

Simulation of Mineral Precipitation and Dissolution in the 5-km Deep Enhanced Geothermal Reservoir at Soultz-sous-Forêts, France

Vero Rabemanana¹, François-D. Vuataz¹, Thomas Kohl² and Laurent André¹

¹ Centre of Hydrogeology, University of Neuchâtel, Emile Argand, 11 – CH-2007 Neuchâtel, Switzerland

² GEOWATT AG, Dohlenweg 28, CH-8050 Zürich, Switzerland

E-mails: Vero.Rabemanana@unine.ch, Francois.Vuataz@unine.ch, Kohl@geowatt.ch, Laurent.Andre@unine.ch

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ABSTRACT

The long-term behaviour of the Soultz-sous-Forêts enhanced geothermal reservoir has been investigated. Major processes susceptible to modify the properties of the reservoir were taken into account. Heat, fluid transport and geochemical reactions are coupled for modelling the reactive flow. As the fluid is a hot brine, a new code was built to simulate the Thermo-Hydraulic and Chemical effects (THC) within the fractures connecting the injection and production wells. The results revealed that re-injection of the formation brine after cooling changes the chemical equilibrium states between the formation fluid and different minerals present in the host rock. Due to their fast reaction rates, carbonate minerals are responsible for most of the reservoir evolution. Amounts of quartz and pyrite deposited are negligible during the period of simulation. In order to improve the hydraulic performance of the reservoir, the effects of acid addition and temporary reverse circulation of fluid were also investigated.

1. INTRODUCTION

In order to forecast the long term behaviour of an enhanced geothermal reservoir under exploitation, interaction between flow, heat transfer, transport and chemical reactions must be considered. This study aims to the coupled modelling of these processes applied to the Soultz-sous-Forêts enhanced geothermal system (EGS), located in Alsace, about 50 km north of Strasbourg (France). The Soultz area was selected as the European EGS pilot site because of its strong temperature gradient in the sedimentary cover (up to 100°C km⁻¹) and its high heat flow reaching locally 0.15 W m⁻² (Kohl and Rybach, 2001).

The project has started in 1987 with the final goal to produce electricity by conversion of heat extracted from a fractured reservoir. The geology of the Soultz region is characterized by a graben structure affected by several N-S striking faults. The crystalline basement is composed of granitic rocks covered by 1400 m of Triassic and Tertiary sediments. Three facies of the granitic host rock are identified: unaltered granite in which fracturation density is close to zero, hydrothermally altered granite facies and veins of alteration (Jacquot, 2000). Hydrothermally altered granite facies is the most porous one (Genter et al., 1997) and veins of alteration are highly fractured. Circulation of fluid takes place mainly within these two last facies, but only the hydrothermalized facies plays the major role in the fluid-rock interaction processes.

The planned arrangement of boreholes is 3-well system with one well for cooled water re-injection, and two wells, on both sides of injector, are used for hot water production.

The bottom of each well is separated by around 650 m from the other ones. Cooled water will be reinjected in the system at a rate of about 100 l s⁻¹. Currently, the three deep boreholes GPK2, GPK3 and GPK4 were drilled at a depth of 5000 m. A temperature of 200°C was measured at the bottom hole. GPK3 should be the injection well, while GPK2 and GPK4 will serve as the production wells. At Soultz, the injection – production system is a closed loop. The fluid used is the formation fluid existing in the granite, namely a brine with a total dissolved solids of 100 g kg⁻¹. The re-injection of the cooled brine disturbs the equilibrium between fluid and reactive minerals. Temperature and pressure in the reservoir will also be changed. In a previous study, a first modelling of deep reservoir at Soultz was presented (Rabemanana et al., 2003).

In the present paper, influence of water-rock interaction on the fluid flow and the reservoir properties is investigated for a longer simulation period.

2. NUMERICAL MODELLING

To predict the long term behaviour of an enhanced geothermal reservoir, it is necessary to understand the chemical phenomena occurring during exploitation. However, the chemical behaviour of a hot brine system is very complex, depending on the fluid composition, the temperature and the pressure. For the case of the formation fluid at Soultz, application of Debye-Hückel theory is not accurate, and existing geochemical codes are not adapted to its temperature. A new numerical simulation model called FRACHEM (Durst, 2002; Bächler, 2003) based on the Pitzer formalism to calculate aqueous speciation has been developed. It is a combination of two existent codes FRACTure (Kohl et al., 1995) and CHEM-TOUGH2 (White, 1995). FRACTure calculates the thermal and hydraulic processes whereas CHEM-TOUGH2 simulates the reactive transport. Some modifications have been implemented to the geochemical module of CHEM-TOUGH2. These modifications consist of the calculation of the activity of the aqueous species, the kinetic laws, the permeability and porosity changes as well as the re-injection process.

