REACTION FRONT FINGERING IN ANHYDRITE CEMENTED SANDSTONE

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SUMMARY - A geothermal bore at Allermöhe (Germany) tapped unintentionally and unexpectedly a reservoir almost totally cemented with anhydrite. Laboratory core flooding experiments were conducted to determine the potential to stimulate the geothermal aquifer. During these experiments sudden permeability increases, due to dissolution of anhydrite, were observed. Preferential flow paths were detected in the cores by X-ray tomography and are here simulated numerically. The numerical code **SHEMAT** a simulation system for coupled flow, heat transfer, species transport, and chemical reactions is used. Data of the core flooding experiments and the numerical studies coincide. A feasibility study to stimulate the geothermal aquifer at Allermöhe shows that the development of preferential flow paths within the vicinity of the bore can be expected. Hence, gentle stimulation of the bore seems possible.

1. INTRODUCTION

The bore Allermöhe 1 (Hamburg, Germany) was deepened to a depth of 3,300 m in 1997. It taps a 70 m thick sandstone aquifer with a temperature of 125°C. Although temperature and thickness of the aquifer agree with the conditions needed for geothermal energy use, the pore spaces, originally open with porosities up to 20 %, are filled to a large extent by anhydrite. Mineralogical investigations showed anhydrite cementation with completely filled pore spaces as well as insular, cloudy, or layered structures. The extractable amount of water, 3 m^3/h, determined by a pumping test, is too low for an economical use of the resource (Baermann et al., 2000a).

To assess the feasibility of a gentle stimulation (no use of chemicals) which would connect the cemented areas in the direct vicinity of the bore to areas of the aquifer with higher permeabilities, laboratory core flooding experiments were conducted using anhydrite cemented sandstone samples fiom the geothermal bore Allermöhe. The experiments showed a sudden permeability increase after an unexpected short time. X-ray tomography images revealed channel like flow paths within the cores (Baermann et al., 2000b).

Prerequisites for preferential flow path formation are described in the literature. Ormond and Ortoleva (2000) showed that interaction between mineral reaction and mass transport in rocks can lead to reaction front instability and the development of channel-like voids.

The numerical code SHEMAT, a subsurface flow and hydrogeothermal simulation system for coupled flow, heat transfer, species transport, and chemical reactions (Bartels et al., 2000) is used to simulate the core flooding experiments of Baermann et al. (2000b). The numerical model of the flooded cores is set up with the help of the graphical user interface Processing SHEMAT.

2. PREFERENTIAL FLOW PATHS

2.1 Model Conception

Flow coupled with transport of solutes generated by mineral dissolution can lead to self-organized enhancement of the heterogeneity in the rock.

System self-organization is not a violation of the second law of thermodynamics, which states that in any spontaneous change the universe evolves toward a state of decreased order. This implies that the disorder of a system plus that of the environment increases with any natural process. An increase in order within a system can be overcompensated by a larger increase in disorder in its environment (Ortoleva, 1994).

If the initial rock texture is perfectly uniform, the resulting reaction front will remain planar while advancing down stream with time. However, if the rock texture is initially even slightly non-uniform, the resulting enhancement of the permeability will be uneven and a fingering reaction front may form (Wei and Ortoleva, 1990).

Fluid flows preferentially in regions with higher permeability. If the fluid is undersaturated with regard to a specific mineral phase, mineral dissolution occurs. Locally restricted, dissolution is faster in the regions of higher permeability. The

increase in porosity leads to an increase in permeability. The permeability increase in turn causes higher flow of undersaturated solution. Due to that positive feedback loop, preferential flow paths arise.

Concentration compensation caused by dispersion or diffusion counteracts the formation, because the undersaturation, which is the driving force of the process, decreases (Ormond and Ortoleva, 2000).

