COMPARING FRACHEM AND TOUGHREACT FOR REACTIVE TRANSPORT MODELING OF BRINE-ROCK INTERACTIONS IN ENHANCED GEOTHERMAL SYSTEMS (EGS)

Laurent André[1], Nicolas Spycher[2], Tianfu Xu[2], Karsten Pruess[2] & François-D. Vuataz[1]

[1] Centre for Geothermal Research – CREGE, c/o CHYN - Emile Argand 11, CH-2009 Neuchâtel, Switzerland
[2] Earth Sciences Division - Lawrence Berkeley National Laboratory - University of California - Berkeley, CA 94720, USA

laurent.andre@unine.ch, nspycher@lbl.gov, tianfu_xu@lbl.gov, k_pruess@lbl.gov, francois.vuataz@crege.ch

ABSTRACT

Coupled modelling of fluid flow and reactive transport in geothermal systems is challenging because of reservoir conditions such as high temperatures, elevated pressures and sometimes high salinities of the formation fluids. Thermal-hydrological-chemical (THC) codes, such as FRACHEM and TOUGHREACT, have been developed to evaluate the long-term hydrothermal and chemical evolution of exploited reservoirs. In this study, the two codes were applied to model the same geothermal reservoir, to forecast reservoir evolution using respective thermodynamic and kinetic input data. A recent (unreleased) TOUGHREACT version allows the use of either an extended Debye-Hückel or Pitzer activity model for calculating activity coefficients, while FRACHEM was designed to use the Pitzer formalism. Comparison of models results indicate that differences in thermodynamic equilibrium constants, activity coefficients and kinetics models can result in significant differences in predicted mineral precipitation behaviour and reservoir-porosity evolution. Differences in the calculation schemes typically produce less difference in model outputs than differences in input thermodynamic and kinetic data, with model results being particularly sensitive to differences in ion-interaction parameters for high-salinity systems.

INTRODUCTION

This work was initiated through the collaboration between the CREGE (Centre for Geothermal Research – Neuchâtel, Switzerland), and LBNL (Lawrence Berkeley National Laboratory – Earth Science Division – Berkeley, California, USA). These two institutions have developed reactive transport simulators applicable to geothermal systems.

The Centre of Hydrogeology of Neuchâtel and now the CREGE have been involved since 1998 in the European Soultz EGS project (Alsace, France), for which the main objective is to build a pilot plant for power production based on the circulation of a geothermal fluid through a deep fractured reservoir. Different studies (Durst, 2002; Bächler, 2003, Rabemanana et al. 2003; André et al., 2005; Bächler and Kohl, 2005) have progressively allowed the development of a geochemical code able to take into account the main characteristics of the Soultz reservoir at a depth of 5 km: 200°C, 500 bars and a fluid salinity of 100 g L^{-1} (ionic strength around 1.8 molal). The resulting code is called FRACHEM.

LBNL developed TOUGHREACT (Xu and Pruess, 2001; Xu et al., 2004) by introducing reactive transport into the existing code TOUGH2 (Pruess et al., 1999). TOUGHREACT has been applied to different problems such as CO_2 sequestration in deep saline aquifers and the prediction of the underground thermal, hydrological, and chemical evolution around nuclear waste geologic repositories. Recent developments of this code included the addition of the Pitzer formalism (Pitzer, 1973) for the computation of activity coefficients in highly saline brines (Zhang et al., in prep).

The aim of this collaboration between CREGE and LBNL, working in similar fields with similar tools, was both to exchange ideas between the two laboratories and to compare the characteristics of the two codes. For best comparison, the two codes were applied to the same geothermal problem: the circulation of cooled brine in a hot porous-equivalent reservoir similar to that at Soultz, between two wells spaced 650 m apart. However, the model conceptualisation was somewhat different with the two codes. Closed-loop injection of fluid under near-reservoir conditions was simulated with FRACHEM, whereas open injection of the same reservoir fluid was simulated with TOUGHREACT (constant fluid composition, without previous circulation in the reservoir). Simulated processes within the reservoir included mineral precipitation/dissolution and changes in reservoir porosity.

