Sequential Model Coupling for Feasibility Studies of CO₂ Storage in Deep Saline Aquifers

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Abstract — Sequential Model Coupling for Feasibility Studies of CO₂ Storage in Deep Saline Aquifers — Modelling carbon dioxide (CO₂) storage in saline aquifers on a reservoir scale is very demanding with respect to computational cost. In realistic scenarios, large heterogeneous geometries need to be described. In addition, the governing physical processes are very complex. The models need to take into account nonisothermal, multiphase, and compositional processes that occur during CO₂ storage. However, in most cases, it is not necessary to describe all the physical processes for the whole simulation time period. The processes which dominate during CO₂ storage operations vary over time. This time-dependent behaviour allows for the use of models of reduced/adapted complexity for the description of the dominant processes within a given timescale. It is shown that by coupling these models of reduced complexity, the model efficiency can be increased without neglecting the relevant phenomena. Sequential coupling of models thus represents a first step in the direction of increased model efficiency.
NOMENCLATURE

- $\kappa$: Component index, either H$_2$O or CO$_2$
- $\alpha$: Phase index, either brine phase (w) or CO$_2$-rich phase ($n$)
- $\phi$: Porosity (-)
- $p_w$: Phase density (kg/m$^3$)
- $\mu_a$: Phase viscosity (Pa·s)
- $k_{\alpha\beta}$: Relative permeability of phase $\alpha$ of the porous medium ($W/(m\cdot K)$)
- $h_{pm}$: Heat conductivity of the porous medium ($W/(m\cdot K)$)
- $\lambda_{dry}$: Porous media heat conductivity for $S_w = 0$ ($W/(m\cdot K)$)
- $\lambda_{sat}$: Porous media heat conductivity for $S_w = 1$ ($W/(m\cdot K)$)
- $\lambda_{BC}$: Brooks-Corey parameter
- $S_{\alpha}$: Phase saturation (-)
- $X_{\alpha}^e$: Mass fraction of the component $\kappa$ in the phase $\alpha$ (-)
- $X_{\kappa}^{eq}$: Mass fraction of the component $\kappa$ in case of thermodynamic equilibrium (-)
- $K$: Intrinsic permeability ($m^2$)
- $g$: Gravity (m/s$^2$)
- $D^\alpha_{\kappa, pm}$: Diffusion coefficient of the component $\kappa$ in the phase $\alpha$ in the porous medium ($m^2$/s)
- $p_{o\alpha}$: Phase pressure (Pa)
- $p_c$: Capillary pressure (Pa)
- $p_{BC}$: Brooks-Corey entry pressure (Pa)
- $u_{\alpha}$: Specific internal energy of phase $\alpha$ (J/kg)
- $h_{\alpha}$: Specific enthalpy of phase $\alpha$ (J/kg)
- $c_T$: Specific heat capacity of the rock (J/kg·K)
- $q_{\omega}, q_h, q_{ir}$: Source or sink term of phase $\alpha$, heat or component $\kappa$

INTRODUCTION

After more than ten years of research the CCS (Carbon Capture and Storage) technology is currently in the stage of large-scale demonstration projects. For the realisation of these projects, a responsible identification and characterisation of suitable CO$_2$ storage sites is crucial. In order to identify appropriate storage sites, detailed geological data is required. Apart from geological information, modelling tools are needed that are capable of describing complex hydraulic, thermal, geomechanical, and geochemical processes on a large range of spatial and temporal scales.

In recent years, different modelling tools have been developed which describe the thermal and hydraulic processes during CO$_2$ storage. Class et al. (2009) give an overview of existing simulators and discuss an intercomparison study by means of three benchmark problems related to CO$_2$ storage. The temporal scale of the benchmark examples was limited to a period of 50 years and the spatial scale did not exceed 10 km (i.e., reservoir scale).

For investigations on pressure build-up and brine migration, reservoir extensions on scales of more than 100 km need to be modelled. Besides the computational requirements with respect to model domain size, the simulation of CO$_2$ storage might need to cover large timescales depending on the problem that needs to be solved. With respect to the risks of CO$_2$ storage and sustainability of the storage, the fate of the injected CO$_2$ plume needs to be modelled over very long periods (thousands of years). Such large-scale and long-term simulations were recently addressed by a follow-up benchmark study (http://org.uib.no/cipr/Workshop/2009/CO2/Benchmark.htm).

