An Interdisciplinary Response to Mine Water Challenges - Sui, Sun & Wang (eds)
©2014 China University of Mining and Technology Press, Xuzhou, ISBN 978-7-5646-2437-8

Enhanced in Situ Leaching (EISLEACH) Mining by Means of Supercritical Carbon Dioxide - TOUGHREACT Modeling

Alireza Arab[1], Martin Bertau[2], Helmut Mischo[3], Broder Merkel[4]
1 TU Bergakademie Freiberg, Institut für Geologie, araba@geo.tu-freiberg.de
2 TU Bergakademie Freiberg, InstitutfürTechnischeChemie, martin.bertau@chemie.tu-freiberg.de
3 TU Bergakademie Freiberg, InstitutfürBergbau, helmut.mischo@mabb.tu-freiberg.de
4 TU Bergakademie Freiberg, InstitutfürGeologie, merkel@geo.tu-freiberg.de

Abstract Waste rock piles and mine tailings exert severe impact on the environment. Especially, groundwater is a critical issue when it comes to subsurface mineral extraction. The objective of this paper is to study the physical and chemical impact of $scCO_2$ on carbonate rock as a minable ore reservoir through numerical simulations by means of TOUGHREACT. Carbonic acid is formed when $scCO_2$ gets mixed with water; as a result calcite dissolution occurs. Subsequently, dissolution of calcite creates voids and increases porosity. Even a small increase in porosity has significant effect on enhancement of permeability and thus leaching efficiency.

Keywords in situ leaching, TOUGHREACT, EISLEACH, supercritical CO_2

Introduction

Decrease of grade of near-surface deposits has resulted in mining deeper deposits and excavating, transporting and processing of much larger volumes of rock per unit production recovered. Continuous increase in labor and energy costs is putting even more constraints on the mining industry which in turn enforces higher required geochemical enrichment for the desired ore to be mineable. Some raw materials like rare earth elements (REE) occur in rather low concentrations which makes mining and extraction difficult and costly.

Conventional mining practices and ore processing thus far have shown to considerably affect the environment. They involve large-scale movements of waste rock and vegetation and the creation of tailings which in turn may release toxic compounds into the water. This contaminated water can pollute the region surrounding the mine and beyond. New mining technologies which have improved efficiency, are less waste producing and have minor environmental impact are a necessity.

Based on experimental work conducted at TU Bergakademie Freiberg, a patented approach was proposed for the first time (Arab et al. 2013) to use supercritical CO_2 ($scCO_2$) to increase rock permeability which can be developed into an applicable process technology for extracting raw earth elements (REE). Injection of CO_2 into a deep carbonate layer was investigated using the non-isothermal reactive geochemical transport modeling tool TOUGHREACT. The injected CO_2 will change into supercritical phase when it passes its critical point (31.1 ℃, 7.38 MPa). Under normal geothermal gradient, depth of about 1 km is well above this critical temperature and pressure. When $scCO_2$ is mixed with water existing in the pore spaces, carbonic acid forms which starts to dissolve the carbonate rock and create voids.

To further improve the procedure and increase the extraction capability, a two stage approach is investigated in this study. First, $scCO_2$ is injected into a low grade ore layer. Compared to water, $scCO_2$ has lower viscosity which enables it to enter hairline fissures and disintegrate siliceous, oxidic or sulphidic species and to transform them into the respective carbonates. Carbonates will not react with supercritical carbon dioxide; however, this changes when water is mixed with carbon dioxide, because then the acidic water dissolves carbonates. Therefore, as the second stage, CO_2 is turned off and water is pumped into the layer. The carbonation of e.g. feldspars is accompanied by volume reduction thereby producing additional pathways for the supercritical CO_2 to migrate through fissures generating an enhanced reaction surface for CO_2 to interact with the mineral species of interest. CO_2 mixed with water is able to dissolve carbonates and thus to increase permeability and leach the desired trace elements including REE.