The circulation of cooled fluid in the reservoir affects its temperature and involves a thermodynamic disequilibrium between rocks and fluid, resulting in the dissolution and precipitation of several minerals. Mineral precipitation and dissolution have an impact on the evolution of reservoir porosity. The two codes and respective thermodynamic and kinetic input data, as well as activity coefficient models, yield significant divergences, most particularly with respect to the computed solubility of carbonate minerals (mainly calcite) and CO_2 partial pressure. Two reasons can be advanced to explain these differences. First and foremost, activity coefficients are computed using different formulations (Pitzer versus extended Debye-Hückel) and/or different sources of ion interaction parameters, thus generating differences in the predicted solubility and precipitation behaviour of calcite, underlining the importance of these data for reactive transport simulations involving saline reservoirs. Second, differences of model conceptualisation (i.e. simulation of closed-loop versus open injection with FRACHEM and TOUGHREACT, respectively) also affect model results, however to a much lesser extent.

THE CODES

FRACHEM

FRACHEM is a THC simulator issued from the combination of two existing codes: FRACTure and CHEMTOUGH2. FRACTure is a 3-D finite-element code for modelling hydrological, transport and elastic processes. It was developed originally for the study of flow-driven interactions in fractured rock (Kohl & Hopkirk, 1995). CHEMTOUGH2 (White, 1995) is a THC code developed after the TOUGH2 simulator (Pruess, 1991), a 3-D numerical model for simulating the coupled transport of water, vapor, non-condensable gas and heat in porous and fractured media. CHEMTOUGH2 presents the possibility to transport chemical species and to model the chemical water-rock interactions as well as the chemical reactions driven by pressure and temperature changes. The transport and reaction are coupled using a one-step approach.

FRACHEM has been built by introducing geochemical subroutines from CHEMTOUGH2 (White, 1995) into the framework of the code FRACTure (Bächler, 2003; Bächler and Kohl, 2005). After an initialisation phase, FRACTure calculates, over each time step, the thermal and hydrological conditions within each element volume and determines the advective flow between each of them. Resulting thermal and hydrological variables are stored in arrays common to FRACTure and the geochemical modules. At this point the program calculates the chemical reactions using a mass balance/mass action approach, the advective transport

of chemical species and the variations of porosity and permeability. Once this calculation is performed, the porosity and permeability are updated and fed into the FRACTure part of the code. The program then returns to the start of the loop until the end of the simulation time (sequential non-iterative approach, SNIA).

FRACHEM has been developed specially for the granitic reservoir of Soultz-sous-Forêts and consequently, specific implementations have been added to the chemical part of this code. The reservoir, at a depth of 5000 m, contains a brine with total dissolved solids (TDS) about 100 g kg^{-1} and a temperature of 200°C. Considering the high salinity of the geofluid, the Debye-Hückel model, initially implemented in the CHEMTOUGH2 routines to determine the activity coefficients, has been replaced by a Pitzer activity model. It should be mentioned here that the activity coefficients calculations are carried out in an indirect manner by means of another code, TEQUIL (Moller et al., 1998). For a given fluid composition (constant ionic strength), the activity coefficients are determined at different temperatures in the range 50 - 200°C using TEQUIL before running FRACHEM. The activity coefficient values obtained at each temperature are regressed as a function of temperature using a polynomial fit, with coefficients then entered into the chemical input file. This approach works well for the case of Soultz simulations because the ionic strength of the circulated fluid remains more or less constant.

Presently, a limited number of minerals are considered, which correspond to the minerals constituting the Soultz granite. The precipitation/ dissolution reactions of carbonates (calcite, dolomite), quartz, amorphous silica, pyrite and some aluminosilicates (K-feldspar, albite, illite) can be modelled under kinetic constraints. The implemented kinetic rate laws are specific to each mineral and taken from published experiments conducted at high temperature in NaCl brines. Thermodynamic data (equilibrium constants) are taken mostly from SUPCRT92 (Johnson et al., 1992) and Helgeson et al. (1978) and are function of temperature and pressure. At last, a supplementary module allows the determination of porosity and permeability variations linked with chemical processes occurring in the reservoir. Considering the alteration of the Soultz granite, the flow is assumed to circulate in a medium composed of fractures and grains. Therefore, a combination of fracture model (Norton and Knapp, 1977; Steefel and Lasaga, 1994) and grain model (Bolton et al., 1996) is used to determine the permeability evolution.