2.1 Thermodynamic model

The original CHEM-TOUGH2 code uses Debye-Hückel model for the calculation of the activity coefficient. Taking into account the high salinity of Soultz fluid, Pitzer formalism was implemented in the geochemical module of CHEM-TOUGH2. According to some assumptions about the Soultz fluid, such as no boiling, no degassing, no condensation, no mixing with a different fluid and conservation of major species such as chloride, the activity a_i of each aqueous species is assumed to be only function of the temperature T (°C) and the molality m_i (mol kg⁻¹) (Durst, 2002).

$$a_i = \gamma_i(T) * m_i \quad (1)$$

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

where B_i are the coefficients of the activity coefficient of the specie i (γ_i).

2.2 Kinetic model

For the kinetic model, a simplified model based on one kinetic law accounting, the temperature, the reaction surface and the distance from equilibrium is proposed to describe each dissolution and precipitation reaction for each mineral.

$$v = k_m(T) \cdot s_m \cdot \left(1 - \left(\frac{Q_m}{K_m} \right)^\mu \right)^n \quad (3)$$

where m is the mineral index, s_m (m^2) is the surface area, k_m ($\text{mol s}^{-1} m^2$) is the rate constant, K_m is the equilibrium constant, Q_m is the ionic activity product and the exponents μ and n are positive empirical factors. Positive values of the reaction rate v (mol s^{-1}) indicate dissolution and negative values precipitation.

The kinetic laws of the Soultz system are derived from published experiments conducted in NaCl brines. Detailed information on the determination of the reaction laws can be found in Durst (2002).

2.3 Permeability and porosity relationships

As the circulation at Soultz mainly occurs in clusters of fractures and in highly altered granite, two porosity and permeability relationships were considered. The first one is a simple cubic Kozeny - Carman grain model based on spheres that relates the permeability k (m^2) to the porosity ϕ by:

$$k = \frac{R_0^2}{45} * \left[\frac{\phi^3}{(1-\phi)^2} \right] \quad (4)$$

where R_0 (m) is the initial spherical close-pack radius.

The second one is the porosity-permeability model for fractured rock after Norton and Knapp (1977). In this model, the permeability for a single set of parallel fracture is given by:

$$k = \frac{\phi * \delta^2}{12} \quad (5)$$

where δ (m) is the fracture aperture and ϕ is the fracture porosity.

This last relation assumes that reactions occur exclusively along the fracture walls and do not within the matrix around the fractures.

2.4 Re-injection process

The last modification implemented in CHEM-TOUGH2 consists of the modelling of the re-injection process. The original version of CHEM-TOUGH2 did not adjust automatically the concentration changes of the injected fluid. This was manually made after each time step. In the modified version, the re-injection of the cooled produced fluid is now possible. The concentration of each species in the volume element where the re-injection takes place is

determined in function of the mass of the fluid injected and of the concentration in the production volume element.

3. APPLICATION OF FRACHEM

3.1 Model set up

The present application of FRACHEM is the modelling of a 2D simplified model with a geometry close to the real Soultz system. The injection and the production wells are linked by fractured zones and surrounded by the granite matrix. The model is composed of 125 fractured zones. Each fractured zone has an aperture of 0.1 m, a depth of 200 m, a porosity of 10%, and contains 100 fractures. Initially the temperature was set to the reservoir temperature of 200°C. One of these fractured zones is modelled with the assumption that the fluid exchange with the surrounding low permeability matrix is insignificant. Due to the symmetrical shape of the model, only the upper part of the fractured zone is considered in the simulation. The area is discretized into 222 2D elements (Figure 1).