2.2 Formation Prerequisites

Whether preferential flow paths develop, can be determined by evaluating the dimensionless Peclet Pe and Damkohler Da number.

$$Pe = \frac{v \cdot l}{D} \qquad (1)$$

In equation (1) v (L/T = Length / Time) is the mean flow velocity, l (L) the characteristic length (in this case the thickness of the system), and D (L^2/T) the dispersion coefficient.

$$Da = \frac{k \cdot l}{v} \qquad (2)$$

Additionally in equation (2) k (1/T) is the kinetic coefficient of the mineral reaction.

The preferential flow path length is determined by the relation between dispersion and advection. If dispersion evens out the concentration within the flow path, the growth of the channel stops. The Peclet number (equation (1)) expresses the relation between advection and dispersion. With infinite Peclet numbers preferential flow paths would grow infinitely long.

The Damkohler number describes the relation between the time a chemical reaction requires to reach equilibrium and the time the fluid needs to flow through a characteristic length. Damkohler numbers smaller than 10^{-2} result in a planar reaction front. Reaction front instabilities occur if the Damkohler number is greater than 1. If the kinetics of the chemical dissolution and precipitation reactions are neglected, which means thermodynamic equilibrium is assumed, the Damkohler number becomes infinite.

2 3 Channel Development

The process of channel development in anhydrite cemented sandstone has been investigated in previous simulations following the numerical studies of Onnond and Ortoleva (2000).

Depending on the width of the initial heterogeneity a varying number of flow paths develops, whereas size and growth rate of the fingers do not depend on the width of the initial heterogeneity. If the initial heterogeneity is wider, two fingers start developing from each downstream comer of it. If the distance between the fingers is too short, one of the fingers stops growing while the remaining one grows wider and develops towards the center of the domain, along the largest pressure gradient (Onnond and Ortoleva, 2000).

The length of an elongated channel depends on the balance between advection and dispersion: If dispersion equilibrates concentration inside the channel before the fluid reaches the channel tip, growth stops (Ormond and Ortoleva, 2000).

3. ALLERMÖHE CASE STUDY

3.1 Core Flooding Experiments

Dissolution of anhydrite and resulting permeability changes were studied in 7 flooding experiments with core material from the bore Allermohe. Samples were 8 cm in length and 6.5 cm in diameter. Experiments were carried out in triaxial apparatuses under 3.8 bars cell pressure and up to 2.8 bars flow pressure at 20°C. Cores were flooded up to 84 days, some with pure water and some with 10 g/L sodium chloride solutions (Baermann et al., 2000b).

Initial permeabilities have been 0.01 to 0.3 mD (1 mD = milli Darcy ≈ $1.0 * 10^{-15}$ m^2) with corresponding porosities of 1 to 5 %. Baermann et al. (2000b) observed a gentle permeability increase until a breakthrough occurred leading to an increase up to 1000 fold the initial permeability.

3.2 Set-Up of the Allermöhe Model

For the simulation a vertical 2D model is used. Model extents are 0.065 m versus 0.08 m. The model was discretized with 82 * 80 cells measuring 0.001 m * 0.0005 m in the center parallel to the core length and 0.001 m * 0.001 m near the edges.

Core properties and boundary conditions are deduced from Baermann et al. (2000b). The experiment with the shortest duration period due to earliest breakthrough (40 days) has been chosen for numerical simulation (experiment P6).

During the 40 days of flooding of core P6 the average porosity increased from 1.9 % to 3.9 % and the permeability from 0.05 mD to 54 mD. We assume that the pore space increase occurred exclusively due to anhydrite dissolution. With the molar volume of the mineral of $4.6 * 10^{-5}$ m^3/mol, it is derived that 0.1 154 mol anhydrite (=15.7 g) have been dissolved. This coincides with the mentioned loss of 16.1 g core weight (Baermann et al., 2000b).

Previous numerical studies yield, that the developing preferential flow path comprises about 17 % of the total area respectively the volume. We assume that the whole amount of anhydrite which leaves the core has been dissolved from the preferential flow path section. The anhydrite

concentration in the rock material is therefore 2550 **mol/m^3**. Thus, the porosity within the channel subsequently increases from **1.9 %** to **13.7** %.