Thus, the scales required for the simulation of realistic storage scenarios are already computationally demanding, even when the model concepts are restricted to flow and transport processes of the CO$_2$ in the reservoirs. However, geomechanical and geochemical processes can become very relevant in certain cases with respect to risk estimation. Such additional processes increase the number of unknowns and thus the computational costs enormously.

Most of the existing simulators are not capable of describing all the potentially relevant physical and/or chemical processes. In particular, when adding geochemistry to the flow and transport model concepts, the computation times for solving large-scale systems increase dramatically. Therefore, these models are often restricted to simplified geometries or small timescales.

One possibility to overcome this problem could be the coupling of different models. Provided that it is possible to identify distinctly dominant processes in subdomains or during certain time periods, it can be a good strategy to model only these processes for the given regions or times (Darcis et al., 2009).

This manuscript focuses particularly on the sequential coupling of models with the aim of efficiently simulating the short and long-term behaviour of a CO$_2$ plume in a storage reservoir. A detailed description of the different time periods that can be distinguished during and after the injection of CO$_2$ into a geologic reservoir is given in Section 1. A sequential coupling concept is presented in Section 2 and an idealised example application is described in Section 3. The results of the application example and two additional variations, taking into account the effects of heterogeneities and grid refinement, are presented and discussed in Section 4, followed by some concluding remarks and perspectives.

1 MOTIVATION FOR MODEL COUPLING

Several characteristic time periods can be distinguished during and, in particular, after the injection of CO$_2$ into a geologic reservoir (see Fig. 1). In the following, this is discussed for a hypothetical injection into a saline aquifer.
In the injection stage, the low mutual solubilities of brine and CO₂ lead to the formation of a multiphase flow system which is driven by large pressure gradients, strong viscous and buoyant forces. Close to the injection well, large changes in pressure increase the relevance of geomechanical processes (deformation). Changes in porosity and permeability or even enlargement of faults or generation of fractures in the caprock might occur. Alterations in hydraulic parameters might also occur due to salt precipitation close to the injection well. The injected CO₂-rich phase is generally undersaturated with respect to water (H₂O) under reservoir conditions. Thus, H₂O evaporates from the saline brine into the CO₂-rich phase, the concentration of the dissolved salts in the residual brine phase increases until finally salt precipitates. Due to the evaporation of H₂O a single-phase CO₂ region evolves in the vicinity of the well.

At early times, temperature changes can affect fluid and rock properties. These temperature changes can be caused by a downhole temperature of the injected CO₂, which differs from the reservoir temperature. Moreover, the Joule-Thompson effect leads to a cooling due to pressure lowering and expansion of CO₂ (Pruess, 2008). However, the large heat capacity of the rock matrix reduces the extent of these non-isothermal effects and due to the diffusive nature of heat transport, their importance reduces with time.

The cessation of the injection gives rise to a second characteristic time period. Advective flow due to viscous and buoyant forces slowly decreases. Thus, the relative importance of compositional effects gradually increases, i.e., dissolution of CO₂ in the brine phase, increase of the brine density, and downward fingering of the CO₂-rich brine. The growing surface of the CO₂-phase plume also causes a shift of the governing processes towards compositional effects. This time period is typically expected to be very long, lasting over tens to hundreds of years.

Eventually, multiphase flow processes come to halt. The plume of the CO₂-rich phase is immobile and gradually CO₂ dissolves into the brine. When considering larger time scales of hundreds to thousands of years, geochemical processes – even with slow kinetics – can become important and affect the properties of the aquifer including porosity and permeability. Since this could potentially trigger new or seal existing flow paths, geochemistry should be considered as interacting with hydraulics, at least in certain sub-domains of the reservoir.

In the modelling of large domains over long periods of time, it can be of substantial benefit to adapt the model complexity both in time and space as required. Sequential model coupling in time and spatial domain decomposition (Toselli and Widlund, 2005) need to be investigated with respect to their application for the simulation of CO₂ storage. While domain decomposition is not the subject of the present study, an approach and an application example for a sequential coupling is presented.

2 SEQUENTIAL MODEL COUPLING

Sequential coupling of models has previously been used for the simulation of contaminant spreading in the vadose zone by Class et al. (2008). The infiltration of a light nonaqueous-phase liquid (LNAPL) into the unsaturated zone, its spreading and transport in the soil air and in the groundwater, as well as a thermally enhanced remediation have been described with a series of three sequentially coupled models.

