



Pore scale study of multiphase multicomponent reactive transport during CO₂ dissolution trapping

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ARTICLE INFO

Keyword:

CO₂ sequestration
Solubility trapping
Multiphase flow
Reactive transport
Porous media
Lattice Boltzmann method

ABSTRACT

Solubility trapping is crucial for permanent CO₂ sequestration in deep saline aquifers. For the first time, a pore-scale numerical method is developed to investigate coupled scCO₂-water two-phase flow, multicomponent (CO₂(aq), H⁺, HCO₃⁻, CO₃²⁻ and OH⁻) mass transport, heterogeneous interfacial dissolution reaction, and homogeneous dissociation reactions. Pore-scale details of evolutions of multiphase distributions and concentration fields are presented and discussed. Time evolutions of several variables including averaged CO₂(aq) concentration, scCO₂ saturation, and pH value are analyzed. Specific interfacial length, an important variable which cannot be determined but is required by continuum models, is investigated in detail. Mass transport coefficient or efficient dissolution rate is also evaluated. The pore-scale results show strong non-equilibrium characteristics during solubility trapping due to non-uniform distributions of multiphase as well as slow mass transport process. Complicated coupling mechanisms between multiphase flow, mass transport and chemical reactions are also revealed. Finally, effects of wettability are also studied. The pore-scale studies provide deep understanding of non-linear non-equilibrium multiple physicochemical processes during CO₂ solubility trapping processes, and also allow to quantitatively predict some important empirical relationships, such as saturation-interfacial surface area, for continuum models.

1. Background

Multiphase reactive transport processes in porous media are widely encountered in scientific problems and engineering processes. Such processes are very complex due to multiple processes, dynamic phase interface evolution, and strong interaction between different processes. Taking carbon dioxide (CO₂) geo-sequestration, which is the topic of the present study, as an example. CO₂ geo-sequestration is currently considered as a method to limit greenhouse gas emissions into the atmosphere. During the sequestration, supercritical CO₂ (herein referred as scCO₂) is injected into deep saline aquifers or hydrocarbon reservoirs. Typically there are four principal trapping mechanisms of CO₂ at different time scales (IPCC, 2005): structural trapping, capillary trapping, dissolution trapping and mineral trapping. The lower density of scCO₂ compared with brine results in upward buoyant migration of scCO₂, which is prevented by low permeability caprocks, and such process is termed structural trapping. Capillary trapping is a process in which microscale scCO₂ bubbles (ganglia) are immobilized by capillary force inside the complex porous structures of the storage rock. scCO₂ dissolves into the surrounding brine through the brine-scCO₂ interface, and decreases the brine pH, resulting in dissolution trapping. Chemical reactions between

the ions dissolved and formation minerals lead to the permanent mineral trapping. From the above description, it can be found that multiple physicochemical processes are involved including scCO₂-brine two phase flow, multicomponent transport, scCO₂ dissolution, and mineral dissolution and precipitation.

While there has been much analysis and observation of macroscale phenomena related to the above processes, the underlying details of the small-scale processes that dominate system behavior, such as phase interface evolution, interfacial mass transport and chemical reactions, pore-scale species concentration distribution, etc., remain unrevealed. Understanding the meso/microscopic phenomena and the interaction between different processes is critical to achieving a better prediction of the related systems and to improving sequestration efficiency. Changes in pore-scale multiphase flow and reactive transport processes sometimes lead to significant variation of macroscopic phenomena. To date, great efforts have been devoted to understanding mechanisms of pore-scale reactive transport processes related to different trapping mechanisms. Capillary trapping can account for 10%–90% of the total injected volume of CO₂ (Iglauer et al., 2011). During capillary trapping, scCO₂ saturation and distribution in porous rocks are affected by lots of factors such as pressure, temperature, brine salinity, pore size distribution,

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wettability, etc. Experiment techniques such as X-ray micro-tomography have been developed to visualize pore-scale scCO_2 distributions under different thermo-physical conditions (Andrew et al., 2014; Chaudhary et al., 2013). To name only a few, Andrew et al. (2014) adopted X-ray to image scCO_2 in carbonates and sandstones under typical storage pressures and temperatures. It was found that the size distribution of residual ganglia obeys power law distributions. Besides, mineral trapping leads to permanent CO_2 sequestration. Luquot and Gouze (2009) experimentally studied the processes of scCO_2 flow-through limestone reservoir samples. Different CO_2 pressure leads to quite different rock dissolution patterns driven by transport-controlled or reaction-controlled mass transfer. Power-law relationship between porosity and permeability during dissolution processes was obtained. With relatively low CO_2 pressure, decrease of porosity was observed due to mineral precipitation (Luquot and Gouze, 2009).

