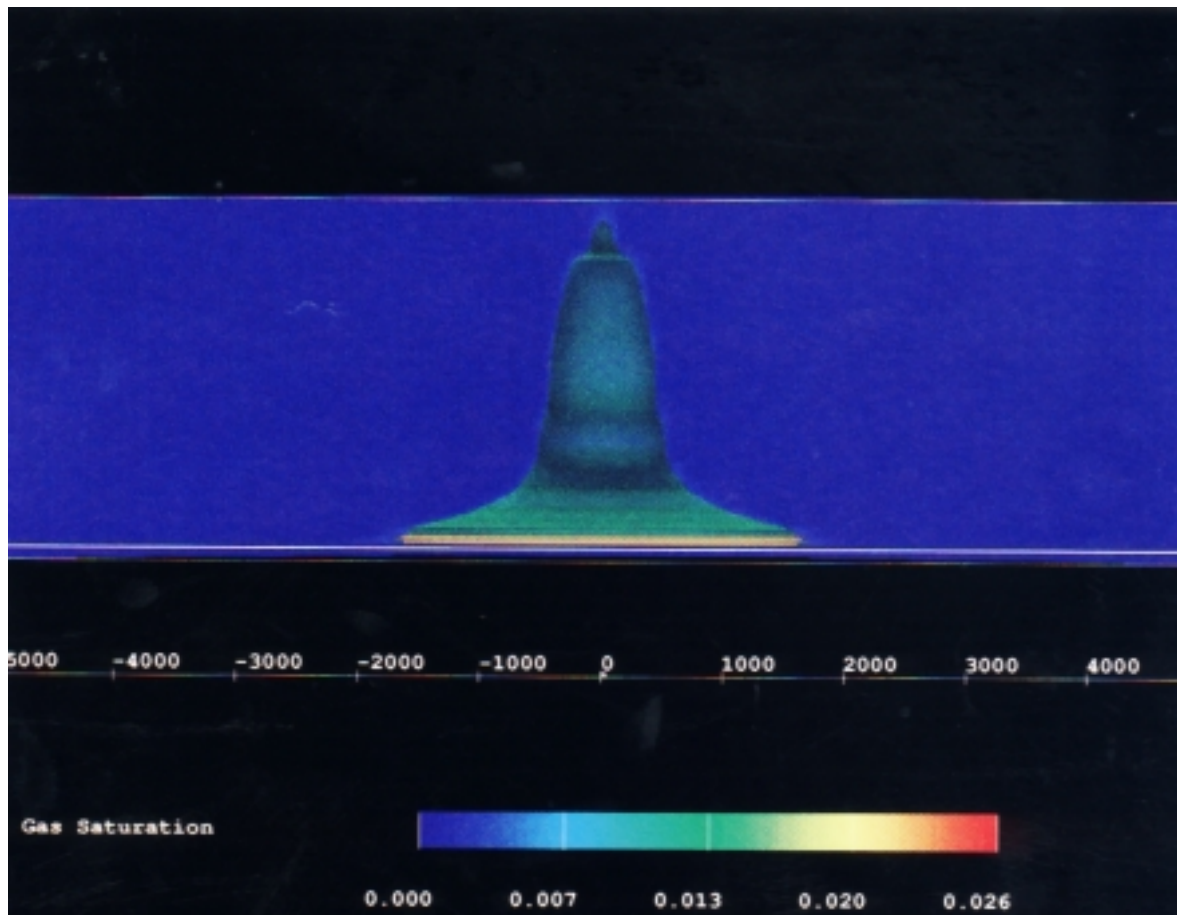


Figure 5: Isosurface plot of 0.025 gas saturation, after 31 years for scenario B. The parallel white lines indicate the confining layer. The horizontal distance scale (in meters) is given below the plot.

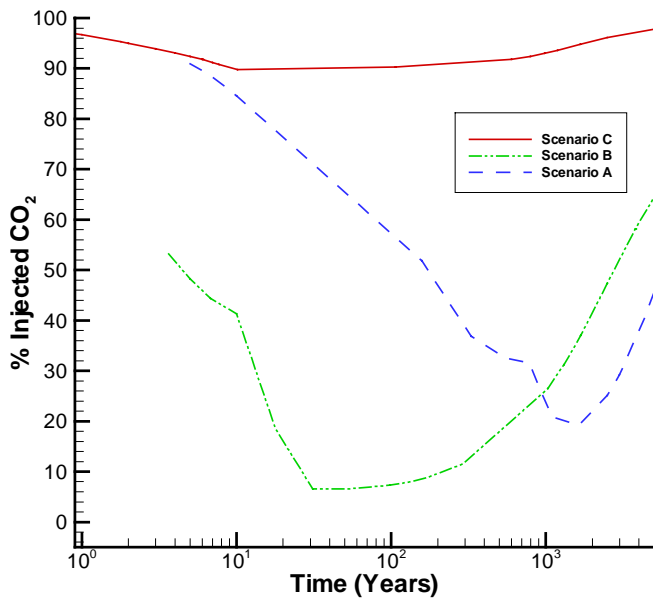


Figure 6: % of injected CO₂ in the lower aquifer for scenario A, B and C.

