Geochemical Constraints on the Operations of High Temperature Aquifer Energy Storage (HT-ATES) in Abandoned Oil Reservoirs

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ABSTRACT

Heat for industrial processes and housing is an important part of the energy consumption. In Germany it accounts for more than 50 % of the total demand. The need for a sustainable heat production and/or the use of existing waste heat is key for the energy transition. Many industrial applications produce large amounts of heat, which cannot be used in the summer months. Heat storage is of increasing importance and technical solutions are widely discussed. In this context, the reuse of abandoned oil reservoirs as aquifer thermal energy storage systems is receiving even greater attention. Their widespread distribution and the very often already existing data and pre-knowledge of the future reservoir is very advantageous. Numerous aquifer thermal storage systems in the low-temperature range (25 °C < T < 40 °C) have been realized worldwide and are currently in operation. High temperature systems (T > 100 °C) with the advantage of higher efficiency and a better match to the requirements of process heat are completely lacking. Especially large temperature differences between storage-, formation- and injection temperatures add complexity to the operations, particularly by increasing significantly the perturbation of the geochemical system, leading to an increased scaling potential in the surface installations as well as in the reservoir. To address this gap of knowledge the Karlsruhe Institute of Technology plans establishing the demonstrator DeepStor in an abandoned oil reservoir below its campus with a foreseen half-yearly cyclic high-temperature energy storage and reproduction.

Herein, the assessment of a reservoir and the hosted geothermal fluid in the marine sandstones of the Meletta beds, part of the Cenozoic graben filling, in around 1250 m to 1350 m depth is demonstrated. Main focus is on the expected fluid-solid interactions with the associated scaling potential. In a first step, the unperturbed formation temperature, key parameter controlling the chemical system and therefore the basis for further analyses, is calculated using the MulT_predict multicomponent geothermometer. Then, the old and rather low-quality wellhead fluid data is corrected applying an equilibrium approach. Finally, the scaling formation is modelled, both for the testing phase (single-borehole application), where formation water needs to be stored in an open pond for later re-injection, as well as during the cyclic storage and production phase with two boreholes (cold and warm side of the reservoir). It is shown that the major scaling phases are iron hydroxides, calcite, celestite and barite. During the testing phase the change of the chemical system to ambient p/T-conditions and the exposition to atmospheric oxygen are the driving forces for the precipitation within the pond. During regular two-well operations a cyclically recurring process of dissolution processes in the reservoir and precipitation in the surface installation is evolving and remains stable in the long term.

1. INTRODUCTION

The global climate change requires a fast transition towards renewable energies. In this process the heat supply is playing a key role. For example, in Germany the share of heat is more than 50 % of the total energy consumption of which still nearly 80 % is provided by fossil fuels (AGEB, 2022; Statistisches Bundesamt, 2021). A sustainable and emission-free heat supply is one of the central tasks of the energy transition. Also the use of waste heat can play a major role in the future efficient utilization of the energy resources. One typical problem for a year-round heat supply is the seasonal fluctuation of the demand. While it is high in the heating period, heat production often becomes uneconomical in the summer months. This leads to heat surpluses from heat production facilities and from industrial waste heat. One concept to use this surplus of heat, which is gaining more and more attention, is geothermal storage, the so-called Aquifer Thermal Storage Systems (ATES).

Numerous ATES operating with low temperatures (nearly all of the installed systems have storage temperatures T < 40 °C) have been built for demonstration purposes or are already in operation (Fleuchhaus et al., 2018). For district heating systems and process heat, however, higher temperatures are usually required, often well above 100 °C. Thermal aquifer storage in this temperature ranges, so-called HT-ATES, has the advantage that the heat can be used directly without additional heat pumps and that high temperature waste heat can be used as the source (Wesselink et al., 2018). However, such systems have not been successfully realized yet. The storage of high temperature fluids is still subject of ongoing research and recently a topic of great interest. In order to close this gap the Karlsruhe Institute of Technology is working on the establishment of the demonstrator site DeepStor on its campus. DeepStor intends to realize the very attractive and advantageous concept of using an abandoned oil reservoir for heat storage. This allows to make use of existing exploration and production data. Such pre-knowledge and operational experience reduce project risks and exploration efforts.

