Geoscientific
Model Development
Discussions

Interactive comment on "A coupling alternative to reactive transport simulations for long-term prediction of chemical reactions in heterogeneous CO_2 storage systems" *by* M. De Lucia et al.

Anonymous Referee #2

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Overview

The paper presents an alternative methodology based on an one-way coupling approach for multiphase reactive transport simulations for carbon storage modelling in geological formations. The authors assume that CO2 is the only driving force for the geochemical reactions and that the plume migration is not considerable affected by eventual precipitation of carbonate minerals. The reservoir rock is predominantly composed of slowly reactive silicate minerals. Their approach consists of independent steps in which the hydrodynamic flow equations are solved, then a reaction path calculation is performed whose result is appropriately scaled for each grid element (the scaling at each element depending on an exposure time to CO2 obtained from the hy-

drodynamic calculations). The method, therefore, relies on a threshold for the minimum dissolved concentration of CO2 that indicates if a given element in the grid is chemically active. The paper then presents numerical experiments to validate their simplified one-way coupling approach against a fully-coupled one.

General comments

The authors have demonstrated the feasibility of a cheap one-way coupling approach for modelling reactive transport as an alternative to the expensive fully-coupled one, which requires many geochemical calculations. However, the presented approach relies heavily on the assumption that the major chemical composition of the fluid and rock does not change over the years that follow CO2 injection and its migration, so that the reactive effects can be later incorporated by solving a reaction path problem from which a proper scaling (depending on an exposure time to dissolved CO2) is used to determine the actual chemical composition of the fluid and rock. They have presented a comparison of their results with a fully-coupled one for only 2000 years (Figures 4 and 5 for the total volume of the reservoir exposed to dissolved CO2). These figures start to show substantial deviations from the fully-coupled approach for times after 300 years, which shows the one-way coupling scheme misses the opportunity to capture a more realistic distribution of dissolved CO2 over the reservoir. Despite this issue, the authors show in Figure 6 and 7 some good matching of their reaction path calculation, for some given grid point, with the evolution of the chemical system from the fully-coupled approach for 2000 years. What is the behaviour of your estimates for times greater than 2000 years? Do every grid point behaves the same as the one shown in Figures 6 and 7? Because the total reservoir volume exposed to dissolved CO2 is considerably greater in the one-way coupling approach, why don't we see this affecting Figures 6 and 7?

Specific comments

Page 6220, line 15: This statement is not true for highly reactive rocks, such as car-

bonates. In a carbonate reservoir, important mineral alterations should be expected to occur at the time scale of the hydrodynamic processes. The feedback between flow and geochemical reactions in this case cannot be neglected. Please specify the type of reservoir this work is aimed to.

Page 6220, line 25: Again, this cannot be generalized to all types of reservoirs. The porosity and permeability of carbonate reservoirs in regions rich in dissolved CO2, and consequently with high acidity, should experience large variations that can in fact affect the hydrodynamic processes at short time scales.

Page 6221, line 10: How does this one-way coupling of Klein et al. (2013) used by the authors to decouple the chemical processes from the transport equations differs from the classical operator splitting scheme so commonly used to solve coupled partial differential equations with multiple chemical and physical processes? The literature is very rich with such strategies. See for example:

âĂć Jacques et al. (2006) "Operator-splitting errors in coupled reactive transport codes for transient variably saturated flow and contaminant transport in layered soil profiles"

Page 6221, line 20. Here it is very important that a more detailed description of the calculation steps be provided. The expression exposure time appears here for the first time after the abstract. Please explain this in the preceding paragraph. Do your hydrodynamic simulations account for dissolution of gaseous CO2 in brine? How is this done? I believe these hydrodynamic equations should be written in the manuscript (at least as an appendix section) to describe the physics and chemistry accounted for (dissolution of gaseous CO2 is also a reactive process). What is characteristic water saturation? I could not find the definition of this expression in the manuscript. Please explain a bit further how the computation of this special water saturation and concentration of dissolved CO2 during exposed time is done.

Page 6224, line 10. More explanation about this analytical scaling is necessary here for a better understanding of the introduction of the reactive processes in the simulation.

Please provide more information on why this is necessary to enhance the clarity of the manuscript. What is the limitation of this approach? Can this be extended to more complex chemical systems, with heterogeneity throughout the reservoir (in both fluid and rock)?

Corrections

1. Change explicitely to explicitly in page 6225, line 15.

2. Change starts to start in line 10 of page 6232. Also improve the grammar in this line.

3. The use of dots for scalar multiplication (throughout the manuscript) is not necessary. This only creates noise in the equations.

Suggestions and discussion

Why not use pH as the control variable for measuring reactivity instead of concentration of dissolved CO2? The pH at every element could be obtained by using some correlation model on the concentration of dissolved CO2 (so that you don't need to perform the expensive chemical equilibrium calculations to determine it).

One could claim that the more dissolved CO2 in the brine, the higher its acidity and so its reactivity. However, if brine contains some dissolved carbonate minerals, then you can still have a concentration of dissolved CO2 above your prescribed threshold, but the pH of brine is perhaps not acidic enough to cause substantial reactions with the rock-forming minerals. By using pH as the control variable, it should be easier to come up with a threshold that indeed reflects the reactivity of the system.