The relationship between permeability and porosity, implemented in the numerical code, is based on the assumption that the shape of the internal surface of rock pores follows a rule of self-similarity. Thus, the theory of fractals can be applied (Pape et al., 1999). In SHEMAT this is approximated by equation **(3)** with the permeability k, the porosity ϕ, and the fractal exponent exp. k_0, ϕ_0 denote the initial values of permeability and porosity (Bartels et al., 2000).

$$k = k_0 \, (\phi / \phi_0)^{exp} \qquad (3)$$

The fractal exponent describing the porosity and permeability relation in this specific core is **3.5.**

An initial heterogeneity is defined in the central inflow region of the core measuring $6*10^{-3}$ m versus $3*10^{-3}$ m. Hydraulic properties are set to **13.7 %** porosity and a permeability of **54** mD. All other cells have a porosity of **1.9 %** and a permeability of 0.05 mD.

The measured porosities, both before and after the core flooding experiment, are considered as average property of the entire core. In contrast thereto the initial permeability is assigned to the entire core but the resulting permeability is taken as the permeability of the preferential flow path.

Initial hydraulic heads are 68 m at the inflow and 38 m at the outflow boundary regarding the experimental data of Bearmann et al. (2000b).

The actual dispersion length, including numerical dispersion, has been $8.0*10^{-4}$ m. The Peclet number is therefore 81. The laboratory experiment showed equilibrium concentration of calcium in the outflow of the core, kinetics of the mineral reaction are therefore neglected, leading to an infinite Damköhler number. The prerequisites for preferential flow path formation are met.

3.3 Results

The growth of the preferential flow path is shown in detail in Figure 1, with the porosity development during the numerical experiment in four snap shots, after **5, 15, 30,** and **45** days.

At the beginning two preferential flow paths develop from the down stream comers of the initial heterogeneity (upper image, 5 days).

Until **15** days after the start both fingers develop in the same way (second image from the top).

After some time one finger stops growing whereas the other one grows wider and bends towards the center of the core (third image from the top, 30 days).

With the experiment continuing the still growing finger combines with the finger which ceased growing, thus both form one preferential flow path (lower image, **45** days). There are 2 cm left until the breakthrough occurs.

It can be seen that the front of porosity change is relatively steep across the entire length of the channel.

