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Research paper

A fast and robust TOUGH2 module to simulate geological CO_2 storage in saline aquifers

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ABSTRACT

A new TOUGH2 module to simulate geological CO_2 storage (GCS) in saline aquifers is developed based on the widely employed ECO2N module of TOUGH2. The newly developed TOUGH2 module uses a new non-iterative fugacity-activity thermodynamic model to obtain the partitioning of CO_2 and H_2O between the aqueous and gas phases. Simple but robust thermophysical correlations are used to obtain density, viscosity, and enthalpy of the gas phase. The implementation and accuracy of the employed thermophysical correlations are verified by comparisons against the national institute of standards and technology (NIST) online thermophysical database. To assess the computation accuracy and efficiency, simulation results obtained with the new TOUGH2 module for a one-dimensional non-isothermal radial and a three-dimensional isothermal system are compared against the simulation results obtained with the ECO2N module. Treating salt mass fraction in the aqueous phase as a constant, along with the inclusion of a non-iterative fugacity-activity thermodynamic model, and simple thermophysical correlations, resulted in simulations much faster than simulations with ECO2N module, without losing numerical accuracy. Both modules yield virtually identical results. Additional field-scale simulations of CO_2 injection into an actual non-isothermal and heterogeneous geological formation confirmed that the new module is much faster than the ECO2N module in simulating complex field-scale conditions. Owing to its capability to handle CO_2-CH_4-H_2S-N_2 gas mixtures and its compatibility with TOUGHREACT, this new TOUGH2 module offers the possibility of developing a fast and robust TOUGHREACT module to predict the fate of CO_2 in GCS sites under biotic conditions where CO_2, CH_4, H_2S, and N_2 gases can be formed.

1. Introduction

Geological carbon storage (GCS) in deep underground geological formations is a promising method to mitigate emissions of CO_2 into the atmosphere. Among potential geological formations for CO_2 storage, depleted oil reservoirs and deep saline aquifers are of special interest because they provide pore space and trapping conditions for long term storage of CO_2. Four trapping mechanisms keep injected CO_2 underground, namely the structural, residual, solubility, and mineral trapping mechanisms. In the structural trapping mechanism, an impermeable cap rock prevents the upward migration of CO_2 due to buoyancy, in the residual trapping mechanism, disconnected CO_2 droplets are held permanently in small pore sizes by capillary forces, in the solubility trapping mechanism, CO_2 dissolves in the formation water and hydrocarbons, and in the mineral trapping mechanism, CO_2 reacts directly or indirectly with native minerals and organic matter forming immobile carbonate minerals (Hitchon, 1996; Zhang and Song, 2014).

Recent studies on the impact of CO_2 injection on the microbial community of saline aquifers using molecular biology techniques (Basso et al., 2009; Itavaara et al., 2011; Kotelnikova, 2002; Morozova et al., 2010; Mu et al., 2014), have shown that methanogenic microbes and sulfate reducing bacteria (SRB) are capable of adapting to the extreme conditions of GCS. A metabolic shift occurs avoiding the complete inhibition of the microbial community. Similar responses have been observed in oil reservoirs (pH < 5, temperature > 50 °C, salinity > 1.0 mol/L) flooded with CO_2, where the concentration of H_2-forming microbes either increased or remained unchanged (Fujiwara et al., 2006; Liu et al., 2015a, 2015b; Sugai et al., 2012), which imply that in depleted oil reservoirs and deep saline aquifers used for long term storage of CO_2, the fate and trapping of CO_2 may be affected by the activity of indigenous microbial communities (Peet et al., 2015).

Estimations of the long term effect of microbial activity on the fate and trapping of CO_2 in depleted oil reservoirs and deep saline aquifers used for CO_2 storage can be obtained by conducting numerical

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simulations. Several commercial and open source software exist to model and simulate GCS in depleted oil reservoirs and deep saline aquifers (e.g., ECLIPSE© (Schlumberger), GEM© (CMG), VIP-Comp© (Halliburton), and TOUGH2 family of codes (Lawrence Berkeley National Laboratory)). The capabilities of these software can be expanded to account for the transport and kinetic growth of indigenous microbes. This can allow predicting the fate and transport of bioproducts (e.g., CH_4, H_2S and N_2), and organic substrates (e.g., fatty acids, alkanes). However, incorporating microbial kinetic models and increasing the number of independent variables (microbes, bioproducts and substrates) would substantially increase the calculation time of these software. Coupling of reactive and multiphase flow processes is usually done through the application of sequential or iterative methods where kinetic and equilibrium reaction equations along with mass and heat conservation equations are solved for each independent variable iteratively (Vilcáez et al., 2013, 2017, 2008).

To the best of our knowledge, there is not any software to simulate the biogenic conversion of CO_2 to CH_4, and/or the biogenic formation of CO_2-CH_4-H_2S-N_2 gas mixtures from the biodegradation of organic substrates at GCS conditions. Accounting for this possibility is particularly important for depleted oil reservoirs where there is an ample source of organic substrates. In depleted oil reservoirs, microbial activity stimulated by the injection of CO_2 and/or nutrients can affect the fate and trapping of not only CO_2 but also the remaining oil in depleted oil reservoirs. For instance, methanogenic biodegradation of alkanes is a well-known process to occur in oil reservoirs (Dolfing et al., 2008; Jones et al., 2008; Larter and di Primio, 2005; Scott et al., 1994), and its stimulation can be seen as a pathway for a new enhanced oil recovery method (Cai et al., 2015a, 2015b; Jones et al., 2008; Vilcáez, 2015a,b). A new TOUGHREACT module with expanded capabilities accounting for the activity of microbes and transport of their metabolic substrates and products, would be advantageous to assess the fate and trapping of CO_2 under biotic conditions as well as the economic value of stimulating methanogenic biodegradation of alkanes in depleted oil reservoirs.