Most previous attempts of establishing an HT-ATES failed due to hydro-chemical problems of scaling and corrosion and reservoir clogging (Fleuchhaus et al., 2018). Scaling is a common issue for geothermal applications. Numerous minerals can precipitate as a consequence of chemical equilibrium perturbation by geothermal production. Amorphous silica, carbonates sulfates, sulfides, hydroxides, halides and native metals have been observed at worldwide geothermal operation facilities (Nitschke et al., 2014). In this context, the

operation of a doublet thermal storage system poses a particular challenge associated with the fact that a water body is moved cyclically from a warm to a cold reservoir and back, interfering the chemical system again and again. The high temperature differences between the cold and the warm side of an HT-ATES cause potentially larger quantities of precipitation and dissolution compared to low-temperature systems. Knowledge and quantification of these processes prior to operations support the design of the operation scheme, can provide insights into the long-term reservoir behavior, and can help to conceptualize mitigation strategies.

Herein, a case study demonstrates the assessment of the chemical system of "reservoir rock-geothermal fluid-scaling" by analyzing an old fluid sample from the nearby Leopoldshafen oilfield. It focuses on the expected mass transfers between the geothermal fluid and the solids under specific operation conditions. The main aspects covered are the determination of the unperturbed reservoir temperature as the basis for further geochemical modelling as well as scaling formation and potential reservoir clogging. The critical minerals often precipitate from the Upper Rhine Graben (URG) fluids under the locally typical production schemes are barite, celestite, and calcite (Nitschke et al., 2017, Scheiber et al., 2014, Herzberger et al. 2010). Beside of these, also iron hydroxides, anhydrite and amorphous silica which could become relevant for specific operation parameters have been considered.

2. DATA & METHODS

2.1 Study Site and Data

The study site is located in the URG in South-Western Germany. Here, in the framework of the DeepStor project, it is planned to develop a HT-ATES as a research and demonstrator facility on the Campus of the Karlsruhe Institute of Technology about 10 km north of the city center. In the URG oil is being or has been produced from several Mesozoic and Tertiary sedimentary units. Due to their hydraulic properties and the depth with associated suitable formation temperatures, the calcite-cemented fine-grained sandstones of the Meletta beds in approximately 1200 m – 1300 m depth have been identified as the targeted reservoir formation. A detailed stratigraphy is displayed in Stricker et al. (2020).

Very nearby, only few kilometers to the north-west, the Leopoldshafen oil field has been operated for decades. Drilling data and geochemical fluid data exist from the late 1950's when first parts of the field have been developed. This study is based on the geochemical analysis of the fluid sample (Leo_13) from the Leopoldshafen 13 well (Tab. 1). The fluid was already sampled and analyzed in 1959. Therefore, the chemical composition is subject to larger uncertainties. Key constituents in terms of scaling forming minerals, such as barium and strontium, have not been analyzed. Hence, a quality assessment and adaption of such data is required prior to scaling analysis (chapter 3.1). For numerically attaining a fluid-reservoir rock equilibrium, the mineralogical composition of the Meletta sandstone given in Banks et al. (2021), who analyzed reservoir rock analogues from a quarry nearby, was used.
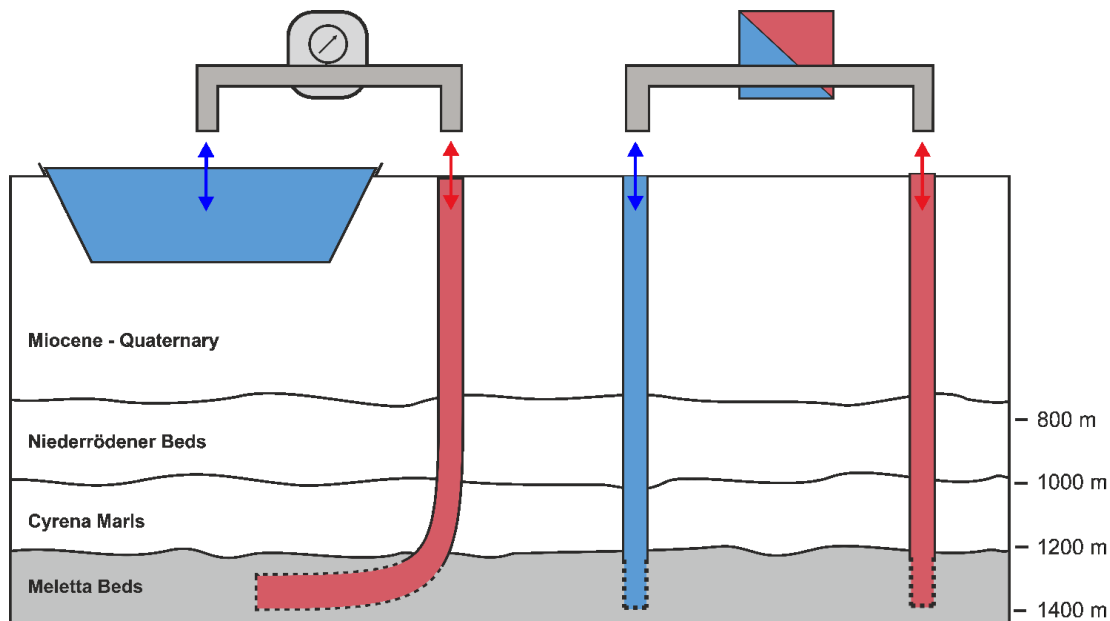