Assuming that a circulation of 10 l/s will be achieved and the solution contain 5 valuable metals at concentrations of 1 ppm, these sums up to 50 g/m^3 and thus 4.32 kg per day. Estimating an average price of 3000 to 8000 € per kg this would result in 12960 to 34560 € value add per day (4.73 to 12.61 million € per year, respectively). At the very moment only rough estimations about the investment and running costs are available. But it is rather likely to produce well beyond the costs of conventional mining plus aboveground processing because costs for labor work are significantly lower.

Method

TOUGHREACT is a non-isothermal reactive geochemical transport model, developed by Xu and Pruess. It was established by introducing reactive geochemistry into the multi-phase fluid and heat flow code $TOUGH_2$ (Pruess 1991). The most recent version of TOUGHREACT provides several EOS (equation of state).

ECO2N EOS was created for CO_2 disposal in deep brine aquifers to simulate fluid flow induced by CO_2 injection into deep aquifers (Pruess and Garcia 2001). ECO_2 provides an accurate description of the thermophysical properties of mixtures of water and CO_2 at conditions that may typically be encountered in deep formations of interest for CO_2 injection ($T > 35$ ℃; $75 \text{ bars} \leq P \leq 400 \text{ bars}$). For analyzing reactive fluid and chemical interaction with rock minerals induced by CO_2 injection, the reactive geochemistry part of the TOUGHREACT code has been combined with $TOUGH_2/ECO_2$, resulting in a new improved reactive geochemical transport simulator $TOUGHREACT/ECO_2$ (Xu et al. 2003).

Temporal changes in porosity and permeability due to mineral dissolution and precipitation can modify fluid flow. This feedback between transport and chemistry can be important and can be considered in the model. Changes in porosity during the simulation are calculated from changes in mineral volume fractions (Pruess et al. 1999).

Space discretization in $TOUGH_2$ and the transport code TOUGHREACT is realized by means of the same integral finite differences (Narasimhan and Witherspoon 1976) because flow and transport equation have the same structure. The IFD discretization approach and the definition of the geometric parameters are shown in Fig. 1. Of course it is as well possible to use any “normal” rectangular finite grid discretization scheme.

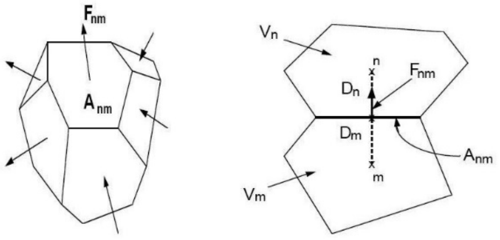

***Fig. 1** Discretization scheme and geometric data according to the integral finite difference scheme (from Xu et al. 2012)*

Model Setup

A TOUGHREACT simulation requires four input files:

- flow.inp- standard TOUGH2 input file and contains the multiphase fluid and heat flow model.
- solute.inp – contains flags and input parameters for calculation of reactive transport.
- chemical.inp – includes chemical parameters and properties.
- thermodb.txt -- The thermodynamic database

To rapidly develop the conceptual model and view results, the PetraSim graphical user interface is used to create a three-dimensional model. PetraSim is an interactive pre-processor and post-processor for the TOUGH family of codes. The carbonate rock ore reservoir is considered as 3 layered (no gravitational effect) with a thickness of 100 m and dimensions of 1 km in X and Y direction at a depth of 2.5 km. Pologonal (Voronoi) mesh is used for the grid system. This type of meshing allows for adequate refinement around the injection well. A total of 1158 cells are used. The injection well is placed at the center of the ore layer (500m, 500m) with a length of 100m (fig. 2). The hydrogeological parameters used in the simulation are shown in table 1.