TOUGHREACT

TOUGHREACT (Xu and Pruess, 2001; Xu et al., 2004) was developed by introducing multi-

component reactive transport into the framework of the existing multi-phase 3-D finite volume fluid and heat flow code TOUGH2 (Pruess, 1991). It is a THC simulator applicable to a wide range of subsurface conditions and to a variety of reactive fluid and geochemical transport problems. Flow, transport, and chemistry are coupled in a sequential manner. Here, a sequential non-iterative approach was applied for consistency with the FRACHEM simulations.

TOUGHREACT takes into consideration many processes such as 1) fluid flow in both liquid and gas phases occurring under pressure, viscous, and gravity forces; 2) heat flow by conduction and convection; 3) diffusion of water vapor and air; 4) thermophysical and geochemical reactions as a function of temperature, such as fluid (gas and liquid) density and viscosity, and thermodynamic and kinetic data for mineral-water-gas reactions; 5) transport of aqueous and gaseous species by advection and molecular diffusion in liquid and gas phases, respectively; 6) temporal changes in porosity, permeability, and unsaturated hydrologic properties owing to mineral dissolution, precipitation and clay swelling.

Geochemical computations are carried out using a mass balance/mass action approach. By default, activity coefficients are computed using an extended Debye-Hückel model (Helgeson et al., 1981) applicable to NaCl-dominant, moderately saline solutions. Recently, a full Pitzer ion-interaction model was implemented as an option, using the formulation of Harvie et al. (1984) (Zhang et al., in prep; see also Zhang et al., 2004). The themodynamic database used for the TOUGHREACT simulations presented here makes use of equilibrium constants mostly from SUPCRT92 (Johnson et al., 1992) and ion-interaction parameters re-evaluated and fitted as a function of temperature by Wolery et al. (2004) (as published by Alai et al., 2005). Other thermodynamic and kinetic data are also functions of temperature.

Mineral dissolution and precipitation can proceed either subject to local equilibrium or kinetic conditions. For kinetically-controlled mineral dissolution and precipitation, a general form of rate law (Lasaga, 1984; Steefel and Lasaga, 1994; Palandri and Kharaka, 2004) is used.

Changes in porosity during the simulation are calculated from changes in mineral volume fractions. Several porosity-permeability and fracture aperture-permeability relationships are included in the model. Here, fracture porosity is related to permeability using the relationship proposed by Verma and Pruess (1988) and described in Xu et al. (2004).

APPLICATIONS

The two codes have been applied to a 2-D geometrical model representing the granitic reservoir at Soultz. Injection and production wells are linked by permeable fracture zones surrounded by low-permeability granite matrix. Each fracture zone has an aperture of 0.1 m, a fixed horizontal depth of 10 m and a porosity of 10% (Figure 1).
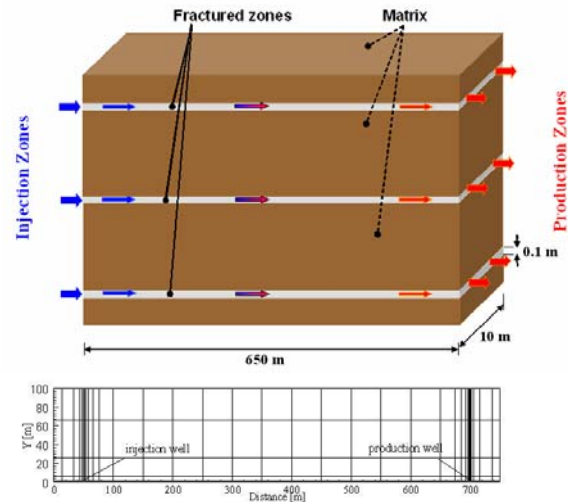

Figure 1: *Simplified model and spatial discretization.*

It is assumed that the fluid exchange with the surrounding low permeability matrix is insignificant. Because of symmetry, it is sufficient to consider only the upper part of one fracture zone in the simulation (Figure 1).

