



UPPSALA
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PROJEKTRAPPORT

CO₂ Sequestration in Saline Aquifers

Investigation of Appropriate Boundary Conditions in
Numerical Simulations

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Project in Computational Science: Report



1 Introduction

In recent years, CCS (carbon capture and storage) has become a highly topical field of research. The idea behind CCS is to capture industrial emissions of carbon dioxide and store it in supercritical form in underground, porous geological formations. Suitable formations could be depleted oil- or natural gas reservoirs, or saline aquifers. Storage in saline aquifers seems to be the most promising alternative, although more research in the area is needed [5].

There is an ongoing debate concerning the amount of carbon dioxide that can be stored in saline aquifers. In a recently published paper, *Sequestering Carbon Dioxide in a Closed Underground Volume* (2009) [1], Economides, et al. conclude that the storability is highly overestimated due to the application of erroneous boundary conditions. If this conclusion holds, CCS would be a less appealing option. The most commonly used boundary condition at the lateral boundary is the Dirichlet (constant pressure/open boundary) condition [6] [7]. Economides, et al. (2009) [1] suggest that the correct boundary condition at the lateral boundary is not a Dirichlet condition but rather a homogeneous Neumann (no flow/closed boundary) condition.

The choice of boundary condition seems to be a critical issue. To study how sensitive the solution is to boundary conditions, simulations are performed using both a Dirichlet condition and a homogeneous Neumann condition at the aquifer boundary, comparing the results. Usually, an aquifer is bounded by a media that has a lower permeability than the aquifer itself. The main aim of this research is to investigate which boundary condition that really models an aquifer surrounded by a large, homogeneous low-permeable media. Here large means large enough to ensure that the conditions beyond this media have no impact on the state of the aquifer. To do this, simulations including the media surrounding the aquifer are performed. The solution is compared to the solutions obtained when using a Dirichlet- and homogeneous Neumann condition at the aquifer boundary. It's also discussed what the solution means in terms of pressure buildup. Furthermore, the question if a Dirichlet condition models an infinite aquifer is addressed. This is a common interpretation [4], which is claimed to be incorrect by Economides, et al. (2009) [1].

The simulations are performed in *TOUGH2* with the *ECO2N* module. *TOUGH2* is a numerical simulator for non-isothermal flows of multicomponent multiphase fluids in porous and fractured media. It was originally developed in the Earth Sciences Division of Lawrence Berkeley National Laboratory [2] [8].

2 Numerical Simulations and Results

2.1 The Model problem

In the simulations the modeled aquifer is in the shape of a disc. The aquifer has a radius of 10 km, a height of 50 m and a permeability of 10^{-12} m^2 . It is located 1 km below sea level, where the carbon dioxide is in supercritical form. The carbon dioxide is injected with an injection rate of 100 kg/s through a well, located in the centre of the aquifer. When investigating which boundary condition that models an aquifer with a low-permeable surrounding media, this surrounding is added to the model aquifer. The media surrounding the

aquifer is assumed to be homogeneous and have a lower permeability than the aquifer, see Fig. 1. To show how the solution depends on the permeability in the surrounding media, the permeability is varied from 10^{-15} m^2 to 10^{-20} m^2 . The values of other parameters used in the simulations are given in appendix A.

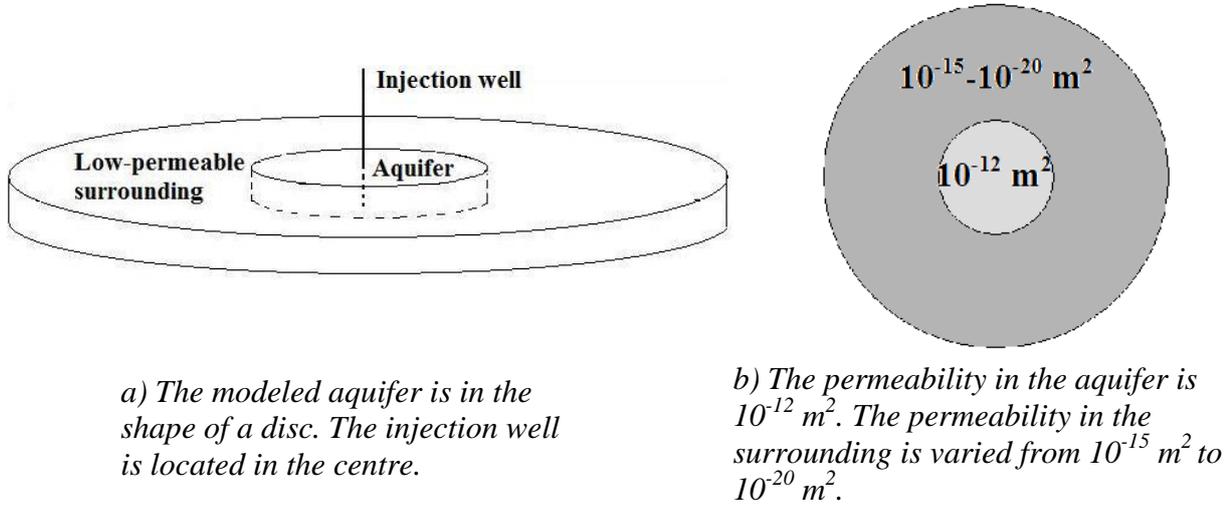


Fig. 1. The low-permeable surrounding is included in the model when investigating which boundary condition that models an aquifer surrounded by a homogenous low-permeable media.