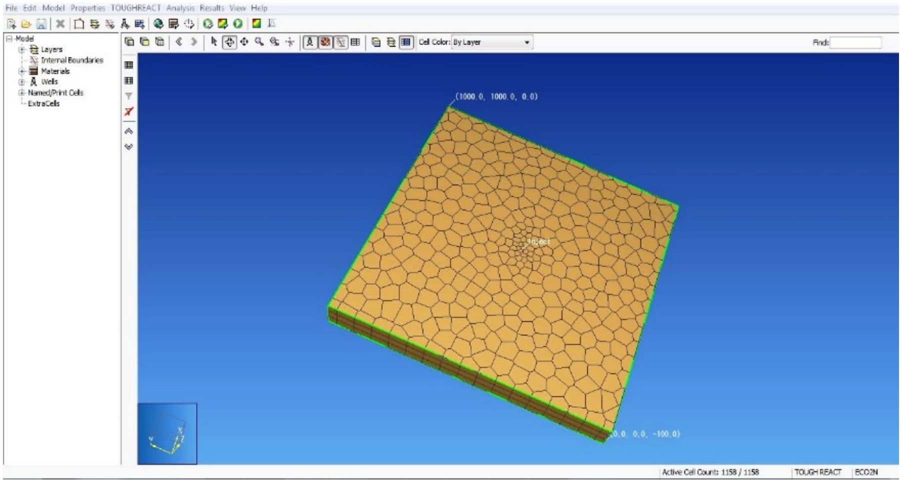

Fig.2 *Carbonate rock layer size and polygonal (Voronoi) grid layout. Injection well is placed in the center of the model. Mesh refinement is implemented around the well.*

Table 1 *Hydrogeologic parameters for the simulation*

Layer thickness	100 m
Permeability	10^{-13} m^2
Porosity	0.12
Compressibility	$1x10^{-8}$ Pa^{-1}
Temperature	75 $^{\circ}$C
Pressure	100 bar
CO_2 injection rate	40 kg/s

In the first stage, CO_2 is pumped into the carbonate layer at a rate of 40kg/s during 70 days. While CO_2 is injected in the carbonate rock layer, pressure and temperature inside the well rises and causes the gas to surpass its critical point and turn into supercritical form as it goes down. As pumping continues, scCO_2 spreads into the layer filling pore spaces and causing increase in pressure throughout the layer.

In the second stage, CO_2 injection is stopped and water is substituted. To do so, the restart feature in TOGHREACT is utilized to load the geochemical condition at the end of stage one as initial conditions for a subsequent run to pump water at a rate of 30kg/s for a period of 70 days.

Results

With a constant injection of $scCO_2$, pressure inside the well and around the well increased from the initial 2×10^7 Pa to 5×10^7 Pa (fig. 3). The area around the well and the area which $scCO_2$ reached had completely dried out and were saturated with $scCO_2$. When water was pumped at a rate of 30kg/s, the water mixed with the injected $scCO_2$ and carbonic acid was formed. Fig. 4 shows the pH value in the ore layer after carbonic acid is formed. Due to the acidic environment the carbonate rock dissolves at a rapid rate. Significant dissolution of calcite occurred in the vicinity of the well due to the presence of carbonic acid (fig. 5). Dissolution of calcite, increases voids and hence increases porosity. Results showed that an increase of 10% in porosity led to an increase of 50% -70 % in permeability especially in the vicinity of the injection well and the areas affected. This increase in porosity and permeability improved fluid flow and also generated enhanced reaction surfaces for the acid to react to the carbonate rock. Mechanical effects on factures due to pressure and porosity increase were not considered.

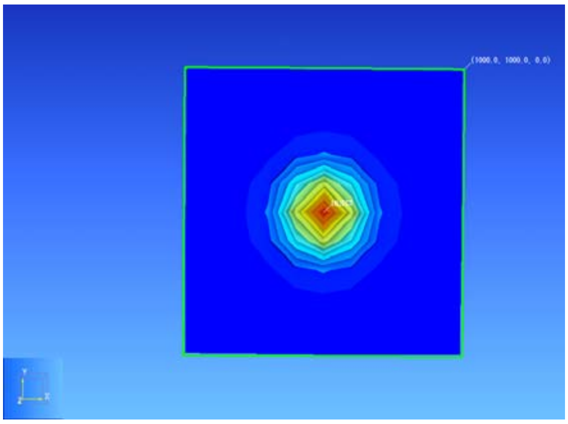

Fig. 3 *Pressure in the injection well reaches to a maximum of* 5×10^7 *a after 70 days of* $scCO_2$ *injection at a rate of 70 kg/s. Initial injection pressure is* 2×10^7 *Pa.*

