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GEOCHEMICAL MODELLING OF THE SOULTZ-SOUS-FORETS HOT DRY ROCK SYSTEM. BRINE ROCK INTERACTIONS IN A DEEP HOT FRACTURED GRANITE RESERVOIR

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ABSTRACT

The European Hot Dry Rock program is located in the Rhine Graben at Soultz-sous-Forêts, in the north-eastern part of France. A future pilot plant for generating electricity will be built on the basis of three 5,000 m deep wells, one for injection and two for production, all drilled in fractured granite and reaching a bottom hole temperature of 200°C. During a previous phase of the project, in 1997, a four-month circulation test was conducted between two boreholes (GPK1, 3590 m and GPK2, 3876 m), separated from each other by a horizontal distance of 450 m in hydraulically fractured granite. A hydraulic connection between the two wells was proven and a production flowrate was kept between 20 and 25 kg/s. The wellhead temperature reached 142°C, whereas the produced fluid and gases were totally reinjected after cooling at 65 °C.

The chemical data obtained from this test as well as the mineralogy given by core samples have been used to build a numerical model of the water-rock interactions in the reservoir. The high salinity of the fluid (TDS = 100 g/l) and the reservoir temperature (165°C) preclude the use of a classical geochemical model. Therefore a new model based on a Pitzer approach has been developed and introduced in the code CHEMTOUGH.

Thermodynamic equilibrium simulations show that, at the vicinity of the bottom of the injection well and during one year of fluid circulation at a rate of 25 kg/s, a maximum of 250 tons of calcite and 290 tons of dolomite are potentially dissolved within the reservoir, but 100 tons of quartz and 40 tons of pyrite can be potentially precipitated.

Kinetic simulation however indicates that the carbonates behaviour is the most relevant process for permeability variations during the time scale considered here (30 years). Simulations carried out for a 1-D fracture during a period of flow between 1 and 12 months do no display significantly different results. Calcite, dolomite and quartz reaction rates allowed calculating respective porosity increase or decrease in the system. This approach is still under work and the geochemical section of the program will then be coupled with the thermo-hydraulic code FRACTure. This procedure will allow to simulate the flow evolution within the reservoir and to estimate the variation of the system impedance, as well as the production fluid temperature.

Keywords: water-rock interaction, geochemical modelling, reservoir modelling, fractured granite, Hot Dry Rock, Soultz-sous-Forêts.

INTRODUCTION

This study is carried out in the frame of the European research programme Joule 4, called European concerted action for the support of Hot Dry Rock geothermal energy R&D activities 1998-2001. This programme takes place in Soultz-sous-Forêts, northern Alsace, France, on the western edge of the Rhine Graben, about 50 km north of Strasbourg. This program started in 1987 with the aim to extract energy from a hydraulically fractured, hot, granitic reservoir. No fresh water was injected after the stimulation experiments and only the highly saline formation fluid was circulated (Baumgärtner et al., 1998).

A 4-month flow test was carried out in 1997 to allow fluid circulation between GPK1 and GPK2 boreholes at a depth of 3200 to 3600 m. At their maximum depth, the two vertical wells are separated by 450 m. The flow rate was maintained at a rate between 20 and 25 l/s and the rock temperature ranged between 150 and 170°C. Wellhead temperature reached 142°C and all the fluid was cooled to about 65°C, without degassing, before reinjection at depth. Since this test, the former production well (GPK2) was deepened to 5,000m where the rock temperature reaches 200°C (Gérard et al., 1999).

As part of the Swiss contribution to the Soultz programme, the project on which this paper is based is aimed toward the coupling of geochemical fluid-rock interactions to thermal, hydraulic, and mechanical processes. As the final goal of the Soultz programme is to build and to test a pilot geothermal power plant, it is necessary to forecast the probable behaviour of the reservoir during exploitation. Prevision can be realised by numerical modelling as well as by laboratory experiments. In addition to temperature changes and thermo-mechanical contractions, a full model should also include geochemical fluid-rock interactions. This would allow an evaluation of both scaling and corrosion risks as well as permeability decrease or increase within the fractured reservoir resulting from mineral deposition or dissolution (Durst and Vuataz, 2000). Our goal is to set up a kinetic geochemical model of the fluid-rock interactions for the Soultz reservoir and then to couple some geochemical modules to the thermo-hydraulic simulator FRACTure in collaboration with the team of the Institute of Geophysics from ETH-Zurich (Kohl and Hopkirk, 1995).

