PROCEEDINGS, TOUGH Symposium 2003
Lawrence Berkeley National Laboratory, Berkeley, California, May 12–14, 2003

TOUGHREACT: A NEW CODE OF THE TOUGH FAMILY FOR NON-ISOTHERMAL MULTIPHASE REACTIVE GEOCHEMICAL TRANSPORT IN VARIABLY SATURATED GEOLOGIC MEDIA

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ABSTRACT

Coupled modeling of subsurface multiphase fluid and heat flow, solute transport and chemical reactions can be used for the assessment of acid mine drainage remediation, waste disposal sites, hydrothermal convection, contaminant transport, and groundwater quality. We have developed a comprehensive numerical simulator, TOUGHREACT, which considers non-isothermal multi-component chemical transport in both liquid and gas phases. A wide range of subsurface thermo-physical-chemical processes is considered under various thermohydrological and geochemical conditions of pressure, temperature, water saturation, and ionic strength. The code can be applied to one-, two- or three-dimensional porous and fractured media with physical and chemical heterogeneity.

INTRODUCTION

By introducing reactive geochemistry into the framework of the existing code TOUGH2 (Pruess, 1991; Pruess et al, 1999), we have developed a comprehensive non-isothermal multi-component reactive fluid flow and geochemical transport simulator, TOUGHREACT.

In this paper, we give some brief description of features, processes, and applications of the TOUGHREACT code. More details can be found in the manual (Xu et al., 2003a). The code is planned to be released in 2003 through the US Department of Energy's Energy Science and Technology Software Center (ESTSC). The TOUGHREACT program modules make use of "self-documenting' features. It is distributed with a number of input data files for sample problems. Besides providing benchmarks for proper code installation, these can serve as a self-teaching tutorial in the use of TOUGHREACT, and they provide templates to help jump-start new applications. The fluid and heat flow part of TOUGHREACT is derived from TOUGH2 V2, so in addition to the TOUGHREACT manual, users must have the user's manual of the TOUGH2 V2 program (Pruess et al., 1999).

Like TOUGH2, TOUGHREACT is written in FORTRAN 77. It has been tested on various computer platforms, including PC, SUN Ultrasparc systems, and Compaq Alpha based workstations. On most machines the code should compile and run without modification.

MAJOR PROCESSES

The present model for fluid flow and geochemical transport has three important features: (1) the gas phase is active for multiphase fluid flow, mass transport and chemical reactions, (2) we consider not only porous media, but also reactive fluid flow and transport in fractured rocks, (3) the effects of heat are considered, including heat-driven fluid flow, and temperature-dependent thermophysical and geochemical properties (such as fluid density and viscosity, and thermodynamic and kinetic data).

Transport of aqueous and gaseous species by advection and molecular diffusion is considered in both liquid and gas phases. Any number of chemical species in liquid, gas and solid phases can be accommodated. Aqueous complexation, acid-base, redox, gas dissolution/exsolution, and cation exchange, are considered under the local equilibrium assumption. Mineral dissolution and precipitation can proceed either subject to local equilibrium or kinetic conditions.

Changes in porosity and permeability due to mineral dissolution and precipitation can modify fluid flow. This feedback between flow and chemistry can be important and can be considered in our model, but a rather large computational penalty has to be paid if this is modeled explicitly. Alternatively, the model can monitor changes in porosity and permeability during the simulation from changes in mineral volume fractions without feedback to the fluid flow. Changes in porosity during the simulation are calculated from changes in mineral volume fractions. Several alternative models for the porosity-permeability relationship are included.

We currently neglect deformation of the porous skeleton. Heat effects from chemical reactions are neglected in our current model, as are changes in thermophysical properties of fluid phases (such as viscosity, surface tension, and density) due to changes in chemical composition.

SOLUTION METHOD

TOUGHREACT uses a sequential iteration approach, which solves the flow, transport and reaction equations separately. The flow and transport in geologic media are based on space discretization by means of integral finite differences (IFD; Narasimhan and Witherspoon, 1976). The IFD method gives a flexible discretization for geologic media that allows us to use irregular grids, which is well suited for simulation of flow, transport, and fluid-rock interaction in multi-region heterogeneous and fractured rock systems. An implicit time-weighting scheme is used for individual components of flow, transport, and geochemical reaction. The transport equations are solved independently for each chemical component, whereas the reaction equations are solved on a grid block basis using Newton-Raphson iteration. The quasi-stationary approximation (Lichtner, 1988) and an automatic time stepping scheme are implemented in TOUGHREACT.