If the surrounding media is large enough, the outer boundary of the domain does not affect the solution at the aquifer boundary. The size of the domain, required to fulfil this is investigated using different sizes of the low-permeable surrounding, simulating with both a closed and an open boundary condition. In these simulations a permeability of 10^{-15} m^2 is used in the surrounding media. The solutions at the aquifer boundary for the different boundary conditions are compared. If the outer boundary is adequately distanced, the solutions agree and the boundary condition used here has no relevance. For both cases, the pressure dependency of time at the aquifer boundary is shown in Fig. 2.

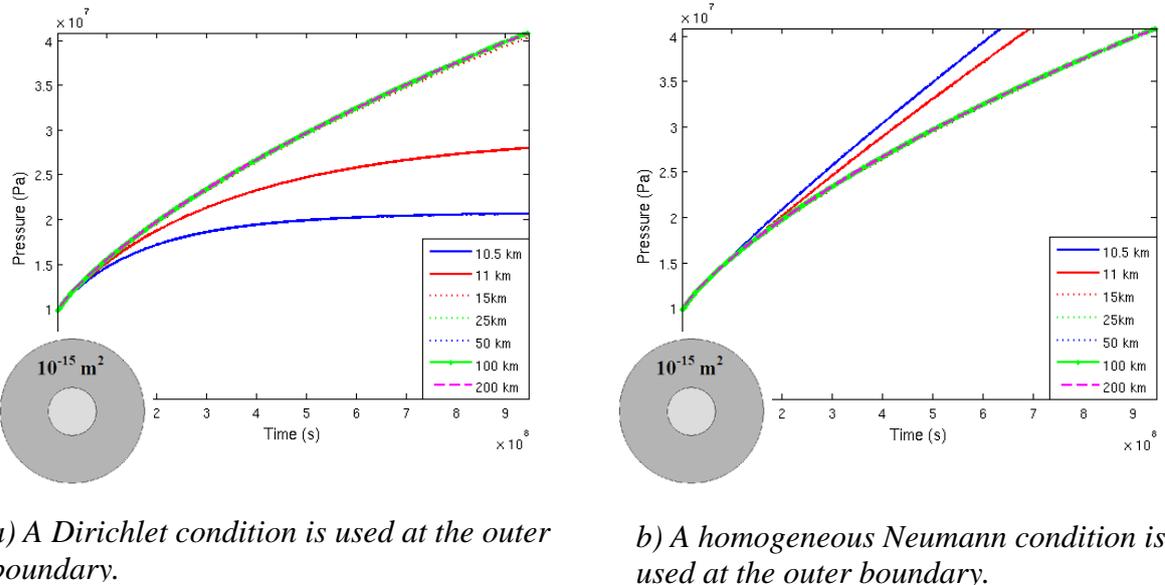


Fig. 2. The pressure dependency of time at the aquifer boundary is computed for different sizes of the surrounding. The permeability of the surrounding media is 10^{-15} m^2 . The solutions coincide if the outer boundary is more than 15 km away from the centre.

There is no difference between the solutions for an outer radius of 25 km to 200 km, and the boundary does not affect the results. For a radius of 15 km there is a slight difference, and the difference increases if the radius decreases further. The limit of 15 km is valid only if the permeability of the surrounding media is 10^{-15} m^2 . With a higher permeability the fluids and the pressure front moves faster and the outer boundary will be reached sooner. Therefore a higher permeability in the surrounding media requires a larger surrounding. A radius of 15 km is hence sufficiently large for permeabilities lower than 10^{-15} m^2 . A scaling of the governing equations is performed in order to generalize this limit of the domain size. By scaling simplified versions of these equations the following relation is obtained:

$$T \sim \frac{L^2 \mu_i \phi}{KP} \quad (1)$$

where T is the time when the outer boundary is beginning to have an impact (i.e. when the pressure front reaches the outer boundary). L is the distance between the aquifer boundary and the outer boundary and μ is the viscosity of the fluids. P is the pressure difference between the aquifer boundary and the outer boundary and K is the permeability of the surrounding. For a derivation of the scaling, see appendix B. From this relation one can see that if L^2/K is held constant the solutions are comparable (presupposing that the other variables in the relation are held constant). The relation between the impact of the outer boundary and the value of L^2/K is shown in Fig. 3. The impact is measured in the l^2 -norm of the difference between the pressure developments at the aquifer boundary using different boundary conditions. For a permeability of 10^{-15} m^2 in the surrounding, this corresponds to calculating the difference between the curves in Fig. 2a and Fig. 2b for the different domain sizes respectively.