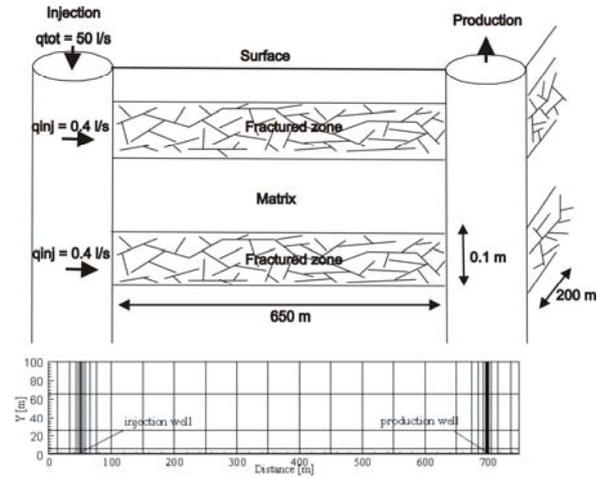


Figure 1: Simplified model and spatial discretization

The size of the elements ranges from a minimum of 0.5 m x 0.5 m near the injection and the production wells to a maximum of 50 m x 35 m. Considering a production rate of 50 l s⁻¹, the fluid was re-injected in each of the fractured zones at a rate of 4 x 10⁻¹ l s⁻¹ and at a constant temperature of 65°C. In this simulation a constant overpressure of 8 MPa was assumed at the injection well and a hydrostatic pressure at the production well. Dirchlet boundary conditions were applied to the upper, left and right side of the model. Due to the sensitivity of the sequential non iterative approach (SNIA) method on the time discretization, the time step used for this simulation is limited to 10² s, meaning that long-term simulations take several days of computer time. The values of thermo-hydraulic parameters considered in the simulation are listed for the Table 1.

Table 1: Thermo-hydraulic model parameters

| Parameters | | Fracture | Matrix | Fluid |
|------------------------|-------------------------|----------------------|-------------------|-------|
| Hydraulic conductivity | K [m ² /Pa] | 7.4 10 ⁻⁸ | 10 ⁻¹⁵ | - |
| Thermal conductivity | λ [W/m.K] | 2.9 | 3 | 0.6 |
| Density | ρ [kg/m ³] | - | 2650 | 1000 |
| Heat capacity | C _p [J/kg.K] | - | 1000 | 4200 |
| Porosity | Φ [%] | 10 | 0 | - |

Table 2 contains the mean composition of the fluid which is derived from the chemistry of the fluid produced during the production test in 1999. The fluid has a pH of 4.9 and a temperature of 200°C. This composition was recalculated assuming an equilibrium between the fluid and the mineral assemblage of the fracture. Initial mineral abundances are based on the synthetically identification of host rock facies (Jacquot, 2000).

Table 2: Mean fluid composition of the 5-km reservoir

| Species | Concentration [mmol kg ⁻¹] |
|------------------|--|
| Na ⁺ | 1079 |
| K ⁺ | 68.5 |
| Ca ²⁺ | 157 |
| Mg ²⁺ | 3 |
| Cl ⁻ | 1452 |
| S | 1.6 |
| C | 19 |
| Fe ²⁺ | 2.4 |
| SiO ₂ | 5.7 |

3.2 Results

3.2.1 Temperature distribution

At first, the reservoir fluid is considered to be in thermal equilibrium at 200°C with the host granite. Re-injection of fluid at 65°C will disturb this equilibrium and will cause mineral dissolution and precipitation. The temperature distribution after 5 years of exploitation is illustrated. For clarity, Figure 2 shows only the half model extension in Y direction. Around the injection well, a thermal front is progressively developed. The temperature decreases due to the thermal diffusion. The front has reached the production well after a simulation period of 5 years and a temperature decrease of 15°C is observed at the production well.