MODEL SCOPE AND APPLICATIONS

TOUGHREACT is applicable to one-, two-, or three-dimensional geologic domains with physical and chemical heterogeneity. The code can be applied to a wide range of subsurface conditions. Temperature can range from 0 to 300 ^{0}C, limited by present by available geochemical databases such as EQ3/6 (Wolery, 1992). Pressures can be from 1 bar (atmospheric pressure) to several hundred bars (at several thousand meter depth). Water saturation can range from 0 to 1 (or from completely dry to fully water saturated). The code can deal with aqueous solutions from dilute to moderately saline (ionic strength up to 4 mol/kg H_2O for NaCl dominated).

The TOUGHREACT code was extensively verified against analytical solutions and other numerical simulators (Xu and Pruess, 1998; Xu et al., 1999; Xu and Pruess, 2001). The LBNL group has applied the code to a variety of field scale problems. A small number of external groups, including UNOCAL, EPDC and ExxonMobil, have participated in beta-testing and have used the program for hydrothermal and petroleum reservoir problems. Major TOUGHREACT application examples are summarized in Table 1.

In combination with different fluid property modules, TOUGHREACT is applicable to a variety of reactive geochemical systems, including (1) mineral deposition such as supergene copper enrichment (Problem 1 in Table 1, EOS9), (2) mineral alteration and silica scaling in hydrothermal systems under natural and production conditions (Problem 2, EOS2), (3) assessment of nuclear waste disposal sites (Problems 3 and 4, EOS3 and EOS4), (4**)** sedimentary diagenesis and CO_2 disposal in deep geologic formations (Problem 5, ECO2), and (5) natural groundwater quality evolution and contaminant transport under ambient conditions (Problem 6, EOS9).

ACKNOWLEDGMENT

We are grateful to Guoxiang Zhang and Lijie Han for reviews of this paper. This work was supported by the Assistant Secretary for Energy Efficiency and Renewable Energy, Office of Wind and Geothermal Technologies, of the U.S. Department of Energy, under Contract No. DE-AC03-76SF00098.

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Table 1. List of major TOUGHREACT application examples.

Problem	*Description*	*Size*	*Reaction type*	*Reference*
1. Supergene copper enrichment (SCE)	Oxidative weathering of pyrite (FeS_2) and chalcopyrite ($CuFeS_2$) causes acidification and mobilization of metals in the unsaturated zone and intense alteration of primary minerals, with subsequent formation of enriched secondary copper bearing sulfide mineral deposits (enrichment blanket) in the reducing conditions below the water table.	2-D, 77 grid blocks, 18 minerals, 46 aqueous species, 1 gas (O_2).	Aqueous complexation, Redox, Gas dissolution/exsolution, Mineral dissolution/ precipitation.	Xu et al. (2001)
2. Caprock hydrothermal alteration	The interaction between hydrothermal fluids and the rocks through which they migrate alters the earlier formed primary minerals and leads to the formation of secondary minerals, resulting in changes in physical and chemical properties of the system.	2-D with fracture-matrix interaction, 742 grid blocks, 14 minerals, 23 aqueous species, 1 gas (CO_2).	Aqueous complexation, Gas dissolution/exsolution, Mineral dissolution/ precipitation	Xu and Pruess (2001)
3. Effects of thermohydrology on geochemistry	The problem is investigated for the Drift Scale Test (DST) problem, in which a strong heat source is emplaced in unsaturated fractured volcanic tuffs at Yucca Mountain, Nevada	2-D with fracture-matrix interaction, 3000 grid blocks, 22 minerals, 28 aqueous species, 1 gas (CO_2).	Aqueous complexation, Gas dissolution/exsolution, Mineral dissolution/ precipitation.	Sonnethal and Spycher (2001)
4. Calcite precipitation and infiltration fluxes at Yucca Mountain	Using reaction-transport model to investigate the relationship between percolation flux and measured calcite abundances.	1-D column with fracture-matrix, 130 grid blocks, 22 minerals, 28 aqueous species, 1 gas (CO_2).	Aqueous complexation, Gas dissolution/exsolution, Mineral dissolution/ precipitation.	Xu et al. (2002)
5. Mineral trapping for CO_2 disposal in deep saline aquifers	Simulations were performed for a commonly encountered Gulf Coast sediment under natural and CO_2 injection conditions in order to analyze the impact of CO_2 immobilization through carbonate mineral precipitation.	1-D radial, 130 grid blocks, 22 minerals, 28 aqueous species, 1 gas (Supercritical CO_2).	Aqueous complexation, Gas dissolution/exsolution, Mineral dissolution/ precipitation.	Xu et al. (2003b)
6. Water quality in the Aquia aquifer, Maryland	$NaHCO_3$ type waters in the coastal plain aquifers of the eastern United States have been related to freshening of the aquifer. The water quality in this aquifer shows zonal bands with changes in concentrations of major cations that have been attributed to cation exchange and calcite dissolution/precipitation.	1-D, 16 grid blocks, 1 mineral, 20 aqueous species, 5 exchanged cations	Aqueous complexation, Mineral dissolution/ precipitation, cation change	Xu and Pruess (1998)