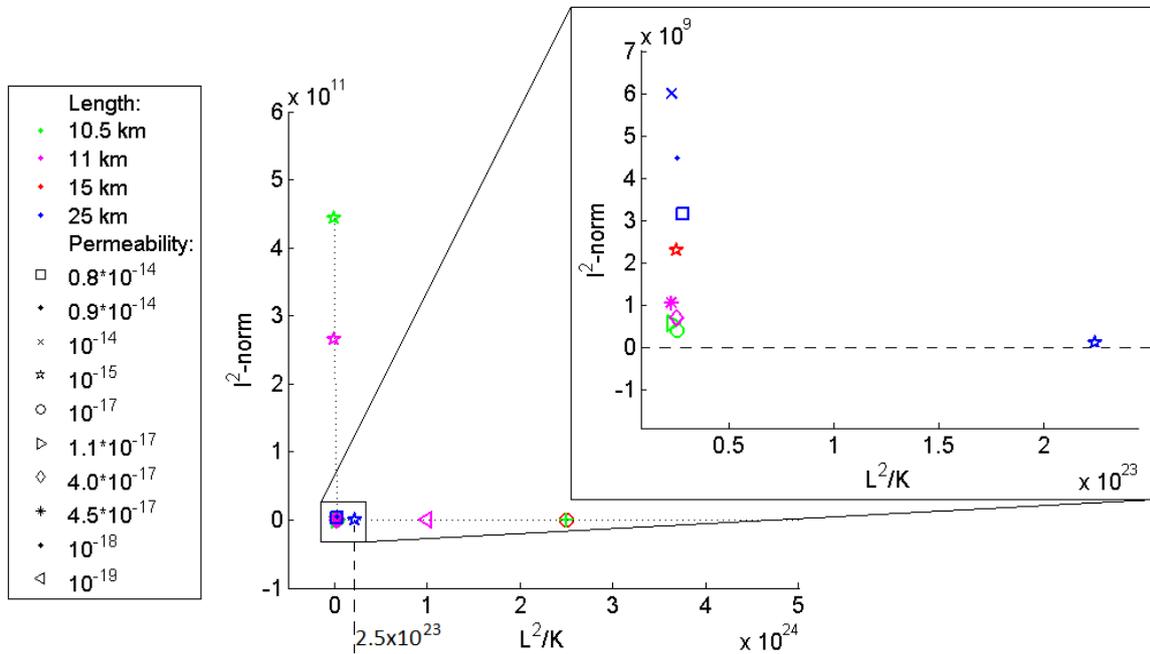


Fig. 3. The impact of the outer boundary is measured as the l^2 -norm of the difference between the solutions retrieved using the two different boundary conditions. There is a sharp limit where the error decreases rapidly. At this limit L^2/K is approximately $2.5 \cdot 10^{22}$. A value of the quotient L^2/K larger than this value means that the error is sufficiently small and that the outer boundary does not have an impact on the solution in the aquifer.

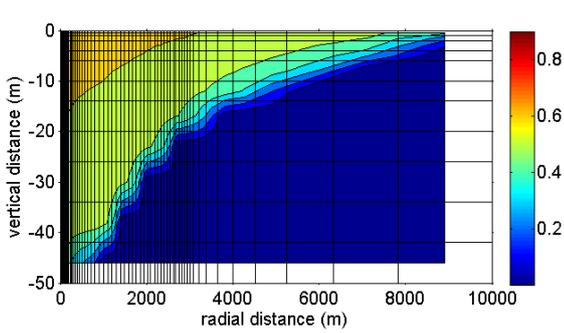
There is a sharp limit where the error decreases rapidly. At this limit L^2/K is approximately $2.5 \cdot 10^{22}$. A larger value of L^2/K implies that the outer boundary has no effect on the solution in the aquifer. Note that this limit is different if parameters affecting the pressure front (injection rate, aquifer size, number of injection wells, permeability in the aquifer etc.) are changed. In the rest of this study, the outer boundary will be 100 km from the centre, which ensures that the outer boundary has no impact for the permeabilities used. All simulations are performed using the constant pressure condition at the outer boundary.

2.2 An Aquifer with a Low-permeable Surrounding Resembles a Closed System

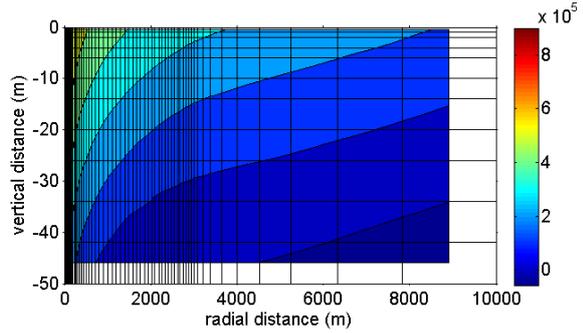
There is a significant difference between the solutions obtained when using a Dirichlet condition at the aquifer boundary, compared to using a homogeneous Neumann condition (Fig. 4a-d). Using a Dirichlet condition, the carbon dioxide plume reaches further than it does when using a homogeneous Neumann condition. Also, the pressure buildup is much greater when using a closed boundary. The solution in the aquifer thus seems to be highly sensitive to the boundary condition used in simulations, and it is an important question finding an appropriate condition.

In Fig. 2 the curves corresponding to an outer radius larger than 15 km show that the pressure is increasing with time at the aquifer boundary. According to the governing equations (see *TOUGH2 user's guide* [2]), steady state will eventually be established and the pressure increase level out. For a time scale of 30 years (used in the simulations) steady state obviously never occurs. This indicates that the commonly used Dirichlet condition is an inappropriate boundary condition.

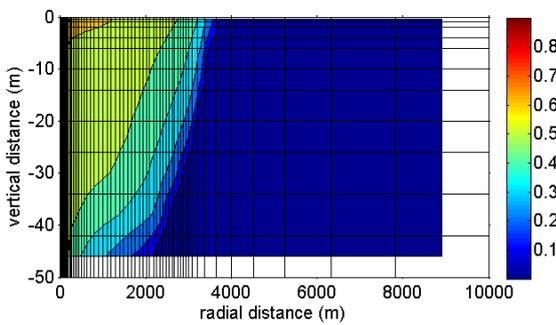
In fact, simulations including the surrounding media show that when the aquifer is surrounded by a homogeneous low-permeable media, the carbon dioxide plume is more similar to the plume received when a closed boundary is used, than when an open boundary is used (compare Fig 4e and Fig 4c and 4a). Also, the pressure buildup has the same order of magnitude as when a closed boundary condition is used.



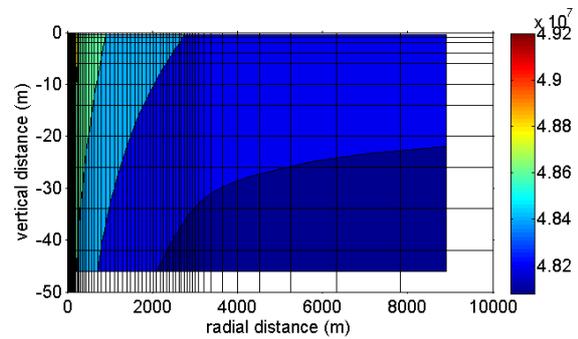
a) Saturation of supercritical carbon dioxide when a Dirichlet boundary condition is used.