THERMODYNAMIC MODELLING

Geochemical interactions in the Soultz-sous-Forêts fractured reservoir were described by Azaroual (1992) and Aquilina et al. (2000). It has been shown that the fluids originate from a brine in Triassic sediments at the border of the Rhine Graben, diluted by a mature meteoric water that dissolves micas and sulphates in the Triassic Buntsandstein and in the granitic basement. Then, the fluids percolate into fractured granites, dissolving plagioclases and precipitating secondary minerals in veins such as quartz, illite, montmorillonite, calcite, dolomite and pyrite. Fluid-rock interaction modelling in the fractured reservoir of Soultz was studied by Jacquot (1998 and 2000), who pointed out the importance of the knowledge of mineral assemblages in contact with the fluid during the circulation. Indeed, the fluid flowing into the fractures is not in contact with fresh granite but with altered granite and newly formed minerals, such as clay minerals, representing almost 40% of this assemblage volume.

The first problem arising in modelling the geochemical behaviour of the Soultz brine is the lack of chemical data. Samples of the 1997 circulation test were completely analysed only 1.5 years after sampling and cannot inform about the original dissolved CO_2 and the distribution between sulphates and sulphides. Physico-chemical parameters such as redox potential and pH, even if measured in situ, can significantly differ from real values at depth and moreover, aluminium analyses are not reliable in such saline fluids. Therefore, it is necessary to present realistic assumptions such as the thermodynamic equilibrium of the fluid with the mineral assemblage observed in the veins.

The second problem, already mentioned before, comes from the high salinity of the fluid (TDS >100 g/kg). Most of the models used to calculate geochemical equilibrium in solution consider the concept and the formula of Debye-Huckel to calculate activity coefficients of the aqueous species. The validity domain of Debye-Huckel formula extends to an ionic strength of 0.8, whereas the ionic strength of the Soultz brine goes beyond 1.6. Consequently for this type of fluid, a calculation method based on the Pitzer equations is more correct (Harvie et al., 1984).

A parallel modelling of the speciation in solution for the Soultz brine was carried out at different temperatures with the codes PHREEQC for the Debye-Huckel concept (Parkhurst, 1995) and TEQUIL for the concept of Pitzer (Duan et al., 1996). The results show that the difference of the saturation indexes foreseen by these models can be really significant as displayed in Figure 1 (Durst and Vuataz, 2000).

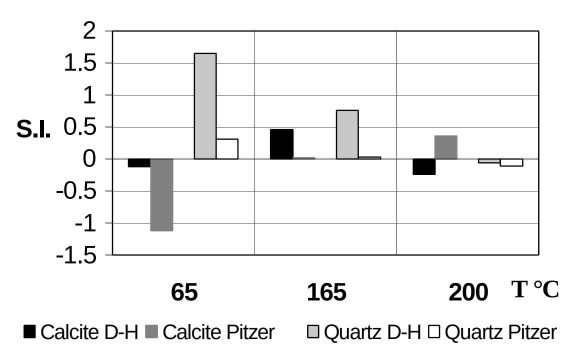

Figure 1. Comparison between saturation indexes (S.I.) calculated with Debye-Huckel (D-H) and Pitzer formalism for three different temperatures.

Unfortunately, the use of Pitzer equations brings new problems. Firstly, they increase the complexity of the code and decrease the performances of the model. Secondly, the equations parameters are unknown for aluminium, and for some species, they are not available at high temperatures (165°C and above). For the simplification of the model, some specifications of the Soultz system were considered :

- The fluid used is the natural formation fluid.
- No boiling, degassing or condensation takes place in the system.
- There is no mixing with a different type of fluid.
- The fluid-rock interactions do not modify the major species concentration (chlorine and sodium).

Given those assumptions, the only variable parameter having a significant influence on Pitzer's equations and thus on activity coefficients is temperature. This allows the use of a two-step method for the thermodynamic model. First, the activity coefficient of each species is determined at different temperatures for a mean Soultz fluid composition (table 1). This composition corresponds to the natural fluid in the reservoir conditions and is recalculated from different analyses.