It should be noted that, with FRACHEM, matrix gridblocks are added above the fracture block for proper thermal behaviour. The modelled area is discretized into 222 2-D elements (Figure 1): 25 for the fracture zone and 197 for the matrix. The size of the element is ranging from a minimum of 0.5 m x 0.05 m near the injection and the production wells to a maximum of 50 m x 35 m. With TOUGHREACT, however, only the fracture zone is modelled, without adjacent matrix blocks. Heat loss in the impermeable matrix is modelled by a semi-analytical solution (Vinsome and Westerveld, 1980) built into the code. As a result, the model contains only 25 elements.

Initially the system temperature was set to the reservoir temperature of 200°C. The geothermal fluid (Table 1) was injected in the fracture zone at a rate of 2×10^{-2} L s^{-1} at a constant temperature of 65°C. A constant overpressure of 8 MPa was assumed at the injection well. With FRACHEM, Dirichlet boundary conditions were applied to the upper, left and right side of the model. The fluid was continuously recirculated from the production well to the injection well. With TOUGHREACT, the fluid was not

recirculated, and constant boundary conditions were simulated by connecting gridblocks of infinite dimensions to the injection well (constant chemical composition, temperature and pressure) and to the production well (constant temperature and pressure).

Table 1: Mean composition of fluid coming from the reservoir at a depth of 5 km.

Species	Concentration [mmol/kg$_{H2O}$]
Na^+	1148
K^+	73.4
Ca^{2+}	169.5
Mg^{2+}	3.2
Cl^-	1648
S	1.7
C	42.5
Fe^{2+}	2.6
SiO_2	6.1

Due to the sensitivity of the sequential non-iterative approach (SNIA) method to time discretization, the time step used for this simulation was limited to 10^2s. The values of thermo-hydraulic parameters considered in the simulation are listed in Table 2.

Table 2: Thermo-hydraulic model parameters.

Parameters		Fracture	Matrix	Fluid
Permeability	[m^2]	5x10^{-13}	10^{-18}	-
Thermal conductivity	[W/m.K]	2.9	3	0.6
Density	[kg/m^3]	-	2650	1000
Heat capacity	[J/kg.K]	-	1000	4200
Porosity	[%]	10	2	-

EFFECTS OF CHEMICAL PARAMETERS

Before presenting results of reactive transport simulations, we evaluate the factors most likely to affect chemistry results: the activity coefficients of dissolved species, the mineral equilibrium constants, and the minerals reaction rates.

The equilibrium constants used in the respective databases are not all derived from the same sources but a reasonably good agreement is observed for the minerals included in the simulations, including carbonates (calcite and dolomite) and silica phases (quartz and amorphous silica). Only small divergences are observed for aluminium silicates.

The reaction rates of calcite, quartz and amorphous silica present a good agreement. For aluminosilicates (K-feldspar, albite and illite), the differences can reach about two orders of magnitude, but this can be explained easily by the fact that, with FRACHEM, the inhibitor effect of Na^+ was taken into consideration, whereas it was not TOUGHREACT simulations.

Of greatest importance here, with a concentrated fluid, is the calculation of activity coefficients, which affect ion activities and consequently the saturation indices of minerals. The effect is most significant with minerals including divalent cations, such as calcium and magnesium carbonates, which have a strong effect on pH. Because we initially observed large differences in pH values computed with FRACHEM and TOUGHREACT (up to 1.5 pH units), we decided to run benchmark simulations for a case of simple cooling without mass transfer to minerals or gases (homogeneous equilibrium) and no transport. This allowed the results of several other popular geochemical codes and thermodynamic databases to be compared with one another for the exact same problem.

A Soultz-like brine, with pH = 4.9 at 200°C was numerically cooled from 200 to 20°C without considering reaction with minerals. The evolution of the brine pH and its saturation with respect to calcite was evaluated using the following different models and databases.

• PHREEQC 2.12 (Parkhurst and Appelo, 1999) with the "phreeqc" (Debye-Hückel) and "pitzer" databases (phreeqc.dat 431 2005-08-23 and pitzer.dat 2005-11-16, as released with version 2.12)

• TEQUIL, incorporating a Pitzer activity coefficient model and data for the Na-K-H-Ca-Cl-SO$_4$-HCO$_3$-CO$_3$-CO$_2$-H$_2$ system (Møller et al, 1998). As noted earlier, TEQUIL results are used by FRACHEM to compute activity coefficients, so FRACHEM was not used directly in this benchmarking exercise.