Figure 1: The schematic concept of the two phases of the DeepStor HT-ATES. During the testing phase (left side) the reservoir is tested via a single well using an open pond as a fluid buffer. In the regular operation phase (right side) a doublet will be used to establish a continuous warm (120 °C) and cold (60 °C) reservoir, respectively.

2.2 Reservoir Temperature Modelling

The unperturbed reservoir temperature is a key parameter for the design of the later operation scheme of the thermal storage system since it governs the equilibrium of the chemical system of fluid and reservoir rock. Any deviation, on the warm side and the cold side of the reservoir will imbalance this equilibrium and may induce dissolution or precipitation of mineral phases. Since the existing temperature

logging data is expected to be strongly influenced by the drilling activity, reservoir temperature determination is done based on the chemical composition of the fluid using multicomponent geothermometry. Such methods (e.g. Ystroem et al., 2020; Palmer, 2014; Nitschke et al., 2017; Spycher and Reed, 1984) have been proven to yield precise results and being very robust against interferences from possible analytical errors and secondary processes such as mineral precipitation and dissolution, dilution or boiling. The reservoir temperature is determined based on the temperature-dependent dissolution-precipitation reaction of reservoir rock minerals and the fluid. The base assumption is the simultaneous equilibrium of the fluid with the reservoir rock minerals at reservoir conditions. The evaluation of the mineral's saturation states over temperature leads to a range of equilibrium temperatures for a pre-defined mineral set.

Herein, the tool MulT_predict was used. The implemented optimization algorithm, back-calculates the fluid sample composition automatically on reservoir conditions correcting from possible alteration by secondary processes. MulT_predict is based on IPhreeqc version 3.7 coupled to MATLAB for the statistical evaluation and optimization. A detailed description of the method and the selection of the mineral set is described in Ystroem et al. (2022).

2.3 Scaling Potential Modelling

The geochemical modelling presented herein was performed using PhreeqC version 3.7 (Parkhurst and Appelo, 2013). Due to the highly saline brine with a TDS of 120 g/L the Pitzer formalism and the associated thermodynamic data (PhreeqC-pitzer.dat) was used to determine the activities of the aqueous species. Scaling assessment modelling was performed for the two phases of reservoir operations, the testing phase with a single well and an open pond and the operation phase with two boreholes establishing a warm and a cold reservoir. The two configurations are displayed in figure (1) and is described in the following sections.

2.3.1 Testing phase

To assess the scaling potential during the testing phase two production cycles are modelled. Firstly, the initial unperturbed formation fluid with and adapted chemical composition (section 3.1) and the numerically determined in-situ temperature (section 3.2) is produced to an open pond. The amounts of precipitates of calcite, barite, celestite and iron hydroxides are modelled. After that the fluid is heated to storage temperature and re-injected to the reservoir, where a new equilibrium is calculated for reservoir conditions enabling the dissolution of the scaling relevant minerals quartz, calcite, barite and celestite. Afterwards, this secondary fluid is also produced to the pond. Modelling the amounts of scaling, the technical processes of cooling, heating, relaxation end exposure to the atmosphere (CO_2-equilibrium and oxidation) are distinguished. The storage temperature is set to 120 °C at hydrostatic pressure of 140 bar. The temperature of the pond is assumed to be 10 °C at atmospheric pressure of 1 bar. All precipitation and dissolution processes have been determined based on equilibrium reactions. Kinetic effects have been neglected.

2.3.2 Operation phase

During the operation phase, the HT-ATES will be operated via two boreholes establishing a permanent warm reservoir and a cold reservoir, respectively. The fluid will be cyclically moved from one reservoir to the other and back undergoing temperature and pressure changes. In order to evaluate the long-term behavior of the HT-ATES, five storage-production cycles have been modelled representing five years of operations. The base case which is considered for this study is 60 °C at the cold reservoir and 120 °C at the warm reservoir at hydrostatic pressure (140 bar). The surface installation pressure is 10 bar. The mass transfers between the fluid and the solids have been assessed separately for the surface installations and reservoir. All precipitation and dissolution processes have been determined based on equilibrium reactions. Kinetic effects have been neglected.