Table 1. Composition of the mean Soultz fluid in mmol/kg

Na	**K**	**Ca**	**Mg**	**Fe**
1064	86.6	172	5.81	0.537
Al	**SiO_2**	**C**	**C l**	**S**
0.0148	2.33	55.8	1564	2.35
Temperature = 165°C pH ≈ 4.7				

This operation is carried out using softwares based on Pitzer's model such as Tequil or EQ3/6 (Wolery, 1992), as well as the assumption that the fluid is in equilibrium with the altered granite at 165°C. The total dissolved carbon was calculated by equilibrium with calcite at the same temperature. The sulphate content given by the analyses leads to a high anhydrite oversaturation at 165°C. Knowing that the Soultz altered granite contains almost no anhydrite but pyrite, the difference between the analysed sulphate and the equilibrium concentration with anhydrite is supposed to represent the sulphide content. The activity coefficients of the species not included in TEQUIL, such as Mg, Fe and sulphide, were calculated at 25°C using EQ3/6, and at 165°C by equilibrium with dolomite and pyrite. These coefficients were then interpolated for temperatures between 65°C and 200°C. However the aluminium behaviour still remains uncertain as the only accurate assumption is its equilibrium with K-feldspar at 165°C, but its molality and activity were extrapolated. At this point, the activity of each aqueous species can be calculated as a simple function of temperature and molality.

$$a_i = \gamma_i(T)*m_i$$

$$\gamma_i(T) = B_0+B_1T+B_2T^2+B_3T^3+B_4T^4$$

where a_i is the chemical activity of species i, γ_i its activity coefficient, m_i its molality, T the fluid temperature and B_i the coefficients determined in this study.

During the deepening to 5km of the production well GPK2 in Soultz, mineral scales were collected on the internal wall of the deep casing. On most of the casing there was only a thin layer which can possibly have been deposited after the circulation test, but at the diameter change of the casing, the amount of mineral deposit was more important. These deposits were analysed and are registered in an unpublished report of the Laboratoire de Géochimie et Métallogénie de l'Université P. et M. Curie in Paris. They are composed of quartz, calcite, aragonite, siderite, as well as iron oxides and iron hydroxides. The precipitation of the carbonates can be due to local degassing of CO_2, even if they are undersaturated in the fluid at temperatures below 165°C. The precipitation of other minerals can be explained by a drop of critical oversaturation indexes. Moreover, limited deposits of galena (lead sulphide) were observed during the 4-month flow test on the plate of the surface heat exchanger.

The thermodynamic model can be used to estimate the maximum precipitation or dissolution that might potentially occur when the 165°C fluid is cooled down to 65°C and reinjected in the reservoir. As a result, each kg of cooled fluid can potentially dissolve 323 mg of calcite and 397 mg of dolomite and can precipitate 14 mg of quartz, 52 mg of pyrite and 0.354 mg of galena. The extrapolation of these values for one year of production in the conditions of the 1997 circulation test (25 kg/s) gives a dissolution of 250'000 kg (90 m^3) of calcite and 290'000 kg (110 m^3) of dolomite as well as precipitation of 100'000 kg (40 m^3) of quartz, 40'000 kg (7 m^3) of pyrite and 280 kg (0.04 m^3) of galena near the bottom of the injection well. On the basis of this thermodynamic model, it appears that dissolution of carbonates (200 m^3) is significantly more important than the precipitation process (47 m^3), presumably leading to an increase of injectivity.

KINETIC MODELLING

The calculation of the real amount of reaction during the exploitation of the reservoir requires a model coupling chemical processes to hydraulic and transport. In this regard, precipitation and dissolution cannot be considered any more as instantaneous and kinetic factors have to be taken in account.

The work of Jacquot (2000) shows that the carbonates seem to be the most reactive minerals in the Soultz HDR system, but it does not consider sulphur minerals. The primary goal of the kinetic model will be to simulate precipitation and dissolution of quartz, calcite, dolomite and pyrite. Nevertheless other minerals like feldspar, anhydrite and hematite should later be incorporated in the model, at least to confirm that they do not play an important role. The kinetic modelling of clays has been abandoned due to the lack of available data on the subject.

For brines, no handy theoretical model is available at present time, therefore the method chosen for this study is to use one law for each reaction. These laws were taken from published experiments, conducted if possible in NaCl brines and they use the available parameters.

$v = F(T, a_i, S)$

where v is the reaction rate in the volume of rock considered in $mol.s^{-1}.m^3$, T the temperature in °C, a_i the activities of the aqueous species and S the area of reaction surface in m^2. The precipitation of a mineral is assumed to occur exclusively on pre-existent surface of this mineral. The selected equations for quartz come from Rimstidt and Barnes (1980) and Dove (1994), those for calcite and dolomite from Morse (1983) and Shiraki and Brantley (1995). Equations for pyrite are from Schoonen and Barnes (1991 a, b, c) and Williamson and Rimstidt (1994).

