European Geosciences Union General Assembly 2016, EGU
Division Energy, Resources & Environment, ERE

Coupling of geochemical and multiphase flow processes for validation of the MUFITS reservoir simulator against TOUGH

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Abstract

A benchmark study is presented comparing the outcome of two different simulators, the established TOUGH and the modern, more efficient MUFITS, with the purpose to evaluate the coupling of geochemical processes to multiphase flow in the context of subsurface CO_2 storage. The results of the two simulators are in excellent agreement. The implemented simplified one-way coupling is a computationally efficient alternative to fully coupled reactive transport simulations. Every coupled process simulation framework benefits from the inclusion of different tools, which is the only way to enable validation and control of numerical results.
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Peer-review under responsibility of the organizing committee of the General Assembly of the European Geosciences Union (EGU)

Keywords: Reactive transport, CO_2 geological storage, benchmark, MUFITS, TOUGH

1. Introduction

Coupled reactive transport modelling at continuum or Darcy scale [1,2] finds increasing attention in the investigation of interacting hydrodynamical, physical, chemical and biochemical processes in natural or engineered systems. In particular, in recent years, it has been extensively employed to assess feasibility of subsurface CO_2 or gas storage in diverse geological settings [3–5]. From a geoscientific point of view, these models are characterized by a large computational demand (CPU-time) and by numerical instabilities and frequent non-convergence of the simulations. These issues limit in practice the applicability to relatively simple systems, usually at a much lower spatial resolution and for much shorter time scales than non-reactive multiphase flow simulations in the same geological settings. Published studies of reactive transport frequently consider only very simple geometries, 1D/2D or extremely coarse 3D grids [5–9]. It is obvious that in this way many geological features such as faults, discontinuities, spatial heterogeneity cannot be represented adequately.

Oversimplifications may concern the chemistry as well, including in the models only a subset of the potentially occurring reactions (or components) of a given system. This has two main reasons: (1) chemistry is usually the

computational bottleneck for coupled reactive transport models, demanding up to 90 % of overall CPU time. (2) the geochemical models are affected by extremely large uncertainties concerning thermodynamic and kinetic parameters [10–12], as well as the mineralogical heterogeneity of the investigated porous media, which makes it difficult to define a representative medium.

In this context, coupled models can be used for qualitative process understanding rather than as reservoir-scale quantitative, predictive tools for subsurface utilization. It is important to stress that purely hydrodynamic (i.e. non-reactive) simulations, being much less demanding in terms of computational efforts, are routinely performed on multi-million element spatial discretizations, several orders of magnitude above what is currently achievable for coupled models.

A promising way to reduce the oversimplifications of the physical systems or the coarsening of simulation grids is represented by **simplifying the coupling** itself [13]. The typical characteristics of CO_2 storage systems in siliciclastic reservoirs are suited to be modelled employing a **one-way coupling** between hydrodynamical and chemical processes. Such a method has already been successfully applied to long-term, reservoir-scale assessment of mineral trapping at the Ketzin pilot site in Germany [14,15].

The complexity of the targeted physical and chemical processes under investigation can only be approached by numerical methods. It is, therefore, crucial to dispose of different tools to benchmark and verify simulation results, ideally using more than one numerical approach, which in practice means more than one simulator. These considerations led us to include in the original implementation of the one-way coupling presented in [13], which is based on the TOUGH family simulators [16,17], with one based on the more modern and efficient multiphase flow simulator MUFITS [18]. The aim of this paper is to benchmark the results of the two simulators with respect to the coupling approach defined in [13]. It is intended as validation of both the simulators and the coupling itself.

In the following sections, the simplified one-way coupling is first outlined, followed by the description of the actual case study. Finally, the predictions by the TOUGH and MUFITS simulators and their results when coupled with geochemistry are presented.