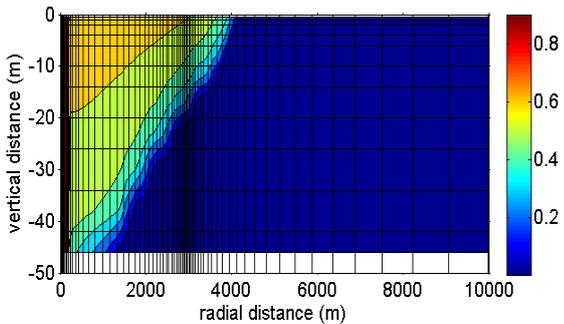
b) Pressure buildup when a Dirichlet boundary condition is used.



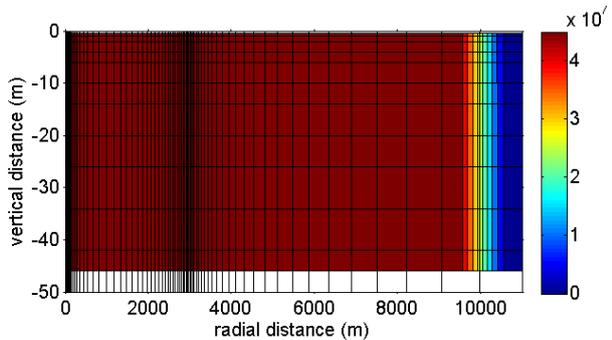
c) Saturation of supercritical carbon dioxide when a homogeneous Neumann boundary condition is used.



d) Pressure buildup when a homogeneous Neumann boundary condition is used.



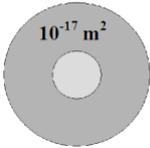
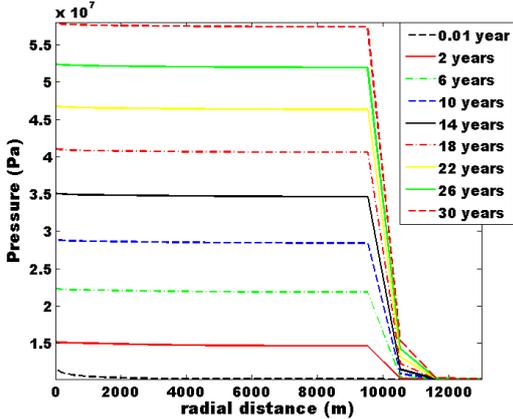
e) Saturation of supercritical carbon dioxide when the surrounding media is included.



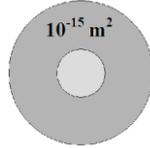
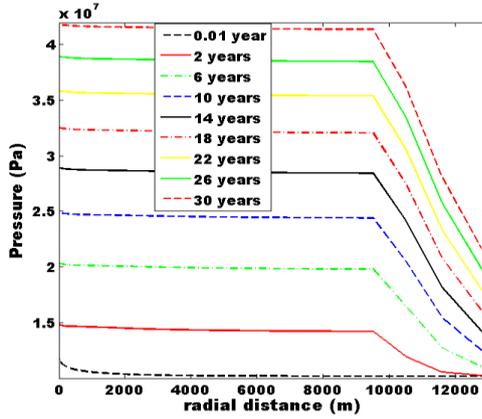
f) Pressure buildup when the surrounding media is included.

Fig. 4. Saturation of supercritical carbon dioxide and pressure buildup after 30 years of injection. The carbon dioxide plume has different shapes using a Dirichlet condition compared to when using a Neumann condition. When including a surrounding with a permeability of 10^{-17} m^2 the plume of CO_2 has a similar shape as when a closed boundary is used. Note that the pressure difference is largest in the upper part of the aquifer. Therefore all of the following pressure distribution graphs show the pressure at the top of the aquifer.

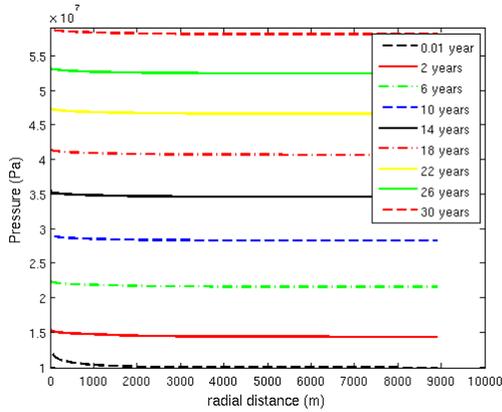
The pressure buildup in the aquifer is a limitation of the storability of the carbon dioxide. The pressure distribution in the aquifer is therefore studied for different injection times. The results obtained using a surrounding media with different permeabilites is compared to the results obtained using a Dirichlet- and a homogeneous Neumann condition at the aquifer boundary, see Fig. 5. There is a striking similarity between the pressure profiles when the aquifer is surrounded by a low-permeable media and the profiles obtained using a closed boundary condition. In both cases, there is a large pressure buildup, as already indicated in Fig. 4d and Fig. 4f.