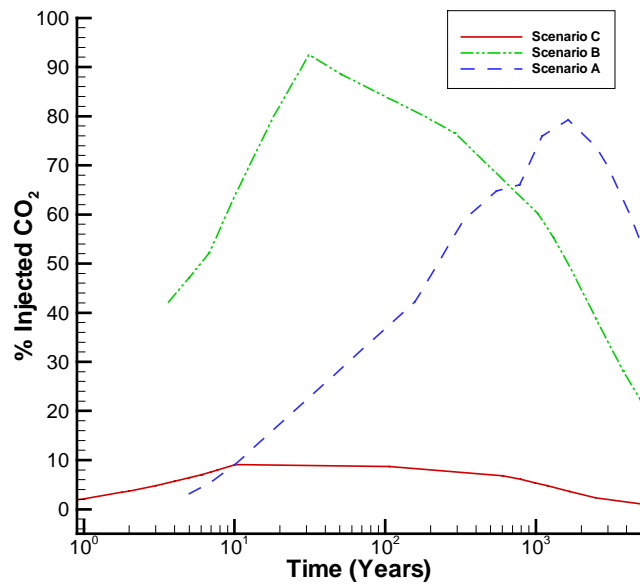


Figure 7: % of injected CO₂ in the upper aquifer for scenario A, B and C

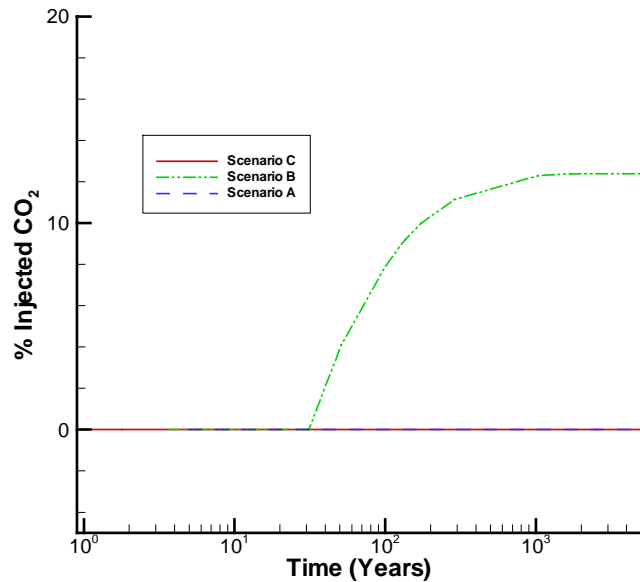


Figure 8: % of injected CO₂ in the upper aquifer for scenario A, B and C

Mineral Sequestration

The CO₂ component of the source fluid lowers the pH of the fluid through the formation of carbonic acid (H₂CO₃). This acidic fluid reacts with the reservoir rocks forming secondary carbonate minerals and clays. Gunter *et al.* (1993, 1997) suggests that siliclastic reservoirs are likely to be the most reactive type of reservoir, with aluminosilicate minerals (in this example feldspars) breaking down to form kaolinite and carbonates. This process is illustrated in Figure 9 which shows the reservoir mineralogy after interaction with a source fluid containing 3.2% CO₂ by weight over a long period of time. Initially K-feldspar is dissolved throughout the high CO₂ (low pH) plume formed above the source region and replaced by Muscovite. This is followed by the dissolution of Ca-feldspar with the precipitation of calcite and kaolinite.