The computer implementation of this geochemical model has been done into the code Chem-TOUGH2, a nonisothermal reactive transport model based on TOUGH2 developed by White (1995). The geochemical module should later be coupled with FRACTure (Kohl and Hopkirk, 1995), which adds thermo-elasticity and turbulent flows modelling. This part of the project will be carried out by D. Baechler (Inst. of Geophysics, ETH-Zurich).

APPLICATION TO A 1-D FRACTURE

The hydraulic part of the simulation of the Soultz system is currently in development at the Institute of Geophysics, ETH-Zurich. Nevertheless the model has been used to simulate the chemical reaction along a 1-D fracture with water injected at 65°C and progressively heated to 165°C, similarly to the 1997 circulation test.

The circulation in the Soultz reservoir takes place in clusters of fractures filled with quartz, calcite, hematite, illite or chlorite, surrounded by porous altered granite. A fine simulation will require a complex geometry at such a small range that it will not allow the modelling of the circulation within the entire system. For this study the circulation zone is reduced to an equivalent porous media.

The model is constituted of twenty volumes as shown in figure 2. The grid has a higher density near the injection point, where the system is more reactive. A total length of 1200 m is chosen to allow a temperature increase up to 165°C along this 1-D geometry. The rock mass is formed by 40% of quartz, 4% of calcite, 1% of dolomite and 1% of pyrite. The remaining 54 % of the mass contains minerals not included in this simulation, principally clays. The values of main parameters for the simulation are listed in table 2.

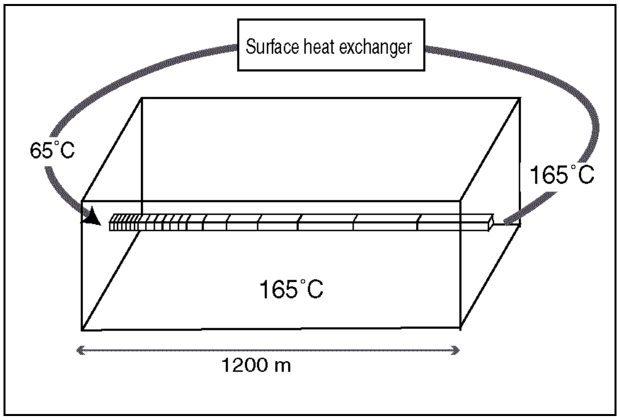

Figure 2. Simulation geometry and temperatures

Table 2.Values of the physical parameters considered for the simulation.

Porosity	10 %
Permeability	10^{-10} m^2
Rock density	2650 kg/m^3
Rock matrix temperature	165°C
Reservoir pressure	40 MPa
Conduit injection rate	10 kg/s
Injection temperature	65°C

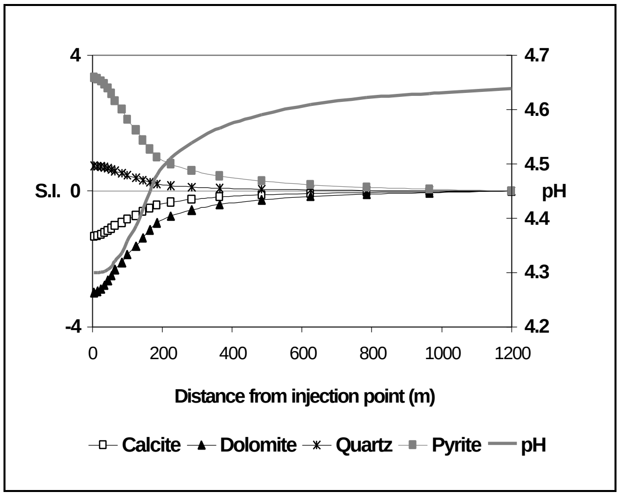

Figure 3. Initial values of the saturation indexes (S.I.) and of pH in the system.

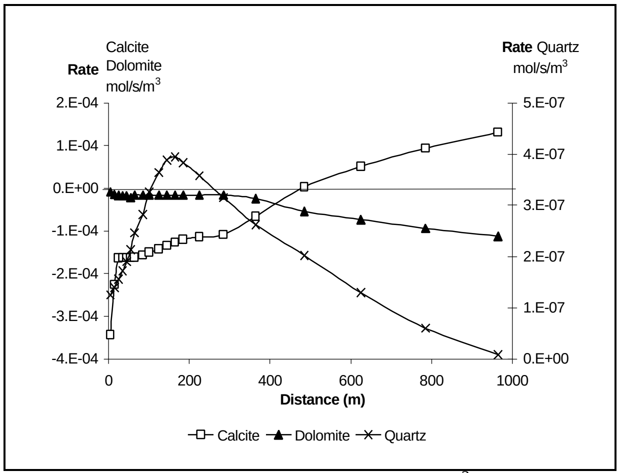

Figure 4. Initial reaction rates (mol/s/m^3) calculated after one day of circulation.