3. RESULTS

3.1 Data Adaption

The quality assessment of the fluid sample Leo_13 reveals that it is rather of low quality. Secondary processes may have altered the composition. It is implausible that calcite is oversaturated under reservoir conditions as well as quartz is strongly undersaturated. Furthermore, the analysis does not comprise data for strontium and barium, which is required for sulfate scaling analysis. Therefore, prior to the scaling assessment, the fluid sample was adapted (Tab. 1).

Barium and strontium concentrations were taken from Sanjuan et al. (2016). They analyzed fluid samples from the same lithological formation of the Landau oilfield wells located approximately 25 km north-west of the study area. For a worst-case calculation the maximum measured concentrations of 7.6 mg/L barium and 308 mg/L strontium have been transferred to sample Leo_13.

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Table 1: Chemical composition of the wellhead sample Leo_13 of the Leopoldshafen 13 oilwell. Parameters in darker grey fields have been numerically adapted back to reservoir conditions or completed by literature values (Landau wells from Sanjuan et al., 2016) to account for the not measured constituents barium and strontium), measurement errors and sample alteration. Concentrations are displayed in mg/L.

Parameter	Leo_13	Adapted composition
pH	6.0	5.84
Na^+	41'000	41'000
K^+	256	256
Ca^{2+}	4'650	4'650
Mg^{2+}	705	705
Fe^{2+}	107	107
Ba^{2+}	-	7.60
Sr^{2+}	-	308
NH_4^+	83	83
$SiO_2(aq)$	6	19
Cl^-	72'800	72'800
SO_4^{2-}	392	392
HCO_3^-/CO_3^{2-}	330	370

To adjust the calcite equilibrium, CO_2 (which potentially has been degassed during sampling) was stepwise added under reservoir conditions decreasing the calcite saturation until equilibrium was attained. The concentration of the carbon species consequently increases from 330 mg/L to 370 mg/L and the pH slightly decreases to 5.84. This fluid was then used to adjust the aqueous SiO_2 concentration by calculate the quartz equilibrium under reservoir conditions. The aqueous SiO_2 concentration increases from 6 mg/L to 19 mg/L.