2. Simplified one-way coupling approach

The idea underlying the one-way coupling is to combine, in a post-processing approach, independently obtained batch geochemical simulations with non-reactive flow simulations. This means that the mutual feedback between chemistry and hydrodynamical processes is actually disregarded. It concerns both the mass balance of fluids or solutes (e.g., minerally trapped CO_2 which is not "available" for transport anymore) and the petrophysical properties of the porous medium (porosity, permeability). For the approach to be valid, however, only three necessary and sufficient conditions have been identified [13]:

1. the time scale of mineral reactions is much larger than that of hydrodynamical processes;
2. the main driving force for mineral alterations is the presence of a limited number of fluid components (e.g., the injected CO_2, either in a separate phase or in dissolved form);
3. the mineral reactions do not affect significantly the petrophysical properties of the medium (porosity, permeability) at least not until the system has reached substantial hydrodynamical equilibrium.

Even if they may appear quite strict, all these conditions are usually met with good approximation in the typical subsurface CO_2 storage system in siliciclastic reservoirs. In fact, chemical reactions capable of significantly altering porosity and permeability of the medium, such as precipitation of carbonates, are expected to be very slow (kinetically-limited), and thus to occur long after abandonment of a storage reservoir. Under these circumstances, the feedback of chemistry on petrophysical properties can be safely disregarded. Conversely, since the transport of solutes will be very slow when approaching hydrodynamical equilibrium, it will not affect chemistry when reactions start becoming significant. Therefore, also the feedback between transport and chemistry will be limited.

The simplified coupling itself proceeds as follows: First, one single batch geochemical simulation and the hydrodynamic simulation of CO_2 injection are run separately for the entire simulation time. From the hydrodynamic simulations, the exposure time to injected CO_2 of each grid element is computed, distinguishing whether CO_2 is dissolved or in a free phase. If only dissolved CO_2 reaches the element, its concentration must exceed a user-defined threshold to consider the element "chemically active". The single batch geochemical simulation is subsequently ap-
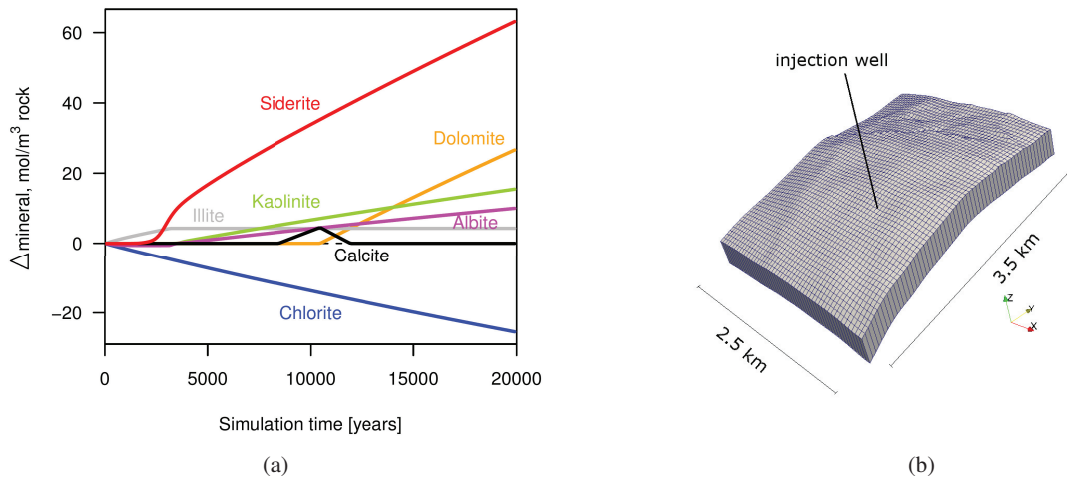
Fig. 1: Illustration of the benchmark study: **(a)** chemical model used in this study (from [13]), inspired by the Ketzin pilot site. The time axis refers to the reference CO_2-rich phase saturation of 0.5 with porosity 0.5. **(b)** 3D one-layer model of the Ketzin anticline used for the validation. The 2.5 km x 3.5 km model comprises 2950 elements. The model is exaggerated by a factor 5 in the vertical direction.

plied to each grid element starting from the arrival time of CO_2 (either dissolved or in a free phase). The state reached at the end of exposure time or, alternatively, after consumption of the CO_2 which entered the element - in order to maintain mass balance - is prolonged until the end of the simulation time. For a more detailed, step-by-step description and discussion of the uncertainty in the definition of the threshold for dissolved CO_2, the reader is referred to [13].