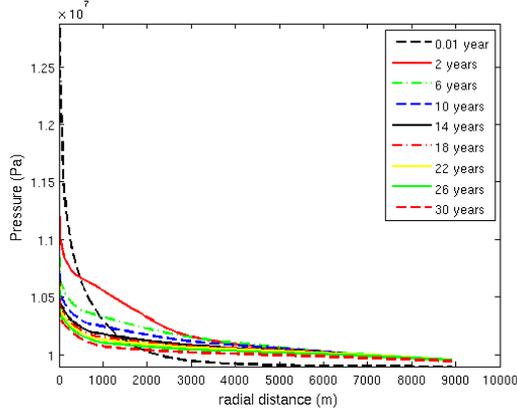
a) The permeability is 10^{-17} m^2 in the surrounding media.



b) The permeability is 10^{-15} m^2 in the surrounding media.



c) A homogenous Neumann condition is used at the aquifer boundary.



d) A Dirichlet condition is used at the aquifer boundary.

Fig. 5. Pressure profiles along the top of the aquifer. Including a low-permeable surrounding media of the aquifer results in a pressure profile similar to the one retrieved when using a homogeneous Neumann condition at the aquifer boundary.

A permeability of 10^{-15} m^2 outside the aquifer leads to a somewhat lower pressure buildup than when the permeability is 10^{-17} m^2 outside. Also, the pressure decrease is not as sharp across the boundary. The pressure buildup in the aquifer is a limitation of the storability because the surrounding rock will fracture if the pressure buildup is too high. How much the pressure is allowed to increase is highly dependent on the aquifer in consideration. In an article by Zhou et al. (2008) [3] a sustainable pressure buildup corresponding to 50 % of the initial pressure is used. In this research, that corresponds to a maximum tolerated pressure of approximately 15 MPa. As Fig. 5a-b shows, this pressure is reached already after two years of injection. The most commonly used boundary condition, the Dirichlet condition, leads to an insignificant pressure buildup and the maximum pressure is never reached. The usage of a Dirichlet condition, when simulating an aquifer with a low permeable surrounding, thus leads to an overestimation of the storability, if the pressure buildup is the only limiting factor. This coincides with the conclusions of Economides, et al. (2009) [1]. When using a Dirichlet condition, the storability is in practice limited by other factors such as leakage of brine.

The results shown in Fig. 4 and Fig. 5 both lead to the conclusion that an aquifer surrounded by a low-permeable media resembles a closed system more than an open system. However, there are some differences. The plume shown in Fig. 4a reaches slightly further into the aquifer than it does using a closed boundary condition and the pressure buildup in Fig 5a-b is not quite as high as in the closed case. This indicates that brine leaks out of the aquifer and that the real boundary condition might be a semi-closed condition. To investigate what the boundary condition is, the pressure and the pressure gradient at this boundary are calculated, using three different permeabilities in the surrounding media. The dependencies between the pressure gradient and the pressure at the boundary are shown in Fig. 6.

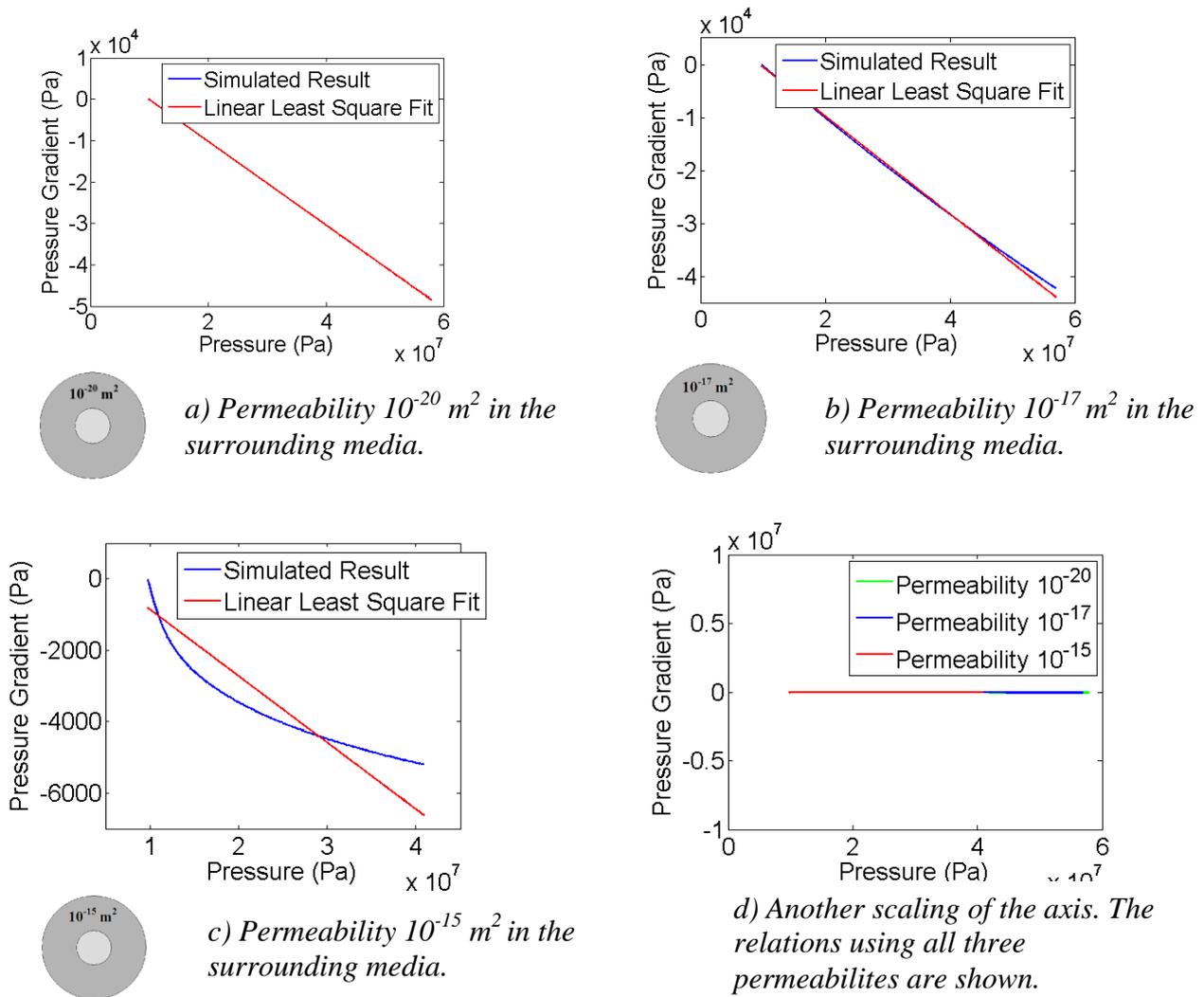
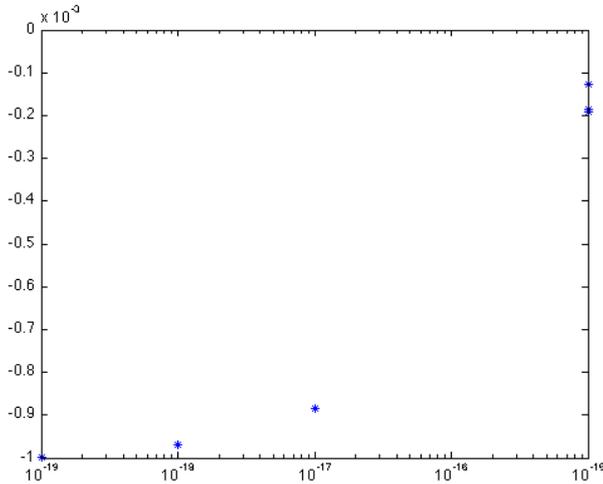