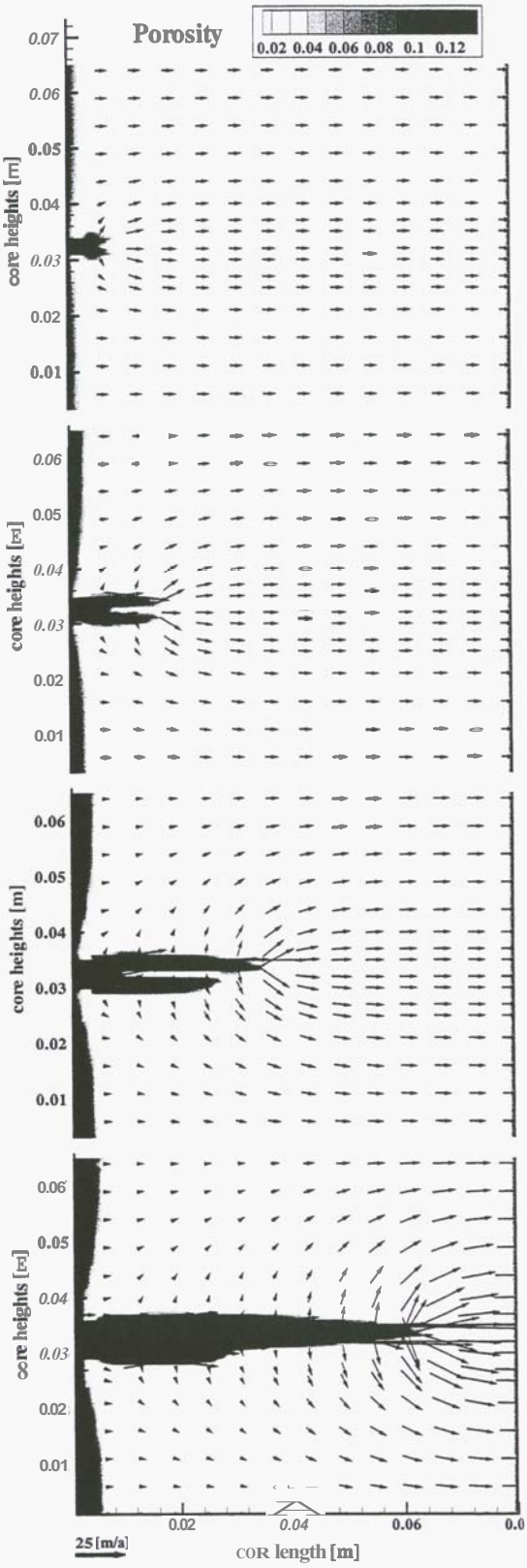

Figure 1: Development of porosity during the numerical core flooding experiment. From top to bottom, **distribution after 5, 15, 30, and 45 days. Arrows depict direction and magnitude** of **flow**

The length of the arrows in Figure 1 depicts the direction of flow. It is obvious that the highest flow velocities occur at the channel tip and that their magnitude increases with progressing simulation time.

The arrow heads trace the flow direction. They show that the area of influence of the channel on the flow field within the entire core increases with the growth of the preferential flow path. With the channel development the area of infiltration, initially just the inlet section (the left boundary of the core), increases and with it the recharged rate of the water.

In Figure2 the temporal development of the channel length, which has been observed during the numerical experiment, within the core is shown. The breakthrough occurs after 48 days, compared to 40 days within the laboratory experiment. The growth rate can be described with an exponential function. The coefficient of total determination is 0.9889.

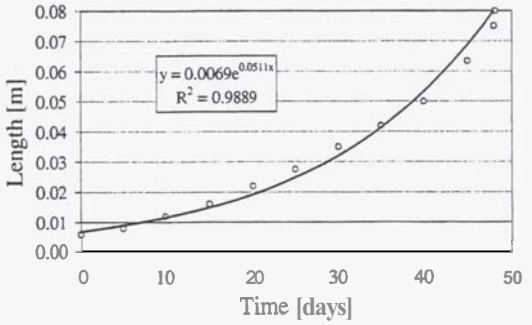

Figure 2: Velocity of growth of the preferential flow path. Breakthrough occurs after 48 days.

With the growing of the preferential flow path the rate of water flooded through the core increases. In Figure 3 the rate is shown in mL/day versus the duration of the numerical simulation. An increase of 35 mL/day to 275 mL/day has been reached after 48 days with the channel extending to the outlet of the core. The laboratory results show an increase to 300 mL/day together with the earlier breakthrough.

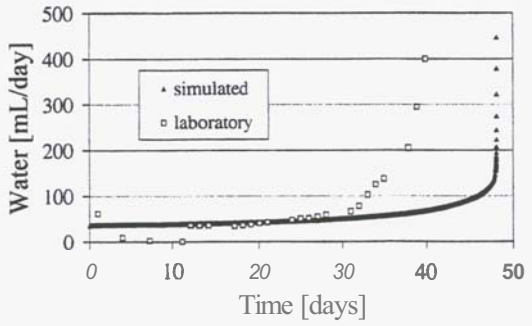

Figure 3: Rate of water in mL per day flooded through the core during the numerical and laboratory experiment.

Figure4 reveals, that the increased recharge rate is due to a continuously increasing permeability of the core. The numerical results are shown in comparison to the laboratory data.