3. The benchmark: simplified Ketzin model

From a geochemical point of view, the benchmark case study reflects the complexity of a typical subsurface storage site. It is loosely based on the published Ketzin reference geochemical model [15] and has already served the purpose of an in-depth analysis of the simplified coupling approach [13]. The considered mineral assemblage comprises the rock-forming phases anhydrite, albite, hematite, illite and chlorite, together amounting to 24 % in volume of the rock. The rest – mainly quartz, which makes up 40 % in volume at the site – is considered inert. In the model, kaolinite and the carbonates siderite, magnesite, dolomite and calcite are included as secondary phases, and allowed to precipitate upon reaching super-saturation. Calcite, dolomite and anhydrite are modelled at local thermodynamical equilibrium; all other minerals are taken into account with a kinetic formulation following Palandri and Kharaka [19]. The single batch simulation used for this study has been obtained using the TOUGHREACT simulator [16]. It is important to stress the fact that any other code or simulator could have been used to perform this simulation [25].

The reactions occurring in a reference batch simulation after contact with injected CO_2 are displayed in Fig. 1a. Anhydrite (not displayed) and chlorite act as main cation donors; iron carbonate siderite starts precipitating after a few hundred years, followed by a transient appearance of calcite, which is then redissolved in favour of dolomite (acting as proxy for Mg-carbonate). The overall amount of reactions is similar to those predicted in site-specific Ketzin studies [15]; their net effect is a minor decrease in porosity after 20,000 years, since the precipitation of carbonates is compensated by dissolution of anhydrite and chlorite. Such decrease can be safely considered negligible with respect to hydrodynamic properties of the medium, amounting to a few per mille in terms of relative change.

The 2.5 km $\times$ 3.5 km simulation grid (Fig. 1b) is composed of one single layer with 2950 elements. It reflects the actual 3D geometry of the anticline of the Stuttgart Formation at the Ketzin pilot site [20–22]. The thickness of the model is about 74 m and thus it encompasses both main stratigraphic units of the Stuttgart Formation at Ketzin. The slope of the anticline structure drives a significant upward migration of CO_2 with time. In the present benchmark, porosity has been set to 0.2 and the isotropic permeability to 1×10^{-13} m^2.

Relative permeability has been set to a Van Genuchten/Mualem model with an exponential parameter $\lambda = 0.64$, residual water saturation $S_{wr} = 0.15$, saturated water content $S_{ws} = 0.98$ and residual gas saturation $S_{gr} = 0.05$. These parameters were found to fit best the experimental measurements performed for the Ketzin site [20]. For the benchmark, no capillary pressure has been defined.

The model starts with fully saturated initial conditions at a hydrostatic pressure distribution, obtained through equilibration runs. All simulations are isothermal at a temperature of 35 °C. 67,000 t of CO_2, the actual amount of CO_2 injected at the Ketzin site, are injected at a constant rate in 5 years in the element highlighted in Fig. 1b, into the flank of the anticline. After injection, the system is let free to evolve until a total simulation time of 3,000 years is reached.

The design of the model ensures a realistic pressure build-up; its dimension is large enough to avoid the arrival of injected CO_2 at the model boundaries during the simulation time. Concerning the boundary conditions, they are set as no-flow at the lateral elements in x-direction, while those in the y-direction are set constant after equilibration.

3.1. Coupled model implementation with MUFITS and TOUGH simulators

The MUFITS simulator [18,23,24] is a modern scientific code designed for efficiency and modularity. Its numerical kernel implements a fully-implicit finite-volume scheme with upwinding. Several fluid properties modules (i.e., EOS-modules) are available, adapted to describe the thermophysical characteristics of different fluids (phases, components, equation of state, density, viscosity, etc).