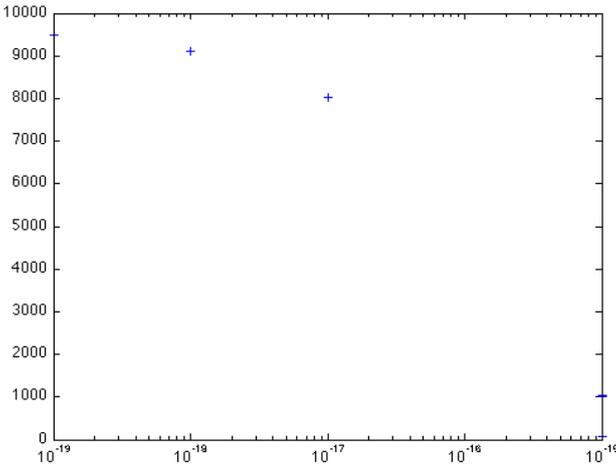
Fig. 6. The pressure gradient varies as the pressure increases at the aquifer boundary. For relatively low permeabilities in the surrounding media the dependency is approximately linear. For a higher value on the permeability in the surrounding media the relation is nonlinear. However, if the values on the axes are scaled to the same order of magnitude it becomes evident that the relation is nearly a homogeneous Neumann condition.

Fig. 6a-c shows that the pressure gradient is slightly dependent on the pressure, which corresponds to a semi-closed condition. When the permeability of the surrounding is 10^{-17} m^2 and 10^{-20} m^2 , the relationship between the pressure gradient and the pressure is approximately linear (i.e. $\nabla p = ap + b$, where a and b are constants). Simulations with a higher permeability in the surrounding media (Fig. 6c) show a clearly nonlinear relationship between the pressure gradient and the pressure. This is due to nonlinearities in the equations, which have a greater impact when the surrounding media has a high permeability. In Fig. 6d the values on the axes are scaled to the same order of magnitude. The figure shows that the pressure gradient is very small compared to the pressure in the aquifer. Hence, setting the pressure gradient to zero is not a rough approximation when concerning the pressure distribution. This corresponds to a homogeneous Neumann condition.

The maximum pressure is not always the main focus of all researches. If the leakage of brine is of interest, a no flow condition is obviously not an appropriate approximation. Nevertheless, it is even worse to use a Dirichlet condition. When investigating leakage one should try to implement a semi-closed boundary condition corresponding to the type of relations shown in Fig. 6a-c. This implementation is complicated since many parameters could affect the conditions at the boundary. As seen in Fig. 6 the condition at the aquifer boundary varies with the permeability in the surrounding. In Fig. 8 it is shown how the coefficients a and b in the linear least square fit dependency of the pressure gradient and the pressure at the aquifer boundary ($\nabla p = ap + b$) varies with the different permeabilities. It is seen that a lower permeability leads to a smaller absolute value of the coefficient a and b . This means that the relation is more similar to a homogenous Neumann condition if the permeability is low outside the aquifer. The conditions at the aquifer boundary may be affected by other parameters as well, such as porosity, compressibility, number of injection wells, aquifer size, injection rate and the temperature. The solution is especially sensitive to the rock compressibility, which affects the pressure distribution just as much as the permeability of the surrounding.



a) The value of the coefficient a varies with the permeability in the surrounding.