• TOUGHREACT, two versions including 1) the released version 1.0 (Xu et al., 2004), making use of the extended Debye-Hückel activity coefficient model of Helgeson et al. (1981) and thermodynamic data from the EQ3/6 database "data0.ymp", and 2) an unreleased version using the Pitzer activity coefficient model and the EQ3/6 database "data0.ypf" (Wolery et al., 2004, as published in Alai et al., 2005).

• EQ3/6 8.0 (Wolery and Jareck, 2003) with 1) the "b-dot" extended Debye-Hückel model of Helgeson (1969) and database "data0.ymp"; 2) the Pitzer model and database "data0.hmw" (after Harvie et al., 1984); and 3) the Pitzer model and "data0.ypf" database (Wolery et al., 2004, as published in Alai et al., 2005).

• SOLVEQ/CHILLER (Reed, 1998; Reed and Spycher, 1998), which incorporates the extended Debye-Hückel model of Helgeson et al. (1981) and the same equilibrium constants as used with TOUGHREACT.

All these codes were applied to the same fluid composition (ionic strength ~ 1.8 molal, Na-Cl dominated Soultz-like fluid), except for the total H$^+$ concentration which initially needs to be computed by each code from the input brine pH of 4.9 at 200ºC.

The cooling of the solution induces shifts in the equilibrium between aqueous species as a result of the temperature effect on equilibrium constants and activity coefficients. Note that the equilibrium constants for calcite were nearly the same in all the databases considered. It was also verified that only those secondary aqueous species called for by the different activity coefficient models were included in simulations.

Significantly different results were obtained. Figures 2 and 3 present the most remarkable parameters: pH and saturation index of calcite. Some variations were expected, in particular the differences between the simulations using the Debye-Hückel model and those using the Pitzer formalism. More surprising are differences between results of simulations making use of the same methodology for computing activity coefficients. Using the Pitzer formalism, major divergences appear with the use of different ion-interaction parameters. Upon cooling, a pH increase is predicted with EQ3/6 when the "data0.ypf" Pitzer database is used, but a decrease similar to that computed with TEQUIL is predicted when the "data0.hmw" is used. Because the TOUGHREACT Pitzer version was used with the EQ3/6 "data0.ypf" database, the two codes are in very close agreement when using this database.

In contrast, results from the other codes and databases all predict a pH decrease (at least initially), with differences reaching more than 2 pH units at 20°C (Figure 2). Consequently, computed saturation indices of minerals such as calcite vary tremendously (Figure 3), in three cases leading to supersaturation with respect to this mineral and a change from retrograde to prograde solubility above ~110ºC, and in most other cases yielding undersaturation and the usual retrograde solubility behaviour.

These results illustrate the importance of input thermodynamic data in geochemical computations, and particularly those affecting activity coefficients when dealing with concentrated solutions. When using the Pitzer formalism, the types of ion-interaction parameters included in simulations (binary, ternary, cation-anion, etc.) and their variation with temperature are critical. When using simpler Debye-Hückel expressions, extrapolations to higher ionic strengths present significant risks. In the present case, differences shown in Figures 2 and 3 are still being investigated. In some databases, some of the ion-interaction parameters are set to 0, or fixed with temperature. "Double counting" between interaction parameters and secondary species is also a possibility. Even with the simpler Debye-Hückel model, consistency between the activity coefficient model and the types and dissociation constants of secondary aqueous species is critical. At this time, for this particular example, we tend to give preference to the results of TEQUIL and those of EQ3/6 (Pitzer version) with the "data0.hmw" database, although we cannot discard the other results until further study. Obviously, differences in calcite solubility behaviour such as those shown on Figure 3 have direct implications on evaluations of long-term reservoir productivity at Soultz, as described in the next sections.
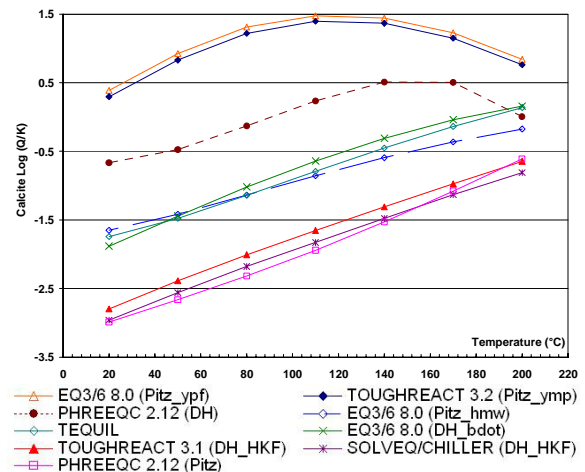
Figure 3: Predicted saturation index of calcite during the cooling from 200 to 20°C of a brine with ionic strength of 1.8.