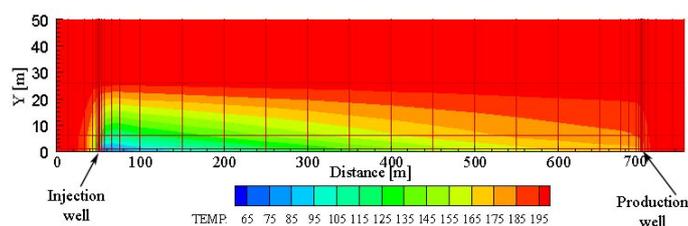


Figure 2: Temperature distribution after 5 years of circulation

3.2.2 Simulation of fluid-rock interaction

Evolution of the fluid-rock interaction is investigated by studying the reaction rate of minerals. For this simulation, only calcite, quartz, dolomite and pyrite have been considered. Other minerals are assumed to be non-reactive because of the lack of their Pitzer coefficients. Figure 3 illustrates the calcite reaction rate. During the first year of simulation, calcite dissolution occurs exclusively in a 300 m wide zone near the injection well. This dissolution process leads to an enrichment of the Ca²⁺ content in the fluid. With the increase of the temperature, calcite starts to precipitate towards the production well. A maximum precipitation rate of $1.59 \times 10^{-4} \text{ mol s}^{-1} \text{ m}^{-3}$ is reached after 3 months of circulation. With increasing the simulation time, the dissolution zone extends towards the production well and the dissolution reaction rate decreases near the equilibrium state.

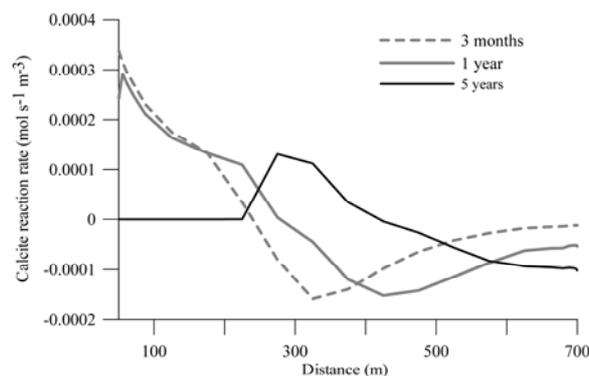


Figure 3: Evolution of calcite reaction rate

Compared to calcite, the behaviour of dolomite is slightly different and the reaction rate is much lower. Over the simulation period, no precipitation of dolomite is observed (Figure 4). Dolomite dissolves all along the fracture zone. After one year of circulation, the dissolution rate reaches $1.60 \times 10^{-5} \text{ mol s}^{-1} \text{ m}^{-3}$ and then decreases during all the simulation.

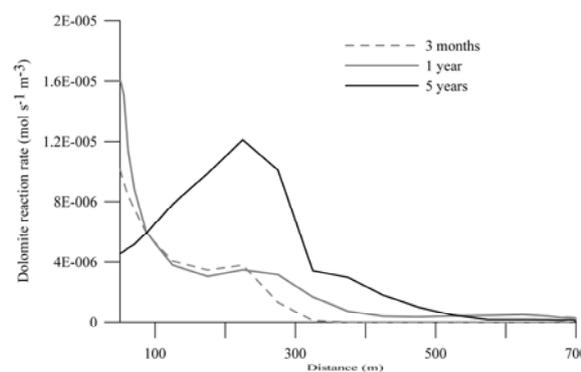


Figure 4: Evolution of dolomite reaction rate

Figures 5 and 6 show that during all simulation period, quartz and pyrite do not dissolve along the fracture zone. Compared to those of carbonates, their reaction rates are much lower. The maximum quartz precipitation rate is observed towards the production well but with increasing of simulation time, this tends towards zero, which is the rate at thermodynamic equilibrium.

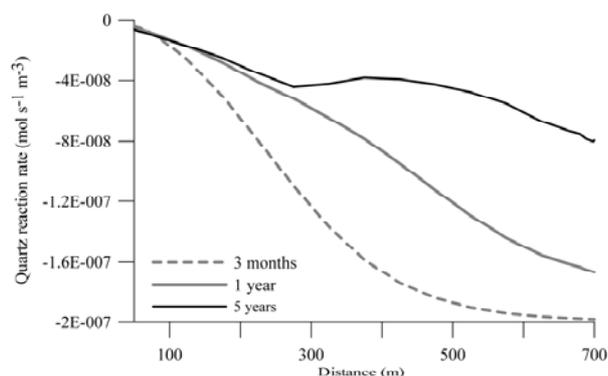


Figure 5: Evolution of quartz reaction rate

Among the minerals taken into account for this simulation, pyrite has the lowest magnitude rate, which can be considered as negligible. A maximum precipitation rate of $3.05 \times 10^{-17} \text{ mol s}^{-1} \text{ m}^{-3}$ is observed and remains constant during all the simulation period. However, with increasing

time, the zone maximum of precipitation moves towards the production well.