As discussed in the previous section, the processes which take place during CO₂ storage in saline aquifers can also be separated into different time periods. In the following, a sequential coupling concept for the simulation of the non-isothermal flow and transport processes which occur during CO₂ storage will be derived. The coupling of geomechanical and geochemical processes will be part of future work.

Two time periods are distinguished here:
- the injection period, and
- the post-injection period.

During the injection period, the CO₂-rich phase mainly spreads due to advective and buoyant forces. However, the flow behaviour is also influenced by temperature effects due to an injection temperature that may differ from the reservoir temperature or due to the Joule-Thompson effect. The dissolution and diffusion of CO₂ in the brine phase only plays a minor role for the short-term CO₂ distribution. Therefore,
the injection period will be described with a nonisothermal two-phase flow model \((2pni)\) model that neglects compositional effects.

In the post-injection period, dissolution, diffusion, and density-driven convection become more important while advection- and buoyancy-driven processes slowly decrease. The initial temperature profile slowly reestablishes due to heat conduction and ambient groundwater flow. Thus, the nonisothermal, two-phase flow model needs to be extended to account for compositional effects. This is realised by sequentially coupling a two-phase, two-component model \((2p2cni)\) to the \(2pni\) model.

Even though it is not part of this work, it is possible to distinguish an additional time period in which the long-term thermal effects are neglected, further reducing the complexity of the model.

### 2.1 Mathematical and Numerical Model

In the model of the first time period, \(i.e., \) the \(2pni\) model, the following mass balance equation is solved for both the wetting brine phase \((w)\) and the nonwetting CO2-rich phase \((n)\):

\[
\frac{\phi}{\alpha} \frac{\partial(S_{w,n})}{\partial t} - \text{div} \left( q_{\alpha} \frac{k_{\alpha}}{\mu_{\alpha}} X_{\alpha} K(\text{grad} P_{\alpha} - q_{\alpha} \mathbf{g}) \right) - q_{\alpha} = 0, \quad \alpha \in \{w,n\}. \tag{1}
\]

Above and in the sequel we use standard notation, Helmig et al. (1997), which is also explained in the nomenclature at the beginning of this paper. Additionally, the energy balance equation of the fluid-solid mixture is solved to describe the nonisothermal processes. Assuming local thermal equilibrium, the energy balance equation can be written as follows:

\[
\frac{\phi}{\alpha} \frac{\partial \left( \sum_{\alpha} q_{\alpha} u_{\alpha} X_{\alpha} S_{\alpha} \right)}{\partial t} + (1 - \phi) \frac{\partial(q_{\alpha} c_{\alpha} T)}{\partial t} - \sum_{\alpha} \text{div} \left( q_{\alpha} \frac{k_{\alpha}}{\mu_{\alpha}} X_{\alpha} K(\text{grad} P_{\alpha} - q_{\alpha} \mathbf{g}) \right) - \text{div} \left( \lambda_{\alpha} \text{grad} T \right) - q_{h} = 0 \tag{2}
\]

Where the summation is taken over the phases \(\alpha \in \{w,n\}\). To close this system of equations, the following auxiliary conditions are applied:

\[
\sum_{\alpha} S_{\alpha} = 1 \quad \text{and} \quad p_{w} = p_{n} - p_{s}(S_{w}) \tag{3}
\]

The model of the second time period \((2p2cni)\) model) solves compositional mass balances for the components CO2 and H2O. It is assumed that the CO2-rich phase consists of a maximum of two components namely CO2 and H2O, whereas the brine phase consists of the three components CO2, H2O and halite (NaCl). However, there is no mass balance for NaCl, since the mass fraction of NaCl is assumed to be constant and determined by the average salinity of the reservoir. The energy equation of the \(2p2cni\) model is similar to Equation (2), but the phase enthalpies and internal energies are functions of the dissolved components in addition to pressure and temperature. The component mass balances are described by:

\[
\frac{\partial \left( \sum_{\alpha} q_{\alpha} X_{\alpha} S_{\alpha} \right)}{\partial t} - \sum_{\alpha} \text{div} \left( q_{\alpha} X_{\alpha} \frac{k_{\alpha}}{\mu_{\alpha}} K(\text{grad} P_{\alpha} - q_{\alpha} \mathbf{g}) \right) - \sum_{\alpha} \text{div} \left( D_{\alpha,pm} q_{\alpha} \text{grad}X_{\alpha} \right) - \sum_{\alpha} q_{\alpha}^{c} = 0 \quad \kappa \in \{\text{H2O, CO2}\} \tag{4}
\]

Besides the equations given in (3), a further auxiliary condition needs to be fulfilled for each phase in the \(2p2cni\) model:

\[
\sum_{\kappa} X_{\kappa}^{c} = 1 \tag{5}
\]

The fluid properties of the CO2-rich phase are calculated as functions of pressure and temperature. The properties of the brine phase additionally depend on the salinity, and in the compositional model, on the CO2 mass fraction.