REACTIVE TRANSPORT RESULTS

Simulations with FRACHEM (Pitzer model of TEQUIL)

The observation of the mineral behaviour shows that all the reactions occur in the first 20 metres of the injection zone. Calcite, a secondary mineral, present within granite fractures in relatively small proportions, is the most reactive.
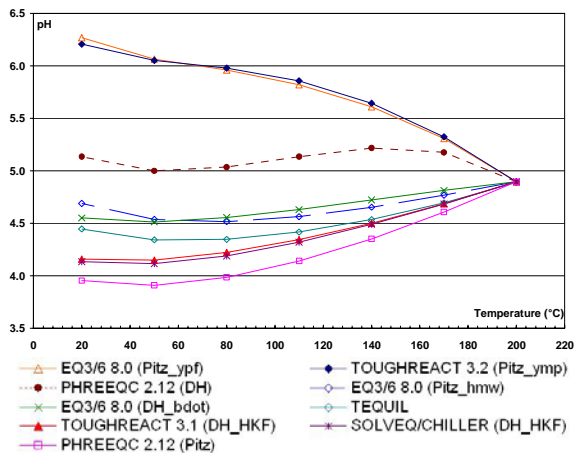
Figure 2: Calculated pH evolution of a brine with ionic strength of 1.8 during its cooling from 200 to 20°C. Variations are observed for different geochemical codes and databases (DH: Debye-Hückel; Pitz: Pitzer; see text)

In the vicinity of the injection well, calcite dissolves whereas it precipitates from about 2 to 20 m. This is due to the retrograde solubility of calcite (solubility decrease with temperature increase). At the onset of fluid circulation within the reservoir, calcite dissolves mainly within the first two metres of the injection well. This dissolution releases calcium in solution which is then available for calcite precipitation further away from the injection well where the temperature increases. With increasing simulation times and decreasing rock temperatures, the dissolution of calcite extends towards the production well and ends when this mineral becomes depleted.

Another carbonate has also been investigated. Figure 4 shows that dolomite dissolves within the first ten metres from the injection well. Similarly to calcite, dolomite dissolution stops when this mineral becomes depleted.

Among silicates, quartz and K-feldspar are major minerals in granite. Contrary to calcite, the solubility of these minerals decreases with cooling. As a consequence, these minerals precipitate near the injection well (note that the fluid always remains undersaturated with respect to amorphous silica in this case) and less so further away as temperature increases.

After 25–30 metres along the fracture, the fluid becomes essentially unreactive with all the minerals shown in Figure 4 as the injected brine becomes close to equilibrium with these minerals.
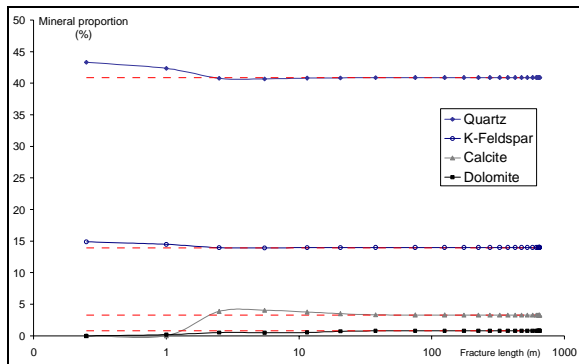

Figure 4: Predicted mineral precipitation and dissolution with FRACHEM after 5 years of circulation (volume percent). The dashed lines correspond to the initial amounts.