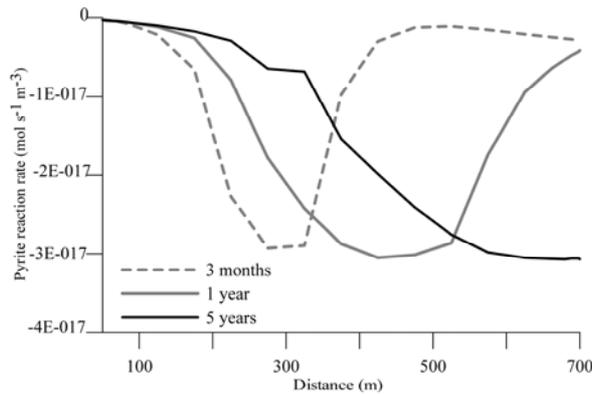


Figure 6: Evolution of pyrite reaction rate

3.2.3 Forecast of secondary mineral transfer

Knowing the reaction rate of each mineral, the amount of dissolved and precipitated minerals per element at the time *t* was calculated from the following expression:

$$m_{i,E} = mw_i * v_E * \phi_E * \sum_{j=t_0}^t (r_{i,j} * \Delta t) \quad (6)$$

m_{i,E} (kg): the amount of mineral *i* within element *E*
mw (kg mol⁻¹): the molecular weight of the mineral
v_E (m⁻³): the volume of element, *φ_E* is the porosity
r_{i,j} (mol s⁻¹ m⁻³): the reaction rate of the mineral *i*
 Δt (s): the time interval.

Table 3 gives the amounts of precipitated and dissolved mineral after 5 years of exploitation. As the calcite reaction is very fast, a significant amount of calcite dissolved and precipitated all along the fractured zone. Due to the retrograde solubility of calcite, amounts of dissolved calcite are most pronounced around the injection well. Quartz and pyrite precipitate in very small quantities.

Table 3: Amounts of precipitated and dissolved minerals within the simulated fractured zone after 5 years of circulation

| Mineral | Dissolved amount (kg) | Precipitated amount (kg) |
|----------|-----------------------|--------------------------|
| Calcite | 2'740 | 1'330 |
| Dolomite | 510 | 0 |
| Quartz | 0 | 1.74 |
| Pyrite | 0 | 0.74 |

3.2.4 Changes of permeability and porosity

One of key questions in reservoir exploitation is how do the reservoir properties evolve with time? In this section, the evolution of the porosity and the permeability is coupled to the reactions taking place along the fractured zone. According to the amount of precipitated and dissolved calcite, porosity and permeability of the reservoir are changing (Figure 7). Occurrence of calcite precipitation results in a decrease of fracture void volume by 40%. The corresponding permeability decrease towards the production well. In contrast, due to the mineral dissolution,

an increase of porosity and permeability are observed along the first 350 m from the injection well.

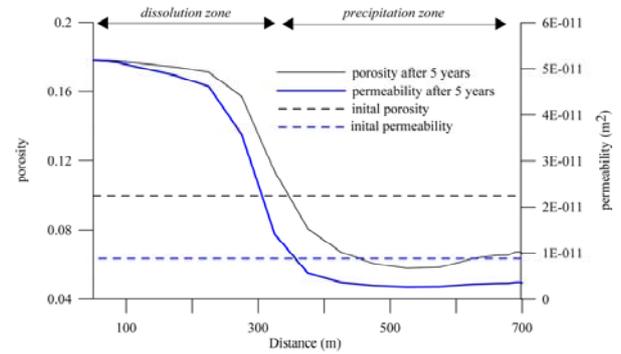


Figure 7: Evolution of porosity and permeability after 5 years of exploitation

3.2.5 Simulation of acid injection

The porosity and permeability are expected to evolve with time during exploitation. A decrease of these parameters values is the result of calcite precipitation and may alter the performance of the fluid circulation after a long-term exploitation. In this simulation, the effect of acid (HCl) injection is examined in order to reduce calcite deposit. To this end, the pH of the brine is changed to 3.7. Figure 8 compares the amount of dissolved calcite near the injection well and all along the fractured zone obtained with the original brine and acidified brine. The result shows that the additional H⁺ ions significantly increased the amount of dissolved calcite. As the permeability and porosity of reservoir are controlled by the occurrence of mineral precipitation and dissolution, this increase of dissolved calcite mirrors an improvement of the reservoir properties.