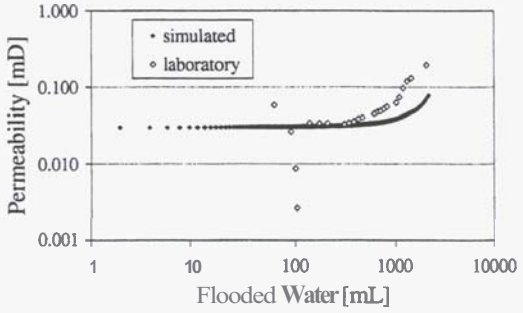

Figure 4: Comparison of the average permeability, determined within the laboratory experiment and calculated during the simulation.

The permeability increase is caused by anhydrite dissolution in the core. The total amounts determined within simulation and laboratory experiment coincide very well (Figure 5). The concentration of calcium at the core outlet during the simulation is constantly 21 mmol/L (840 mg/L). The laboratory experiment shows comparable Ca concentrations about 20 mmol/L. After the breakthrough a rapid decrease of the Ca concentration occurs in both experiments.

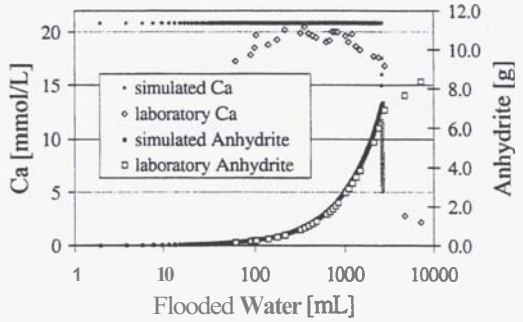

Figure 5: Comparison of laboratory and simulated data of the concentration of calcium in the solution and the total amount of anhydrite dissolved during the simulation versus the amount of water flooded through the core.

4. DISCUSSION

4.1 Comparison of Experiment and Simulation

Set up of the Allennohe model is done following the published data of Baermann et al. (2000b). Without knowing the exact initial distribution of anhydrite in the core sample P6, it is assumed that an initial heterogeneity at the center of the core inlet develops to a preferential flow path in the otherwise homogeneous core. At first two channels develop until one stops growing and the remaining flow path grows wider, towards the center of the core, and incorporates the second one. Thus, one finger survives and reaches the outlet of the model (Figure 1).

For further simulations it is planed to use digital X-ray tomography images to deduce the real initial anhydrite distribution within the core samples.

However, it is shown that the approximations done here provide very good correspondence between theory and praxis. The results of the numerical simulation coincide with the observations during the core flooding experiment. Breakthrough occurs after 40 days in the experiment and 48 days within the numerical simulation (Figure 2).

The dissolution of anhydrite results in an increase of porosity and permeability within the preferential flow path. The hydraulic conductivity of the entire core is determined by the permeability of the channel, because the main water body flows through it. The resulting permeability development within the laboratory and numerical experiment is similar (Figure 4). The dissolved mineral amounts in the experiments coincide extremely well (Figure 5).

The concentration of calcium in the solution is about 20 mmol/L, which reflects almost the concentration of anhydrite in thermodynamic equilibrium with pure water at 20°C. When the breakthrough occurs the calcium concentration decreases rapidly to resulting 2.2 mmol/L. An explanation for the low calcium concentration is, that water which is still in equilibrium with anhydrite is diluted by water flowing through the flow path. This water does not get in contact with anhydrite because in the preferential flow path the anhydrite is completely dissolved and is therefore free of Ca^{2+} ions.

In both the laboratory as well as the numerical experiment, it is shown that in the case of channel formation, due to reaction front instability, a sudden permeability increase can occur after short time periods. In the case study a stable reaction front, which means total dissolution of anhydrite within the core, would require more than sixfold the observed time.

4.2 Feasibility Study to Stimulate the Allermöhe Aquifer

The numerical simulation of the core flooding experiment shows that the permeability development in the anhydrite cemented sandstone is induced by reaction front instability and resulting channel formation. The question arises: can the aquifer around the bore hole be stimulated like the core samples? If this is the case, it might be possible to connect the less permeable area around the bore to areas with higher permeability in short time scales. Therefore, the possibility of preferential flow path development around the bore Allermohe is checked evaluating the Peclet and Damkohler numbers.