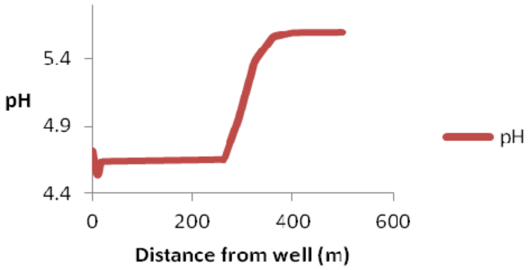

Fig. 4 *pH around the injection well after 70 days of water injection*

Conclusions

The present study investigated the use of supercritical carbon dioxide to enhance in situ mineral extraction from deep ore bodies. A three dimensional model using TOUGHREACT reactive transport code was setup. $scCO_2$ was injected into a deep carbonate layer at a rate of 40 kg/s for a period of 70 days. Afterwards, water was substituted and was pumped at a rate of 30 kg/s for another period of 70 Days. Carbonic acid formed when water mixed with the injected $scCO_2$ and as a result calcite dissolution occurred.

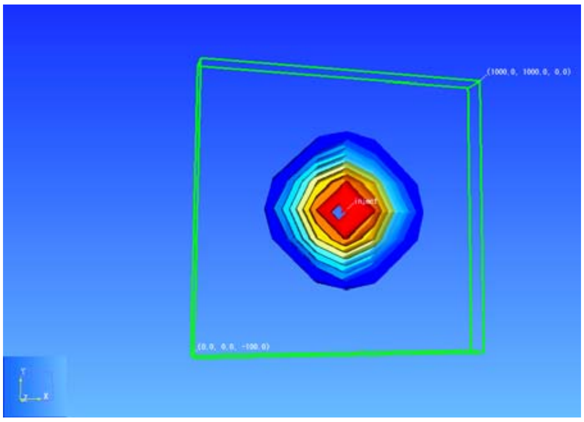

Fig. 5 *Calcite dissolution around the well after 70 days of scCO2 and water injection*

In turn, the dissolution of calcite created more voids and increased porosity. Even a small increase in porosity has a significant effect on permeability enhancement. The increase in porosity and permeability improves fluid flow and also generates enhanced reaction surfaces for the acid to react to the carbonate rock.

With the injection of $scCO_2$ pressure inside the layer increased significantly. This may lead to opening up of existing fissures and also creating new ones which in turn will further enhance porosity and permeability. The $scCO_2$ fracturing technology in combination with CO_2/water extraction has to be investigated and developed as in-situ ore processing with representative examples of different types of rocks and deposits. In-situ fracturing with $scCO_2$ has not been performed until now.

This study showed that the new proposed method of enhanced in-situ leaching mining (EISLEACH) by means of $scCO_2$ has great potential to become the future technology of mining.

References

Arab A, Bertau M, Mischo H, Merkel B (2013) Enhanced in-situ leaching - modeling with TOUGHREACT, WissenschaftlicheMitteilungen 44 Mine Water Symposium Proceedings 73- 80

Narasimhan T N, Witherspoon PA (1976) An integrated finite difference method for analyzing fluid flow in porous media, Water Resources Research 12: 57-64

Pruess K (1991) $TOUGH_2$: A general numerical simulator for multiphase fluid and heat flow. Lawrence Berkeley Laboratory Report LBL-29400, Berkeley, California

Pruess K, Oldenburg C, Moridis G (1999) $TOUGH_{2:}$ User’s guide, Version 2.0. Lawrence Berkeley Laboratory Report LBL-43134, Berkeley, California

Pruess K, García J (2001) Multiphase flow dynamics during CO_2 disposal into saline aquifers. Environmental Geology, Springer 42: 282-295

Xu T, Apps JA, Pruess K (2003) Reactive geochemical transport simulation to study mineral trapping for CO_2 disposal in deep arenaceous formations. Journal of Geophysical Research, 108 (B2): 2071, doi:10.1029/2002JB001979

Xu T, Spycher N, Sonnenthal E, Zheng L, Pruess K (2012) TOUGHREACT User’s Guide: A simulation program for non-isothermal multiphase reactive transport in variably saturated geologic media, version 2.0