In this work, the recently developed GASSTORE module [24] has been employed for the simulations. It covers non-isothermal three-component three-phase solid-liquid-gas flow of water, gas and salt components and salt precipitation and dissolution. Thermophysical properties of different gas components (viscosity and density) are tabulated as function of pressure and temperature for efficient spline interpolation, and derived with an approach similar to that implemented by the ECO2N module of the TOUGH simulator [17]. The two thermophysical packages are in excellent agreement [24].

There is a slight difference concerning the chosen implementation of boundary conditions in the two simulators. In TOUGH, large pore volume multipliers are set to provide constant conditions throughout the simulations. In MUFITS, further auxiliary elements are placed contiguously to the northern and southern boundaries of the model (short sides). A further slight difference is represented by the calculation of the relative permeability function. In TOUGH it is internally computed by a corresponding routine; in MUFITS it is externally computed and provided to the simulator as a table, discretized in steps of 0.005 units of water saturation. Such values are internally interpolated to generate the actual values for relative permeabilities.

Performing the simplified coupling is a matter of post-processing the simulation results of different simulators. The current implementation is written in the high-level open source language R (`www.r-project.org`) and consists in routines capable of loading and processing simulation results from the data formats employed by the simulators. It is a component to our flexible simulation environment [25].

4. Results

Fig. 2 shows the distribution of gaseous and dissolved CO_2 after 3,000 years, respectively, in free phase (gas) and dissolved form for the two simulators. Reservoir pressures and thermophysical properties of the fluids in the two simulations are in excellent agreement within a range of 0.1 %. Visible discrepancy between the models only concerns a few grid elements located at the forefronts of the migrating CO_2. Since these elements are reached by very small concentrations of dissolved CO_2, they do not contribute significantly to the chemistry.

The good agreement between the two simulators is confirmed by the analysis of the cumulative volumes exposed respectively to gaseous and to dissolved CO_2 (Fig. 3, left). More importantly, the calculated amount of mineralized CO_2 in newly formed carbonates (Fig. 3, right) is in excellent agreement, confirming that the slight discrepancies in the predicted flow of CO_2 have no significant impact on chemical reactions.

This benchmark is particularly challenging because of the large volumes of the grid elements, which make the simulations prone to numerical errors due to tiny concentrations of dissolved CO_2 and gas saturations reached in the elements. The simulations appear to be sensitive to the numerical interpolation of relative permeability functions,
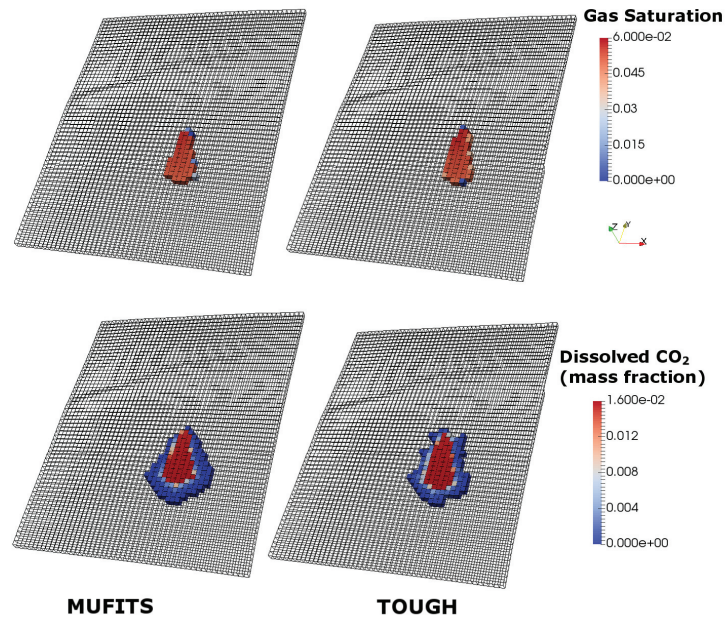
Fig. 2: Visual comparison of the gaseous (top) and dissolved (bottom) CO$_2$ migration predicted by MUFITS (left) and TOUGH (right) simulators after 3,000 years.
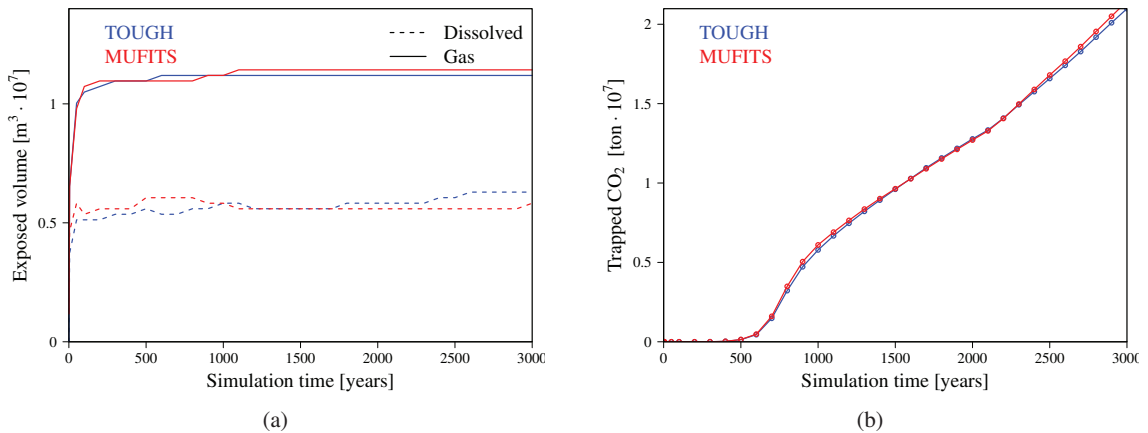