Simulations with TOUGHREACT and the Extended Debye-Hückel model

The carbonates are the most reactive minerals. As presented in the case of the FRACHEM simulations, calcite and dolomite dissolve near the injection well (Figure 5).
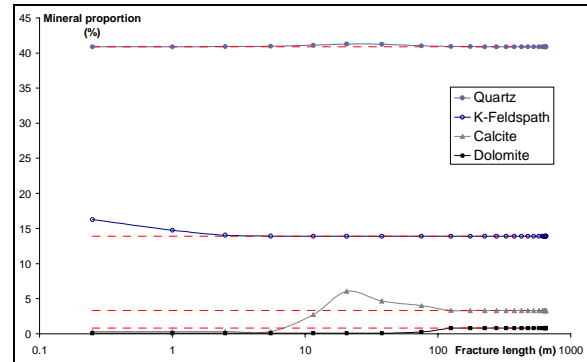

Figure 5: Predicted mineral precipitation and dissolution with TOUGHREACT and extended Debye-Hückel model, after 5 years of injection (volume percent). The dashed lines correspond to the initial amounts.

These minerals are predicted to dissolve faster than in the FRACHEM simulations because of small differences in reaction rates. Calcite dissolves for the first 10 metres (when temperature is below 145°C), then precipitates between 10 and 100 metres from the injection well. Dolomite dissolves within the first 100 metres and remains unaffected further out. Quartz precipitation is negligible, whereas feldspars precipitate in the close vicinity of the injection well.

Simulations with TOUGHREACT and the Pitzer model

The behaviour of silicates is mostly similar to preceding results obtained with the Debye-Hückel model and with FRACHEM. However, the behaviour of calcite is notably different. We showed earlier that cooling simulations using the extended Debye-Hückel model, and the FRACHEM simulations, yield a low pH at 65°C (4–4.5 range) (Figure 2) and consequently undersaturation with respect to calcite at this temperature (Figure 3). In contrast, the Pitzer database used here (Wolery et al., 2004) yields a significantly higher predicted pH (near 6.2) in the fluid cooled to 65°C and, as a result, significant supersaturation with respect to calcite (Figure 3). Consequently, the injection of this solution in the system leads to important calcite precipitation along the first 10 metres of the fracture zone and a more moderate precipitation between 10 and 400 metres from the injection well (Figure 6).
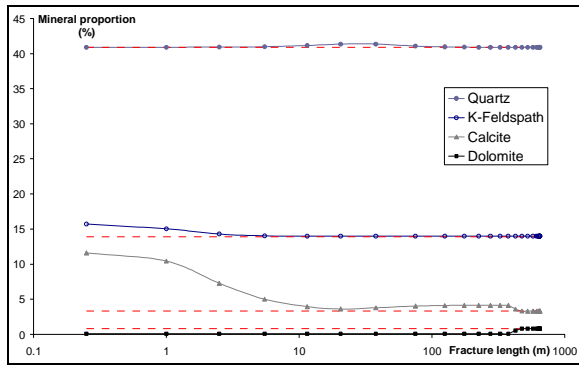
Figure 6: Predicted mineral precipitation and dissolution with TOUGHREACT and the Pitzer model, after 5 years of injection (volume percent). The dashed lines correspond to the initial amounts.

Implications on predicted reservoir porosity

The predicted evolution of reservoir porosity (Figure 7) is determined according to the mineral reactions occurring in the reservoir. As expected from the results presented above, the three models give rather different results.
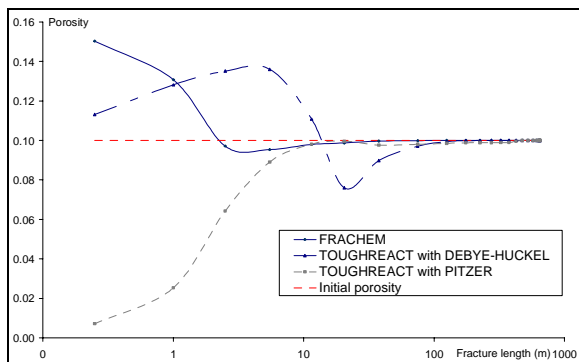

Figure 7: Evolution of the reservoir porosity after 5 years of circulation.