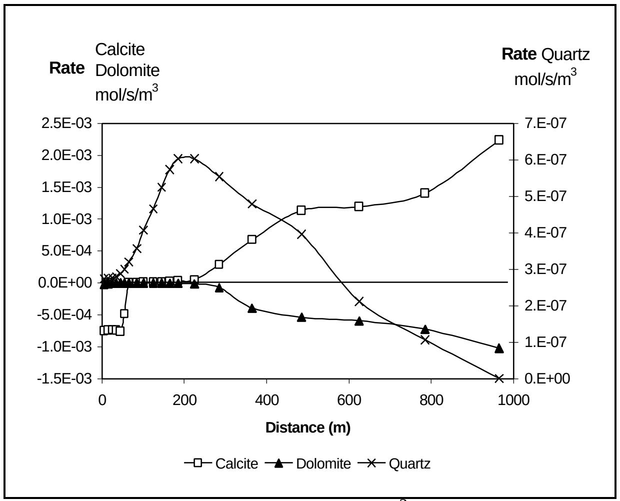

Figure 5. Reaction rates (mol/s/m^3) calculated after one month of circulation

The simulation starts at thermal equilibrium. The pH of the fluid and its saturation indexes with regard to calcite, dolomite, quartz and pyrite are shown in figure 3. The desequilibrium observed near the injection point induces the reaction of the system, namely dissolution of carbonates and precipitation of quartz and pyrite. A first kinetic simulation presents the reaction rates at the end of the first day (figure 4). Considering the theoretical thermal equilibrium, calcite dissolves in the first 500 m and then precipitates in the rest of the flow path. Dolomite dissolves all the way but its dissolution rate increases simultaneously with calcite precipitation. Quartz precipitates in the whole flow path with a maximum at 200 m from the injection point.

After one month of circulation, as shown in figure 5, calcite dissolves only in the fist 100 m of the path and starts to precipitate at 200 m. Dolomite still dissolves in parallel of calcite precipitation. At the vicinity of the injection point, for one month, it represents 7 % of relative porosity increase. Farther in the circuit, the volumes of precipitated calcite and dissolved dolomite equilibrate themselves. The quartz precipitation does not vary significantly during the simulation period of one month. At its point of maximum precipitation, the quartz can induce 0.1% of porosity decrease per month.

During the next 12 months, the system evolution remains similar with a slow diminution of carbonates reaction rates. The quartz precipitation rate does not vary significantly in the time scale considered (1 year). Simulation carried out only with quartz seems to indicate that quartz behaviour remains more or less constant, even for a 30-year period.

DISCUSSION

Thermodynamic modelling of the Soultz brine showed that the use of the Pitzer concept is limited, due the scarcity of the data for high temperature and the absence of certain important minerals. However, the lack of data can be overwhelmed by the use of hypotheses on the state of fluid equilibrium in the reservoir. Nevertheless, the consequence was the abandonment of aluminosilicates modelling and redox processes. Another limitation of the Pitzer equations is due to the long time of calculation, in case it is coupled to a thermo-hydraulic model. The method used for thermodynamic modelling, based on the specifications chosen to represent the system (see above), allows bypassing the slowness of the calculation of the Pitzer model.

The maximum values of precipitation/dissolution processes computed by the thermodynamic model are revised to lower values with the kinetic modelling. Results of the simulation of reaction rates for calcite and dolomite within a 1-D conduit do not differ significantly for a 1-month or a 12-month circulation. Moreover, a simulation for quartz alone gives similar reaction rates for a period up to 30 years. It appears that the stability of the reaction rates and the setting up of an almost permanent state depends on the thermal equilibrium of the system.

The simulation carried out indicates that dissolution of calcite in the vicinity of the injection point leads to a 30% augmentation of the porosity in the first 100 m during the first year. This percent-increase is obtained for a porous-equivalent medium and is probably higher than the real value. Farther, the effects of calcite re-precipitation on the porosity are compensated by the dissolution of equivalent volumes of dolomite. However, the precipitation of quartz does not seem to significantly influence the porosity.

The ongoing research consists first to integrate a process to control the redox potential, which will allow a realistic simulation of pyrite and hematite, and second to model the effects of the geochemical reactions on the permeability.

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