In the compositional model, the mutual solubilities of brine and CO2 are calculated as a function of the brine salinity, the pressure and the temperature according to Spycher and Pruess (2005). For further information on the fluid property functions, the reader is referred to Bielinski (2006).

Equations (1, 2), and (4) are discretised with a fully implicit Euler scheme in time and with a vertex-centered finite volume method (BOX method) in space (Helmig et al., 1997). This is implemented in the new multi-scale, multi-physics toolbox DuMuX (Flemisch et al., 2007, http://www.dumux.de).

### 2.2 Transfer of Primary Variables and Mass Conservation

The \(2pni\) model solves for a constant set of primary variables \(i.e., \) the brine-phase pressure \(p_{w}\), the CO2 saturation \(S_{n}\) and the temperature \(T\).

The set of primary variables in the \(2p2cni\) model, however, changes with the phase state. The phase state is a control volume specific information and can either be brine phase only, CO2-rich phase only or both phases. If only one phase is present, either brine or CO2, the value of \(S_{n}\) is constant and a mass fraction is used as primary variable. For a single-phase brine system, the mass fraction of CO2 in the brine phase is the primary variable. For a single-phase CO2 system, the mass fraction of H2O in the CO2-rich phase is the primary variable. If the mass fraction reaches the
value of maximum solubility $X_{eq}^{CO2}$, a second phase appears, and the primary variable is switched to the CO$_2$ saturation $S_n$ (Tab. 1). For saturation values of 0.0 and 1.0, the model switches back to brine phase only or CO$_2$-rich phase only, respectively.

<table>
<thead>
<tr>
<th>Primary variables</th>
<th>Brine phase only</th>
<th>CO$_2$-rich phase only</th>
<th>Both phases</th>
</tr>
</thead>
<tbody>
<tr>
<td>$p_w$, $X_w^{CO2}$, $T$</td>
<td>$p_w$, $X_w^{H_2O}$, $T$</td>
<td>$p_w$, $S_n$, $T$</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Mass fraction</th>
<th>Brine phase only</th>
<th>CO$_2$-rich phase only</th>
<th>Both phases</th>
</tr>
</thead>
<tbody>
<tr>
<td>$X_w^{CO2} &lt; X_{eq}^{CO2}$</td>
<td>$X_w^{H_2O} &lt; X_{eq}^{H_2O}$</td>
<td>$S_n$ $X_w^{CO2}$ = $X_{eq}^{CO2}$ $X_w^{H_2O} = X_{eq}^{H_2O}$</td>
<td></td>
</tr>
</tbody>
</table>

During the switch from the 2p2dni model to the 2p2cnii model, the primary variables need to be transferred on each node of the grid in a way that guarantees the conservation of the CO$_2$ mass. In regions where only one phase is present in the 2p2dni model, either brine or CO$_2$, the mass fraction of CO$_2$ in the brine phase $(X_w^{CO2})$ or the mass fraction of H$_2$O in the CO$_2$-rich phase $(X_n^{H_2O})$ are initialised with zero accordingly in the 2p2cnii model.

If both phases are present in the 2p2dni model, the CO$_2$ saturation in the 2p2cnii model needs to be corrected locally after the model coupling. The reason for the correction is the additional CO$_2$ mass in the brine phase due to the assumed maximum solubility $X_{eq}^{CO2}$ (Tab. 1). The equality of the CO$_2$ mass in a given control volume before and after the model coupling leads to the following equation:

$$[S_n \rho_w]_{2p2dni} = [S_n X_w^{CO2} \rho_w + S_w X_w^{H_2O} \rho_w]_{2p2cnii} \quad (6)$$

Replacing $[S_w]_{2p2cnii}$ by inserting the first auxiliary condition (Eq. 3) and solving for $[S_n]_{2p2cnii}$ leads to:

$$[S_n]_{2p2cnii} = \frac{[S_n \rho_n]_{2p2dni} - \{X_w^{CO2} \rho_w\}_{2p2cnii}}{\{p_n X_w^{CO2} - \rho_w X_w^{H_2O}\}_{2p2cnii}} \quad (7)$$

Since the value of $S_n$ is reduced by this transformation, the capillary pressure is lowered, and as a consequence, the value of $p_n$ decreases. A decrease in the CO$_2$ pressure causes a change in the maximum solubility of CO$_2$ in the brine phase which also has an influence on the brine density. Thus, a few iterations with updated 2p2cnii terms on the right hand side are performed for Equation (7).