TOUGHREACT is a numerical simulation program for chemically reactive non-isothermal flows of multiphase fluids in porous and fractured media. It was developed by introducing reactive chemistry into the multiphase flow code TOUGH2 (Xu et al., 2006). Hence, several TOUGHREACT modules could be derived from available TOUGH2 modules to simulate a variety of multiphase and multicomponent reactive transport processes (Pruess, 1991). ECO2N module of TOUGHREACT allows for the simulation of not only the dissolution but also the mineral trapping of CO_2 in deep saline aquifers. The latest version of TOUGHREACT includes microbial capabilities (Xu et al., 2014). However, ECO2N module of TOUGH2 can only handle pure CO_2 (Pruess and Spycher, 2007), limiting the applicability of TOUGHREACT-ECO2N to simulate the fate and trapping of CO_2 under biotic conditions. Accounting for the biogenic formation of CO_2, CH_4, H_2S, and N_2 is important because they can affect the trapping and conversion of both the injected CO_2 and available organic substrates (e.g., alkanes).

Because of the complex numerical schemes needed to couple biogeochemical and multiphase processes and the large number of independent variables which would substantially increase the calculation time of a new TOUGHREACT-ECO2N module, the aim of this research is to reduce the calculation time of the ECO2N module of TOUGH2 and expand its capabilities to simulate the multiphase flow of CO_2-CH_4-H_2S-N_2 gas mixtures. The approach consists of using simple correlations to obtain the thermophysical (density, viscosity and enthalpy) properties of gas mixtures along with a new non-iterative fugacity-activity thermodynamic model which can predict the mutual solubility of CO_2-CH_4-H_2S-N_2 gas mixtures in brine (Shabani and Vilcáez, 2017).

Similar to this research but with different objectives, other TOUGH2 modules have been developed based on other TOUGH2 modules. EOS7Cm was developed based on EOS7C module (Oldenburg et al., 2004) to simulate the flow of CO_2-CH_4-H_2S gas mixtures under high temperature, pressure, and salinity conditions (Lei et al., 2016). EOS7Cm uses an iterative thermodynamic model to solve the mutual solubility of CO_2-CH_4-H_2S gas mixtures in brine. TMGAS is another TOUGH2 module developed based on the TMVOC module to simulate the injection of gas mixtures into deep geological sites (Battistelli and Marcolini, 2009). TMGAS also uses an iterative thermodynamic model to solve the mutual solubility of acid gases (CO_2 and H_2S) in brine. The inclusion of iterative thermodynamic models in simulating the flow of gas mixtures has been proven to result in long calculation times (Battistelli and Marcolini, 2009). Thus, the necessity of using non-iterative thermodynamic models to reduce the calculation time of GCS is evident.

Prior to its coupling to TOUGHREACT, the efficiency of the new module developed based on the ECO2N module of TOUGH2 is verified by conducting simulations of CO_2 injection using synthetic and real one-dimensional (1D) and three-dimensional (3D) geological models.

2. Methodology

In the newly developed module, possible thermodynamic states include a single aqueous phase with or without dissolved CO_2, a single CO_2-rich phase that might be either gas or liquid CO_2 with dissolved H_2O, and a two-phase aqueous-CO_2 rich state. Supercritical CO_2 is treated as a gas phase, and salt (NaCl) is treated as component whose concentration in the aqueous phase remains unchanged over time and space. The possibility of a separate solid salt phase forming due to the precipitation of halite (NaCl) is not considered. This simplification is valid for the co-injection of CO_2 and brine, or the injection of CO_2 into a geological formation containing relatively low salinity formation waters, where microbial activity is relevant and the formation of a solid separate salt phase is not possible and/or can be neglected. This simplification is made based on CO_2 storage capacity enhancements obtained through co-injecting CO_2 and brine into saline aquifers (Rathnaweera et al., 2016), as well as on proposed methods to stimulate microbial methanogenesis in depleted oil reservoirs that involves the co-injection of CO_2 and brine amended with nutrients (Vilcáez, 2015b).

Keeping the approach used in the ECO2N module of TOUGH2, the thermodynamic state of a single-phase system is defined by pressure, CO_2 mass fraction in the aqueous phase, and temperature, whereas the thermodynamic state of a two-phase system is defined by pressure, gas saturation, and temperature. For single-phase isothermal systems, the list of primary thermodynamic variables reduces pressure and CO_2 mass fraction in the aqueous phase, whereas, for two-phase isothermal systems, the list of primary thermodynamic variables reduces to pressure and gas saturation. The equilibrium gas and liquid CO_2 mass fractions at the specified temperature and pressure conditions are calculated using the non-iterative fugacity-activity thermodynamic model of Shabani and Vilcáez (2017).

2.1. Governing equations

TOUGH2 uses the integral finite difference method (IFDM) to solve the mass and heat balance equations for a system with NK components and NPH phases in equilibrium:

$$\frac{d}{dt}\int_{V_n} M^{(i)} \cdot dV = \int_{\Gamma_n} F^{(i)} \cdot n \, d\Gamma + \int_{V_n} Q^{(i)} \quad dV \tag{1}$$

where subscript i is the index for the NK components in the system, M is the mass or energy accumulation term, F is the mass or energy flow term, and Q is the sink/source term of mass or energy.

The mass accumulation per volume of component i is given by:

$$M^{(i)} = \varphi \sum_{\beta=1}^{NPH} S_\beta \rho_\beta X_\beta^{(i)}, \quad i = 1, NK; \quad \beta = 1, NPH \tag{2}$$

where β is the phase index, φ is porosity, S_β is saturation of phase β, ρ_β is density of phase β, and X_β^i is the mass fraction of component i in phase β.