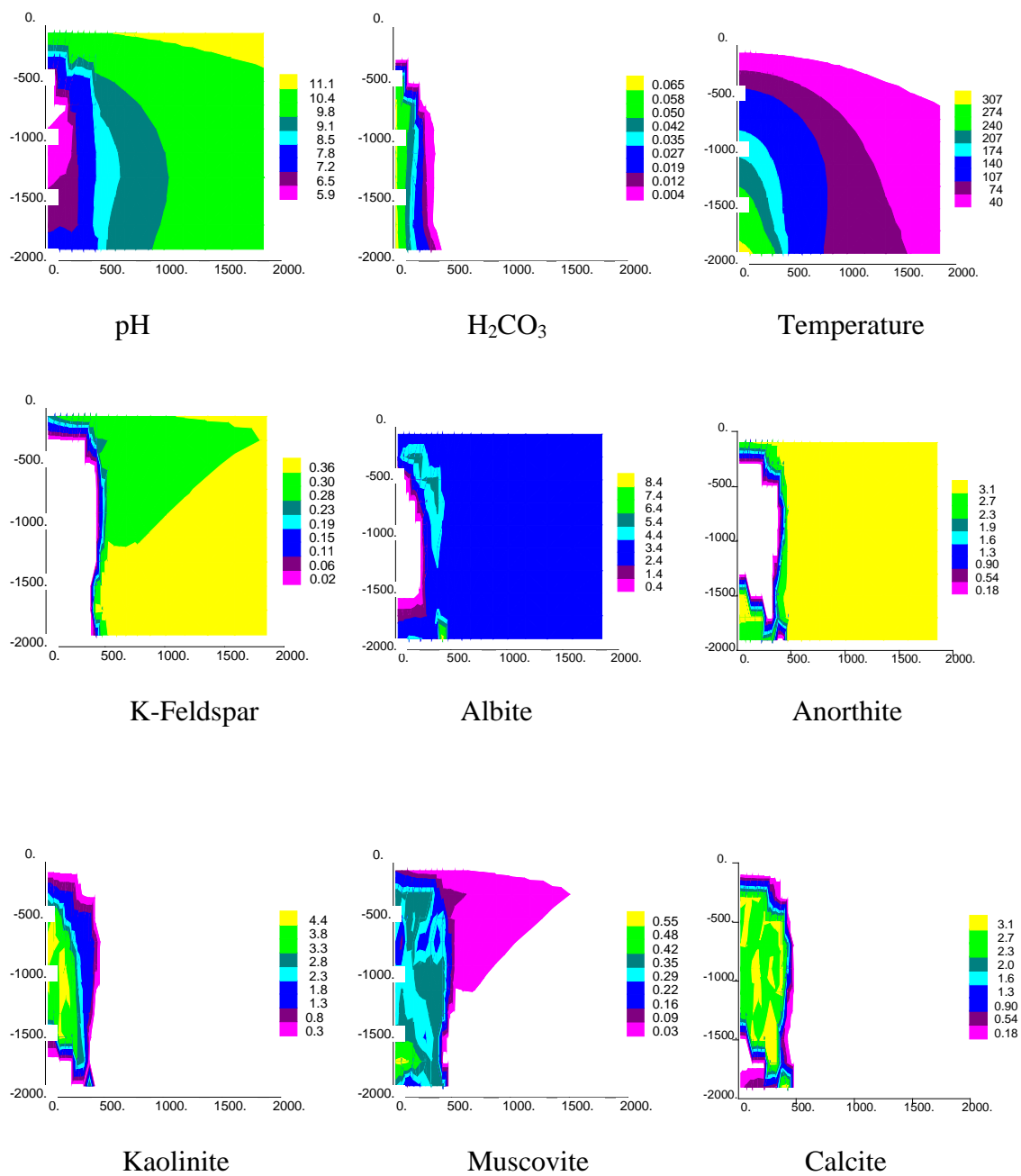


Figure 9: Mineral assemblage and some aqueous species concentrations after reaction between CO₂ rich reservoir fluid and initial reservoir rock. Aqueous concentrations are in Moles/kg and solid concentrations in Moles/liter of fluid.

Conclusions

Simulation of CO₂ injection into a saline aquifer provides a useful tool to determine the effectiveness of such an aquifer for long term sequestration. Providing the simulator is capable of modeling the transport of CO₂ in the liquid and gas phases and reactions between reservoir fluids and reservoir rocks then realistic estimates of the storage potential of such reservoirs should be obtained.

Further Work

Our research is just beginning. Currently we are developing a model of the Farnham dome region as a basis for modeling of reservoir and mineral sequestration of CO₂. This model contains a realistic (if somewhat simplified) representation of the geology of the region and will be used to test containment of injected CO₂.

Some of the important questions to be answered to complete this work are:

- What reaction rates and what reactions for the water rock interactions are appropriate?
- What reactive surface areas are appropriate for the reservoir?
- What porosity - permeability relationship is appropriate for the reservoir?
- What relative permeability and capillary pressure functions are appropriate for the reservoir?
- How can model predictions be tested?

Acknowledgements

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