On the edge of the CO$_2$ plume, small values of $[S_n]_{2p2dni}$ can occur. These values lead to a negative result for $[S_n]_{2p2cnii}$ in Equation (7) which means that there is not enough CO$_2$ present to reach the maximum solubility in the brine phase. In this case, the phase state in the 2p2cnii model is set to brine phase only, and the CO$_2$ mass fraction is calculated with the following equation:

$$\{X_w^{CO2} \rho_w\}_{2p2cnii} = \{S_n \rho_n\}_{2p2dni} \quad (8)$$

Like in Equation (7), a few iterations are necessary to take the influence of the resulting CO$_2$ mass fraction on the value of $[\rho_w]_{2p2cnii}$ into account. This method guarantees the conservation of the CO$_2$ mass in the system, however, due to the modifications in saturation and composition it does not conserve the mass of H$_2$O.

### 3 APPLICATION EXAMPLE

In this section, the implemented sequential coupling concept is tested using an idealised, radially symmetric application example.

The model domain is shown in Figure 2. Taking advantage of the radial symmetry, a 30° slice of the reservoir is modelled. It has a thickness of 20 m and a radius of 800 m. The inner radius of the slice is 0.05 m representing the injection well. The vertical discretisation is 1 m, whereas the cell size increases exponentially, and in angular direction the discretisation is 7.5°, which leads to a grid of 3 200 hexahedral elements.

The reservoir has a porosity of 0.25 and a permeability of 200 mD in $x$-$y$-direction and 20 mD in $z$-direction. For the description of the capillary pressure $p_c$ and the relative permeabilities $k_r$, the relations given by Brooks and Corey, 1966, are applied. The Brooks-Corey parameters are set to $\lambda_{BC} = 2$ and $p_d = 5 000$ Pa. The residual saturations of the brine and the CO$_2$-rich phase are set to 0.25 and 0.025 respectively. The rock has a density of 2 650 kg/m$^3$ and a heat capacity of 800 J/(kg·K). The heat conductivity of the porous medium is calculated as a function of brine saturation according to Somerton et al. (1974):

$$\lambda_{pm}(S_w) = \lambda_{drp} + \sqrt{S_w} \cdot (\lambda_{sat} - \lambda_{drp})$$

where $\lambda_{sat} = 2.7$ W/(m·K) and $\lambda_{drp} = 0.32$ W/(m·K). The salinity of the brine is 0.1728 kg/kg in the whole domain and is assumed to stay constant.

Initially the pressure and the temperature are determined by the hydrostatic pressure distribution and a geothermal gradient of 0.032 K/m starting at the reservoir bottom with a pressure of 259.74 bar and a temperature of 353.47 K. On the outer boundary, the values of $p_w$ and $T$ are set according to the initial conditions. On the top, bottom, and lateral boundaries, Neumann no-flow boundary conditions are
applied. The CO\(_2\) is injected at a rate of 0.1 kg/s with a temperature of 308 K for a period of 1 year. After the injection stops, the system is modelled for another 1000 years.

In the sequentially coupled model, the switch from the 2pni to the 2p2cni formulation is performed as the injection stops. For comparison, a reference model that applies the 2p2cni formulation over the whole timescale is run (Fig. 2).

In addition, two variations of the base case example are simulated. The first one applies a grid, which is refined once in radial, angular and vertical direction, leading to 25,600 hexahedral cells. The second one applies a heterogeneous permeability field (Fig. 3) created with the geostatistical package gstat (Pebesma, 2004). Even though the examples shown here are far below a realistic scale, they are well-suited to show the general advantages and disadvantages of sequential model coupling. A more detailed study on the effects of heterogeneities, larger geometries, and injection rates will be the subject of future work.