In the FRACHEM simulation, the porosity increases near the injection well due to the dissolution of carbonates (calcite and dolomite). It increases by 30% in the first metre from the injection well before decreasing quickly between 1 and 10 metres because of calcite reprecipitation. The predicted maximum porosity decrease is close to 6%.

With the Debye-Hückel TOUGHREACT model, the general trend is somewhat similar to that predicted with FRACHEM. The porosity increases near the injection well and in the first ten metres of the fracture zone, due to carbonates dissolution. Calcite precipitation between 10 and 100 metres decreases the porosity within this interval by a maximum of about 25%. Feldspar precipitation near the injection well (Figure 5) reduces the magnitude of porosity increase near the well. At first, porosity increases to ~0.14 due to carbonates dissolution and then, when

carbonates have disappeared, silicates precipitate, resulting in a porosity decrease.

With the Pitzer TOUGHREACT model, the porosity evolves differently. Calcite precipitation leads to a large decrease of porosity within the first ten metres from the injection well. Because the same fluid (in this case supersaturated with respect to calcite at 65ºC) is continuously injected in the system, calcite precipitation never stops. The small amount of dolomite dissolution in this zone (Figure 5) cannot compensate for the porosity reduction caused by calcite precipitation and, after five years of circulation, the porosity at the injection well head becomes close to zero.

It should be noted that these respective evolutions of porosity are also affected by the simulated injection mode. In the FRACHEM simulation, the injected fluid circulates in a closed loop; the brine is heated in the system in contact with rock, and then cooled to 65°C before injection. In the TOUGHREACT simulations, the same fluid (initially equilibrated with the reservoir) is continuously injected. This difference in conceptualization affects the behaviour of silicates. These minerals precipitate near the injection well because of the temperature decrease. However, no significant dissolution of silicates occurs within the reservoir (slower dissolution than precipitation), such that the amounts of dissolved silica and aluminium initially deposited near the injection wells are never fully replenished when the brine circulates through the reservoir. Consequently, less silicate precipitate close to the injection well in the case of closed circulation, compared to open injection. This effect is most visible after injection periods longer than five years, and is negligible compared to the large difference in calcite precipitation behaviour between the models.

CONCLUSIONS

The goal of this work was to compare two geochemical transport codes, FRACHEM and TOUGHREACT, to model complex systems like geothermal reservoirs. Significant differences in model results were found to be caused primarily by differences in activity coefficient models and their parameters.

First, this study highlights the importance of the databases in the results of geochemical modelling in particular for brines. Depending on the implemented activity coefficient models and sources of input parameters for these models, variations reaching 2 units have been observed in the predicted brine pH. Such pH differences have a strong impact on saturation indices of minerals such as carbonates. In the FRACHEM simulations (implementing a Pitzer model) and those with an extended Debye-Hückel TOUGHREACT model, significant undersaturation

is predicted with respect to calcite. In contrast, the Pitzer TOUGHREACT model together with a newly acquired Pitzer database results in strong supersaturations. These differences have a tremendous influence on the predicted evolution of reservoir porosity. In the first case, the porosity is predicted to increase due to the dissolution of carbonates (calcite and dolomite) near the injection well, then slightly decrease further away because of calcite reprecipitation. In the other case, calcite is predicted to precipitate within the first ten metres of the injection well, to the point of nearly plugging off the injection zone.

It is difficult for the time being to forecast what will be the real evolution of the reservoir. A circulation test was performed in 1997 within the shallow reservoir at 3500 m at Soultz-sous-Forêts. The initial reservoir temperature was only 165 °C but the mineral composition of the granite was very similar to the one of the deep reservoir (5000 m). During this circulation of 140 days, the pressure at the injection well decreased, indicating an increase of the injectivity around the injection well. This process is probably due to the dissolution of carbonates, the most reactive minerals. This scenario is supported by FRACHEM simulations, as well as by simulations using the currently released TOUGHREACT version. Further work is underway to evaluate currently available ion interaction parameters required for implementing the Pitzer model with the codes discussed in this study.

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