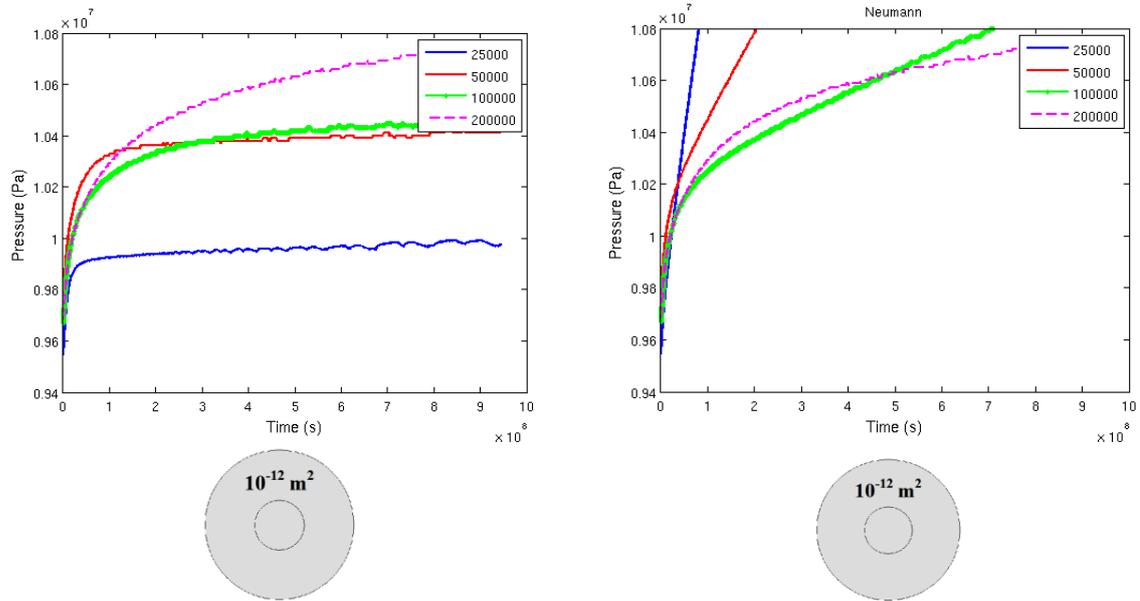
b) The value of the coefficient b varies with the permeability in the surrounding.

Fig. 8. The absolute values of the coefficients a and b , in the linear least square dependency of the pressure gradient and the pressure, decreases if the permeability increases in the surrounding.

2.3 A Dirichlet Condition is Not Modeling an Infinite Aquifer

The homogeneous Neumann boundary condition seems to be a much better approximation of the boundary than the commonly used Dirichlet condition when the aquifer is surrounded by a low-permeable media, as Economides, et al. (2009) [1] also concludes. Economides, et al. (2009) [1] also state that the constant pressure boundary has been mistaken to mimic an effectively infinite aquifer [4]. To investigate if a Dirichlet condition really simulates an infinite aquifer, the permeability is now set to 10^{-12} m^2 in the surrounding as well as in the aquifer. Since the permeability in the surrounding is higher than 10^{-15} m^2 it is possible that the outer boundary has to be further away than 100 km from the injection well as not to affect the result in the aquifer. To find out how far away the outer boundary has to be, the scaling relation (1) is used. The outer boundary has no impact when L^2/K is larger than about $2.5 \cdot 10^{22}$. With a permeability of 10^{-12} m^2 , L should be larger than approximately 160 km. Since L is the distance between the outer boundary and the aquifer boundary, and the aquifer has a radius of 10 km, the outer radius should be about 170 km. To verify this estimate, an investigation similar to the one in the previous section is performed. The pressure dependency on time at a radius of 10 km is calculated for different sizes of the surrounding media. In Fig. 7a a Dirichlet condition is used at the outer boundary and in Fig. 7b a homogeneous Neumann condition is used. In both cases, domains with a radius from 25 km up to 200 km are modeled. Using a radius of 200 km, the pressure dependencies are the same in both Fig. 7a and Fig. 7b. Hence, as the scaling also implies, the solution is not affected by the outer boundary using a surrounding of this size or larger and the aquifer can thus be considered to be infinite.

The pressure curve in Fig. 7 corresponding to an outer radius of 200 km also shows that the solution does not reach steady state during the 30 years of injection. Consequently the pressure at the imagined domain boundary, at 10 km from the injection well, is not constant and the condition at this boundary does not correspond to a Dirichlet condition.



a) A Dirichlet condition is used at the outer boundary.

b) A homogeneous Neumann condition is used at the outer boundary.

Figure 7. The pressure dependency on time at an imagined domain boundary, located 10 km from the injection well, is computed for different sizes of the domain. The permeability is 10^{-12} m^2 in the whole domain. The solutions agree if the outer boundary is more than 200 km away from the centre.

3 Conclusions

Simulations including a low-permeable area outside the aquifer – large enough to ensure that the conditions beyond this is of no importance – show that the pressure at the aquifer boundary is not constant, in fact it is increasing in time. This indicates that a Dirichlet condition is not an appropriate boundary condition. The actual boundary condition is a semi-closed, nonlinear condition. However, the pressure gradient at the aquifer boundary is very small compared to the pressure in the aquifer. Hence, a homogeneous Neumann condition is a fairly good approximation when considering the pressure distribution. The pressure is however not always the main focus of all investigations. If leakage of brine is the main consideration, it is of course not appropriate to use a homogeneous Neumann (no flow) condition. For such cases, it would be interesting to implement the nonlinear semi-closed condition.

The storability of the carbon dioxide is limited by the pressure buildup in the aquifer. When the aquifer is surrounded by a low-permeable media, the pressure buildup is similar to the buildup in a closed system. The maximum allowed pressure is reached after only a few years of injection. In contrast, there is an insignificant pressure buildup when using the Dirichlet condition. In this case, the maximum pressure is never reached. Consequently, using a

Dirichlet condition when simulating an aquifer with a low-permeable surrounding media lead to an overestimation of the storability if the pressure buildup is the only limiting factor. This coincides with the conclusions of Economides, et al. (2009) [1].

Economides, et al. (2009) [1] also state that the constant pressure boundary has been mistaken to mimic an effectively infinite aquifer. Simulations show that the pressure at an imagined boundary inside a large aquifer is not constant in time and thence the condition there does not correspond to a Dirichlet condition.