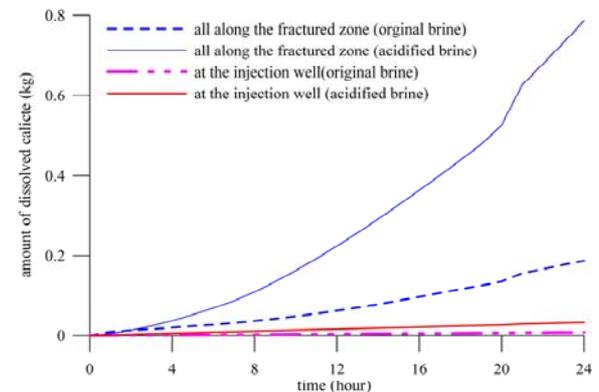


Figure 8: Evolution of amount of dissolved calcite after acid addition

3.2.6 Effect of a temporary reverse circulation

Another scenario has been tested with a temporary reverse fluid circulation after a given period of exploitation. The simulation of fluid-rock interaction showed that calcite is more soluble around the injection well and that an increase of temperature favours calcite deposits. In order to improve porosity and permeability around the production well, the fluid circulation has been reversed during one month after one year of exploitation. This means the production well is used for injection and vice versa during this period. The fluid chemistry and mineral components are updated and the boundary conditions are inverted to this configuration. Figure 9 shows a comparison of the porosity simulation with and without reverse circulation after 2 years of exploitation.

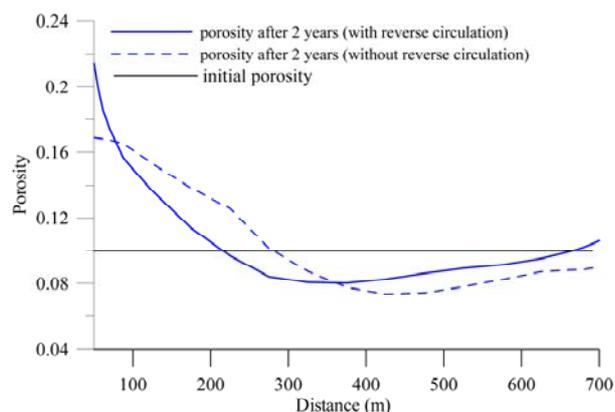


Figure 9: Evolution of the porosity by temporary reversing the fluid circulation

The porosity evolution is different for these two scenarios of exploitation. Calcite reaction rate is significantly important for the forward circulation as confirmed by the observed greater amount of calcite deposited and dissolved (Table 4). However, by including a reverse circulation of the fluid during a period of one month, the porosity changes are significantly improved around the wells. The porosity is slightly increased of 6% at the production well and the decrease of the porosity caused by calcite precipitated is less pronounced, compared to that obtained without reverse circulation.

Table 4: Comparison of amount of calcite deposited and dissolved after 2 years of circulation

| Weight of calcite (kg) | Forward circulation | Reverse circulation (1 month) after 1 year of forward circulation |
|------------------------|---------------------|---|
| precipitated | 917 | 825 |
| dissolved | 1'412 | 975 |

4. CONCLUSION

A Thermo-hydraulic and chemical (THC) coupled model has been developed and applied for the Soultz-sous-Forêts enhanced geothermal system. This study is focused on the prediction of the long-term behaviour of reservoir properties. It has been shown that re-injection of the cooled formation brine changes the chemical equilibrium states between the formation fluid and different minerals in the host rock. At the beginning of the simulation, carbonates are quickly dissolved near the injection well. With increasing simulation time, the carbonates are progressively consumed and the reaction rates decrease significantly. Small amounts of quartz and pyrite precipitate during the simulation period. Compared to those of carbonates, their reaction rates are much lower. It can be concluded that carbonates reaction predominate, especially the calcite reaction. As a consequence of calcite precipitation,

permeability is reduced from its initial value of $9 \times 10^{-12} \text{ m}^2$ to $2 \times 10^{-12} \text{ m}^2$ at the production well. This alteration of the hydraulic performance of the reservoir could be improved by carrying out a temporary reverse circulation of the fluid between the injection and production wells or by injecting acid to dissolve calcite deposited around the production well.

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