### 4 DISCUSSION OF THE RESULTS

#### 4.1 Simulation Time

**4.1.1 Base Case**

In Figure 4, the CPU time is plotted versus the simulation time. The simulation of the one-year injection period and the subsequent 1000 years of relaxation takes 10.59 h with the reference model and 3.19 h with the sequential model on one core of an Intel Core 2 Duo E6300 processor with 1.86 GHz. Thus, the sequential coupling leads to a speed-up of approximately factor 3. As expected, a large speed-up takes place during the injection period, when the less complex 2pni formulation is applied in the sequential model. In addition to its stronger nonlinearities, the reference model is slowed down by the large number of phase switches that occur during this period. The phase switches along with the change of primary variables generally reduce the convergence rate of the Newton algorithm applied in each timestep.

However, the sequentially coupled model also remains faster in the first few years after injection stop (Fig. 4, bottom) even though both models apply the 2p2cni formulation at this point. This can be explained by the presence of the single-phase CO\(_2\) region that only develops in the reference model during the injection, the residual brine in the vicinity...
of the well does not dissolve in the sequential model. After injection stops, this single-phase region gradually dissolves due to brine imbibition. This transformation from a single-phase system to a two-phase system requires an additional number of phase switches in the reference model. Figure 5 (top) shows the timestep size of both models during the injection period. Even though the sequential model is faster due to the less nonlinear equation system that needs to be solved, the main part of the speed-up is caused by the much better convergence and thus the larger timesteps of the \( 2 \text{pni} \) model compared to the compositional model during the injection period. This is also shown in Figure 5 (bottom) where the number of Newton iterations per timestep is plotted against the model time. Even though another setting of the timestep management can have an influence on the exact value of the resulting speed-up factor, the convergence of the compositional model will always be reduced due to the phase switches.

### 4.1.2 Refined Grid and Heterogeneities

The simulation of the heterogeneous case is also performed on an Intel Core 2 Duo E6300 processor with 1.86 GHz whereas the refined-grid case required a parallel simulation on eight Intel Xeon E 5440 CPUs with 2.83 GHz. In Table 2 the CPU times for all three cases are shown as well as the speed-up factor. For both, the heterogeneous and the refined-grid case, the speed-up of the sequential model with respect to the reference model is increased compared to the base case. As in the base case, the main speed-up takes place during and shortly after the injection period.

<table>
<thead>
<tr>
<th>Case</th>
<th>Sequential model CPU time (h)</th>
<th>Reference model CPU time (h)</th>
<th>Speed-up factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Base case</td>
<td>3.19</td>
<td>10.59</td>
<td>3.3</td>
</tr>
<tr>
<td>Heterogeneous case</td>
<td>4.74</td>
<td>31.64</td>
<td>6.7</td>
</tr>
<tr>
<td>Fine-grid case</td>
<td>8.3</td>
<td>38.75</td>
<td>4.7</td>
</tr>
</tbody>
</table>

It is observed, that for both variations the number of Newton iterations per timestep in the reference model increases with respect to the base case, which leads to a further reduction of the timestep size (Fig. 6). In both cases the reduced convergence of the reference model is caused by a larger number of nodes which encounter a phase switch either due to the finer grid or due to the increased surface area of the plume in the heterogeneous case.

### 4.2 \( \text{CO}_2 \) Phase Distribution

In Figure 7 (top), the total \( \text{CO}_2 \) mass and the mass of \( \text{CO}_2 \) in the \( \text{CO}_2 \)-rich phase is plotted over time. The total \( \text{CO}_2 \) mass is the same in both models since the same amount of \( \text{CO}_2 \) is injected. For the large timescale of 1 000 years, the mass of \( \text{CO}_2 \) in the \( \text{CO}_2 \)-rich phase calculated by the reference and by the sequential model shows a good agreement.
If the phase distribution within the first four years is considered (Fig. 7, bottom), small differences between the formulations can be observed. The deviation with respect to the CO₂ mass in the CO₂-rich phase during the first year is caused by the different formulations. In the sequential model, all the CO₂ is in the CO₂-rich phase, while in the reference model, there is already some amount dissolved in the brine phase.

When the injection is shut down, the sequential model switches to the 2p2cni formulation. After the switch the phase distributions in both models are in very good agreement.

The refined-grid case and the heterogeneous case show the same behaviour with respect to the CO₂ phase distribution. The phase distribution of CO₂ is an integral measure that is evaluated for the whole reservoir. It does not guarantee that the model results are equal. For a more detailed comparison, the local CO₂ distribution needs to be considered. This is done in the next section.