To summarize, an aquifer surrounded by a homogeneous, low-permeable media – large enough to ensure that the conditions beyond this media have no impact – shows far more similarities with a closed system than an open. From this research, it is evident that the boundary conditions used at the lateral boundaries are of great importance. The usage of a Dirichlet condition, when considering an aquifer with a large low-permeable surrounding, leads to a heavy overestimation of the amount of carbon dioxide that can be stored. This points out the importance of further research regarding the aquifer surroundings in order to find out if CCS is still a feasible method.

Acknowledgements

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Appendix A - Parameters

Table 1. Parameters used for simulations in TOUGH2.

Parameter	Value
Rock grain density	2600 kg/m ³
Porosity of the aquifer	0.15
Porosity of the well area	0.18
Absolute permeability in the aquifer for all directions	1.0·10 ⁻¹² m ²
Formation heat conductivity	2.51 W/m°C
Rock grain specific heat	920 kJ/kg°C
Pore compressibility	4.5·10 ⁻¹⁰ m ² /N
Relative permeability function	Brooks-Corey-Burdine*
Capillary pressure function	Brooks-Corey
Initial pressure	1.55·10 ⁷ Pa
Injection rate	100 kg/s

* The Brooks-Corey-Burdine function for relative permeability is

$$k_{rl} = \bar{S}_l^{3+\frac{2}{\lambda_B}}$$

$$k_{rg} = \left(\frac{S_g - S_{gr}}{1 - S_{lr} - S_{gr}} \right)^2 \left(1 - \bar{S}_l^{1+\frac{2}{\lambda_B}} \right)$$

where

k_{rl} is the relative permeability for the liquid, wetting phase (the brine, which can contain solved carbon dioxide)

k_{rg} is the relative permeability for the non wetting phase (the carbon dioxide)

S_l is the saturation for the wetting phase and S_{lr} is the residual saturation for the wetting phase, i. e. the saturation when the relative permeability k_{rl} is zero.

S_g is the saturation of the non wetting phase and S_{gr} is the residual saturation for the non wetting phase

λ_B is the pore size distribution index which is set to 0.762 in the simulations

\bar{S}_l is the effective saturation, $\bar{S}_l = \frac{S_l - S_{lr}}{1 - S_{lr}}$

which by the Brooks-Corey function for capillary pressure is

$$\bar{S}_l = \left(\frac{P_d}{P_c} \right)^{\lambda_B}$$

where

P_c is the capillary pressure

P_a is the capillary pressure when S_1 is equal to 1.

Appendix B – Scaling of the equation

The scaling is based on a simplification of equations used by *TOUGH2*. The presence of NaCl is neglected, and so is the gravity. The fluids are assumed to be immiscible and the relative permeability function is for simplicity equal to the saturation, $k_{r,i} = S_i$.

$$\nabla \cdot \left(\frac{\rho_i S_i}{\mu_i} \mathbf{K} \cdot \nabla p \right) = \frac{\partial}{\partial t} (\phi \rho_i S_i) \quad i = c, w \quad (2)$$

S_i is the saturation, ρ_i is the density, μ_i is the viscosity, \mathbf{K} is the absolute permeability tensor, p is the pressure and ϕ is the porosity. The equation is assumed to be valid for the water (w) and the carbon dioxide (c) independently.

The low-permeable surrounding of the aquifer is considered for the scaling and therefore the permeability is constant in the whole domain. The porosity can in this approximation be treated as a constant value. The variables are scaled in the following way :

$$\begin{aligned} \rho_i &= R_i \tilde{\rho}_i \\ p &= p_0 + P \tilde{p} \\ r &= L \tilde{r} \\ t &= T \tilde{t} \\ \nabla &= \frac{1}{L} \tilde{\nabla} \\ \frac{\partial}{\partial t} &= \frac{1}{T} \frac{\partial}{\partial \tilde{t}} \end{aligned}$$

where all the tilde variables are scaled variables in the interval [0,1], r is the radius. The capital letters are representing a characteristic value for the problem. Substituting these variables into equation 2 leads to a scaled equation:

$$\frac{1}{L^2} \frac{R_i}{\mu_i} \mathbf{K} P \underbrace{\tilde{\nabla} \cdot (\tilde{\rho}_i S_i \tilde{\nabla} \tilde{p})}_{\mathcal{O}(1)} = \frac{R_i}{T} \underbrace{\frac{\partial}{\partial \tilde{t}} (\phi S_i)}_{\mathcal{O}(1)} \quad (3)$$

By identification from this equation the following relation is obtained.

$$T \sim \frac{L^2 \mu_i \phi}{K P} \quad (4)$$

This relation is validated by the fact that the right hand side has the unit seconds, and by simulations. If the only parameter varied is the permeability, K , in the surrounding media, the limiting radius, L , when the boundary starts to affect the solution can be retrieved. If then the

fraction L^2/K is constant for the (L,K)-pairs, then the scaling is valid. For different values of the permeability the following limits are obtained, see *Table 2*.

Table 2. The value of the permeability in the surrounding media gives a corresponding radius limit where the boundary starts to affect the solution. The fraction, L^2/K , is approximately constant, though.

L(K)	K	L^2/K
5000	$1.0 \cdot 10^{-15}$	$2.5 \cdot 10^{22}$
15000	$0.8 \cdot 10^{-14}$	$2.81 \cdot 10^{22}$
1000	$4.0 \cdot 10^{-17}$	$2.5 \cdot 10^{22}$
500	$1.0 \cdot 10^{-17}$	$2.5 \cdot 10^{22}$

The fraction L^2/K is as constant as one can expect from such a major simplification, therefore the scaling can be assumed to be valid.