Chemical reactions play significant roles on scCO_2 dissolutions, mineral dissolution and precipitation. In the literature, mineral dissolution and precipitation reactions have been extensively studied at the pore-scale, both experimentally and numerically. Different mineral dissolution processes including face-dissolution, uniform dissolution and channel dissolution (wormhole phenomena) have been identified under different flow, mass transfer and reaction rates (Luquot and Gouze, 2009; Kang et al., 2002; Tartakovsky et al., 2007; Noiriél et al., 2009; Szymczak and Ladd, 2009; Kang et al., 2010; Yoon et al., 2012). For the dissolution trapping, studies related to pore-scale scCO_2 dissolution processes are scarce (Soltanian et al., 2017), mainly for two reasons. On the one hand, scCO_2 dissolution has long been treated as an equilibrium process. Thus immiscible experiments, in which scCO_2 and water are equilibrated prior to injection, are usually performed in the literature focusing on multiphase flow processes without scCO_2 dissolution (Pentland et al., 2011; Andrew et al., 2013). However, due to non-uniform scCO_2 distribution and limited mass transport rate, scCO_2 dissolution is actually a non-equilibrium process requiring a prolonged time to deplete scCO_2 (Paterson et al., 2013; Akbarabadi and Piri, 2013). On the other hand, during scCO_2 dissolution, multiple physicochemical processes are involved including phase interface evolution, fluid-fluid interfacial chemical reaction, multicomponent mass transport, and reactions at fluid-solid interface. It is really challenging for current experimental techniques to capture all the physical fields interested (velocity, concentration, phase interface, etc.) within limited space of porous media. To the best of our knowledge, until very recently there are a few experimental studies available in the literature (Buchgraber et al., 2012; Chang et al., 2016; Chang et al., 2017). These studies are benefited from advanced microfluidic techniques which allow direct observation of scCO_2 -brine distribution and dissolution processes. Buchgraber et al. (2012) adopted 2D silicon micromodels represented Berea sandstone to investigate scCO_2 dissolution processes. Chang et al. (2016,2017) further performed comprehensive studies of scCO_2 dissolution under imbibition and drainage conditions. A set of scCO_2 dissolution experiments were performed in a heterogeneous 2D sandstone-analog micromodel at supercritical conditions. Based on the high resolution images of scCO_2 -brine distribution and phase evolution, several scCO_2 dissolution types were identified depending on local pore structure and mass transport rate.

Multiphase flow reactive transport in brine- scCO_2 -rock system depends on several parameters, such as pressure, temperature, flow rate, porosity, reactive surface area, tortuosity, wettability, etc. (Jun et al., 2013). The behavior of such coupled processes is unpredictable without the help of numerical modeling. Besides, compared with experimental studies, details of multiphase flow and reactive transport processes, especially the transport processes and species concentration distributions in different fluid phases, can be well captured by numerical modeling. However, due to the complex porous structures, multiple physicochemical processes, and strong interactions between different processes, it remains challenging to develop corresponding pore-scale numerical models. Such models should have the following abilities including capturing multiphase flow and distributions, taking into account both homo-

geneous and heterogeneous reactions, multicomponent mass transfer, and possible solid skeleton alteration. The aim of the present study is to develop an advanced pore-scale multiphase reactive transport model which can be applied to, but not limited to the reactive transport processes during scCO_2 dissolution process, and further to investigate the underlying details of scCO_2 -water distribution and evolution, concentration fields and time evolutions of important parameters. To the best of our knowledge, this study is the first one for numerically studying at the pore-scale coupled multiphase flow and chemical reactions in porous rocks during scCO_2 sequestration.

2. Numerical method

With the improvement of computational resources, pore-scale modeling, in which the complicated structures of a porous medium are explicitly resolved, has become a critical technique for exploring microscopic multiple physicochemical processes and underlying mechanisms. Both “top-down” or “bottom-up” numerical methods have been adopted for pore-scale modeling (Chen et al., 2013a). Conventional CFD methods such as finite volume method (FVM), finite element method (FEM), and finite difference method (FDM), solve the Navier–Stokes equation (mass, momentum, energy and concentration, etc.) directly and thus belong to “top-down” methods. The main challenge encountered by such methods is grid generation of complex porous structures and boundary treatment at complicated phase boundary. Molecular dynamics (MD), which falls in the category of “bottom-up” method, requires unacceptable computation resources, thus limiting its application to problems of engineering interest. The lattice Boltzmann method (LBM, also a bottom-up method), which is a mesoscopic method occupying the middle ground between conventional CFD methods and MD, has developed into an alternative and promising numerical approach for simulating transport processes in recent years (Chen and Doolen, 1998; Zhang, 2011; Chen et al., 2014). Due to its kinetic intrinsic, the LBM has powerful capacity to treat complex boundaries and thus is particularly suitable for transport in porous media. At present, the LBM has been successfully adopted for investigating a variety of transport phenomena including porous flow, multiphase flow, heat transfer, chemical reaction, turbulent flow, slip flow, non-Newtonian flow and electrokinetic flow (Chen and Doolen, 1998; Zhang, 2011; Chen et al., 2014).

For the coupled multiphase flow and reactive transport processes studied in the present study, multiple sub-processes are involved, and thus the pore-scale numerical model developed must be able to handle the following major fundamental issues: to capture the deformable liquid-gas interfaces, to simulate mass transfer in the multiphase systems, and to treat both homogeneous and heterogeneous chemical reactions. There have been a few numerical studies about pore-scale multiphase reactive transport processes in porous media (Huber et al., 2011; Chen et al., 2013b, 2015a). In our previous studies, several LB models were selected and were delicately coupled to develop an advanced pore-scale model for coupled multiphase flow, single component transport, and reactive transport processes during nuclear waste storage (Chen et al., 2013b). Later, the above single-component multiphase pore-scale model was extended to multicomponent multiphase flow during shale gas exploitation (Chen et al., 2015a). In Ref. Chen et al. (2015a), the mass transport of each component was solved. This, however, is not necessary, because by writing chemical reactions in the canonical form components can be classified into primary and secondary components and only the total concentration of primary components is required to be solved (Kang et al., 2006). Note that in Ref. Kang et al. (2006) only single-phase reactive transport processes were studied. In this study, we further develop and improve our pore-scale multicomponent multiphase flow model (Chen et al., 2015a) based on such scheme to delineate the primary and secondary components. The numerical schemes and LB models are briefly introduced as follows.

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