4.3 Local Differences

4.3.1 CO₂ Saturation Distribution

In this and in the next Section the z-ratio of the figures is increased by a factor of 5 to also display the vertical CO₂ distribution. Figure 8a, b shows the CO₂ saturation distribution after one year of CO₂ injection for the reference model and the sequential model, respectively. At this point, the switch from the 2pni to the 2p2cni formulation has already taken place in the sequential model leading to a reduced CO₂ saturation due to the saturation correction (Sect. 2). Apart from that, the reference model allows the aforementioned development of a single-phase CO₂ region in the vicinity of the well during the injection period compared to the sequential model which always holds a residual brine phase. The resulting larger CO₂ accumulation of the reference model close to the well leads to a slightly reduced lateral spreading of the plume. In addition to that, the small amount of CO₂ that dissolves in the brine further reduces the plume extent.

These effects can be seen in Figure 8c which shows the resulting saturation difference between the reference and the sequential model. Figure 9 shows the saturation distribution of the reference model and saturation differences compared to the sequential model 100 years after the injection phase. There are still small local saturation differences between the models, however, the maximum difference is 0.015 which
is more than an order of magnitude less than the saturation differences at the end of the injection phase (Fig. 8c).

4.3.2 CO₂ Mass Fraction Distribution

Figure 10a, b shows the CO₂ mass fraction distribution of the reference model and of the sequential model after the switch from the 2pmi to the 2p2cni formulation. The differences between the models (Fig. 10c) result from the differences in the saturation distribution. Thus the plume of dissolved CO₂ is slightly larger for the sequential model corresponding to the larger extent of the CO₂-phase plume. 1000 years after the injection phase, almost all the CO₂ is dissolved in the brine. Note that only a small amount of CO₂ is injected. The heavier, CO₂-rich brine leads to the development of density-driven fingering and convective mixing. In Figure 11a, b the mass fraction distribution shows that a large amount of the dissolved CO₂ already has sunk to the bottom of the reservoir. The fingering in a homogeneous reservoir like the one regarded here is induced by minimal numerical inaccuracies and is always heavily influenced by the grid. Since the initial mass fraction distribution is not completely the same, the triggering of the convective fingers is not identical either (Fig. 11c).

4.3.3 Injection Pressure

With respect to the well-block injection pressure a difference of approximately 0.2 bar can be observed between the reference and the sequential model (Fig. 12). In the reference model the injection pressure is smaller since the injected volume is reduced due to the dissolution of CO₂ in the brine phase. As the differences in saturation and mass fraction distribution show, the pressure differences will not have a large effect on the long-term behaviour of the CO₂ plume. However, they show that with respect to a pressure study, sequential coupling will lead to an overestimation of the pressure increase. Nevertheless this study is not aiming for an investigation of the injection pressure since this would require to take into account many other factors like the effects of boundary conditions and grid resolution, the compressibility of the rock matrix, and changes in the hydraulic parameters due to mineral precipitation or matrix deformation.

4.3.4 Influence of Grid Refinement and Heterogeneities

For the variations with the refined grid and the heterogeneous intrinsic permeability field the magnitude of local differences between the reference and the sequential model does not increase. In the heterogeneous case the final mass
fraction distribution shows less deviations between the reference and the sequential model, since the downward migration is mainly influenced by the heterogeneities and not so much by the shape and the local occurrence of the convective fingers.

**CONCLUSIONS AND PERSPECTIVES**

This study presented a first implementation of a sequential coupling of models of different complexity during and after the injection of CO₂ into a reservoir. It could be shown that:
- the model predictions for the distribution of CO₂, both in phase and dissolved in brine, are in excellent agreement when large time periods are considered;
- the sequentially coupled approach leads to a significant improvement of model efficiency.

It is expected that random processes like the triggering of fingers by instabilities is 'in reality' always dominated by permeability perturbations or local heterogeneities. Thus, the deviations in the mass fraction distributions of dissolved CO₂ are considered as non-limiting for the applicability of the proposed approach.

Both model domain and injection period in this numerical study are far below a realistic scale. Thus, ongoing work is currently concerned with the transfer of these results to relevant spatial and temporal scales. It is of particular importance to develop criteria of 'when' and 'how' such coupling can be done, and what the error introduced by neglecting some of the processes during certain periods is.

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