Fig. 3: **(a)**: total reservoir volume in contact with injected CO$_2$, in both gaseous and dissolved states as predicted by the two simulators. **(b)**: CO$_2$ mineralization calculated with MUFITS and TOUGH via the simplified coupling.

which in this benchmark often are about the irreducible gas saturation value, where the changes in relative permeability are at a maximum.

For these reasons, finer spatial discretizations are expected to actually increase the agreement of different simulators. Moreover, the slight differences introduced by the definition of boundary conditions in the two simulators are the reason for the discrepancies in the hydrodynamical simulation results. However, these discrepancies play a negligible role when considering the coupling with chemistry. In presence of slow reactions, it is preferable to introduce approximations in the coupling instead of coarsening the simulation grid.

5. Conclusion

A simplified one-way coupling with chemistry has been applied to TOUGH and MUFITS simulations with excellent agreement between their results. The benchmark is constituted by a simplified, one-way coupling approach to reactive transport in an exemplary case of subsurface CO_2 storage in a saline aquifer and siliciclastic reservoir inspired by the Ketzin pilot site in Germany, which displays ideal characteristics for the decoupling of processes. In this very efficient approach, the underlying processes, multiphase flow and chemistry, can be calculated by means of different, arbitrary tools, providing the modeller with the opportunity to verify the results of complex simulations. The agreement of the calculated mineralization starting from two otherwise very slightly discrepant hydrodynamical simulations proves that the method is robust and captures very well the ongoing main processes.

The approximated approach can be used to perform sensitivity analyses on geochemistry and to use fine spatial discretizations with million elements grids, which is otherwise unfeasible for fully-coupled reactive transport simulations. The small loss of accuracy in the predictions with respect to target variables is more than compensated by the ability to run hydrodynamic simulations on finely discretized grids, thus achieving a better description of geological features and providing results which are also numerically more stable.

The applicability of the one-way coupling can be further extended beyond systems which strictly comply to the three necessary and sufficient conditions: (1) separability of time scales; (2) only one transport-limited reactant (i.e. CO_2); (3) amount of chemical reactions not sufficient to alter petrophysical properties of the medium. A promising way is represented by combining the one-way and fully-coupled approach in different regions of a simulation domain and at different times. Where and when the three above mentioned conditions are met, the simulations could switch from a tight, full coupling to the simplified, one-way approach.

Finally, we stress that it is not only good practice but crucial to include different concurrent numerical simulators in the modelling workflow. Comparing different numerical approaches is namely the only way to cross-validate complex simulation results.

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