The Allermohe bore has a depth of 3305 m. The thickness of the reservoir is 73 m and the aquifer has a temperature of 125°C. The thermal water is of high salinity with concentrations of Na^+, Cl^-, Ca^{+2}, and SO_4^{2-} and others resulting in 218 g/L total dissolved solids.

The dimensionless numbers of Peclet and Damkohler are used to predict preferential flow path development within the flow system. According to equations (1) and (2) the channel formation depends on the appearing flow velocities in the system. Therefore, it is not sufficient to prove channel development in core flooding experiments only, but to show it for the natural systems, too.

The kinetic coefficient for the mineral reaction has to be determined. The kinetic coefficient k in equation (2) is:

$$k = r \cdot w_{mol} \cdot A \left[\frac{mol}{m^2 \cdot s} \frac{kg}{mol} \frac{m^2}{kg} \right] \qquad (4)$$

Anhydrite dissolution rates have been determined to be between $r = 0.2*10^{-7}$ in a solution of 100°C without sodium chloride and $3.9*10^{-7}$ mol/m²*s in a solution of 25°C and a concentration of 200 g/L. For the conditions of the bore Allermohe (100°C, 200 g/L) $2.0*10^{-7}$ mol/m²*s has been detected (Bartels and Iffland, 2000). With the molar weight of anhydrite w_{mol} of 0.136 kg/mol and the mean of the inner surface A of 149 m²/kg the kinetic coefficient is $4*10^{-11} s^{-1}$.

The characteristic length l, in this case the thickness of the aquifer, is 73 m. The velocity around the bore due to injection can be determined from Darcy's law:

$$v = \frac{q}{\phi} = \frac{k_f \cdot I}{\phi} \qquad (5)$$

The hydraulic conductivity k_f at 125°C corresponding to 3 mD is $1.33*10^{-7}$ m/s. The sphere of influence around the bore R can be estimated after Sichardt (1928) with the draw down s. The technical limit of 50 bar excess pressure is used here corresponding to a draw down of 500 m.

$$R = 3000 . s \cdot \sqrt{k_f} = 547 \text{ m} \qquad (6)$$

The hydraulic gradient I can be calculated as:

$$I = s / R = 0.9142 \text{ [-]}. \qquad (7)$$

This leads to a velocity v of $2.03*10^{-6}$ m/s.

Dispersion coefficients have to be estimated. Gelhar (1986) compiled an extensive number of dispersivity data from many field sites around the world. Most of the observed dispersion lengths lie within 0.01 to 1 times the observation length, additionally a trend of asymptotic behavior of the dispersion length versus observation length could be determined. Assuming a relatively homogene-

ous aquifer at Allermöhe and considering the long observation length a dispersivity between 5 m and 10 m seems reasonable.

The resulting Damköhler number is 144 which is above 1, therefore, leading to an instable reaction front. The resulting Peclet number will be between 14.6 and 7.3, for dispersion lengths of 5 m and 10 m, respectively.

All together the development of preferential flow paths within the vicinity of the bore Allermöhe seem possible, if the calculated flow velocities can be reached.

5. CONCLUSIONS

Simulating the core flooding experiment with the numerical code SHEMAT leads to acceptable results. Breakthrough time, mass of water, calcium concentration and amount of dissolved anhydrite coincide with the laboratory experiment.

The behavior of the Allermöhe core samples within the laboratory experiments could be explained with the development of preferential flow paths. This has also been shown by mineralogical investigations and emphasized with the numerical study shown here.

After further digitalization of mineralogical data the core model will be improved and calibrated. After verification the model will then be extended to simulate the vicinity of the Allermöhe bore as well as to further estimate the possibility of gentle stimulation.

6. ACKNOWLEDGMENTS

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