3.2 Unperturbed Reservoir Temperature

The temperature determination with MulT_predict after optimization yield a mean temperature of 57 °C (Fig. 2). The determined temperature was used as the initial state of the chemical system minerals-fluid at reservoir conditions for the later assessment of scaling and reservoir clogging risk. At these conditions neither dissolution nor precipitation occur. Changes to these conditions caused by the technical operations will change this equilibrium. The consequences in terms of mass transfer between fluid and solids are analyzed in the following sections.
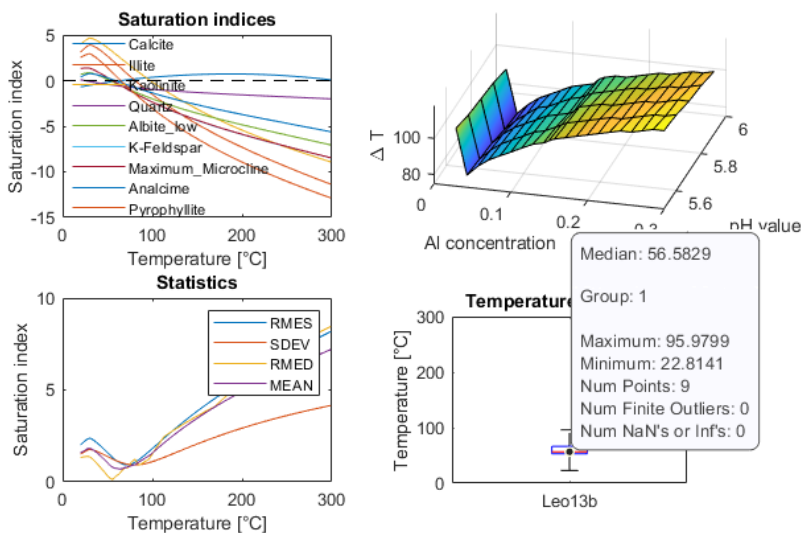
Figure 2: Results of the unperturbed reservoir temperature in Meletta beds in 1250 m in the Leopoldshafen 13 well using MulT_predict. The mean temperature is determined to be 57 °C.
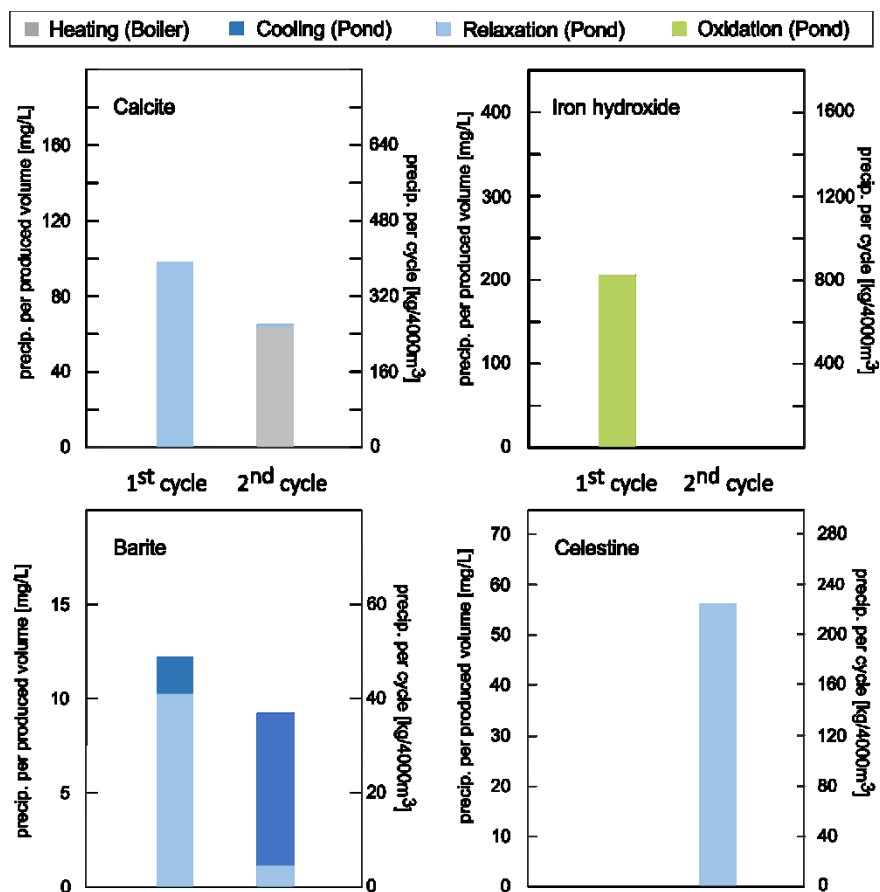

Figure 3: The amounts of scaling (calcite, iron hydroxide, barite, and celestite) that are expected during the testing phase. Grey color indicates precipitation induced by heating, dark blue by cooling, light blue by fluid relaxation and green by oxidization.

3.3 Scaling During Testing

During the testing phase it is planned to store the fluid after production in an open pond. This leads to cooling and relaxation to ambient conditions (10 °C, 1 bar) including gas equilibrium with the atmosphere (in particular CO_2 concentration of 415 ppm). For testing the re-injection, the fluid is heated up with a boiler to 120 °C.

Calcite, barite, celestite and iron hydroxide are oversaturated at ambient conditions in the pond and will form scalings (Fig. 3). The initial production of the reservoir fluid to the pond leads to a precipitation of ~100 mg/L (~400 kg/cycle) calcite due to relaxation and equilibrium with the atmosphere. Having a retrograde solubility ~60 mg/L (~240 kg/cycle) calcite will form as a consequence of the heating in the boiler. Being exposed to the atmosphere the aqueous iron(II) is oxidized and immediately precipitates as iron(III) hydroxide. Virtually the entire dissolved iron, ~200 mg/L (~800 kg/cycle), will precipitate at the first production cycle. If no large scale reducing processes happen, the once oxidized fluid has no iron dissolution capacity despite being iron depleted. It will not cause any more iron precipitation being produced again. During the first production cycle ~12 mg/L (~48 kg/cycle) barite is formed. The main driver is the relaxation of the fluid to ambient conditions. The cooling only plays a minor role. During the second cycle almost ~10 mg/L (~40 kg/cycle) are forming. This time, cooling controls the precipitation. During the first cycle no celestite scaling is to be expected. During the second cycle, fluid relaxation causes nearly 60 mg/L (~240 kg/cycle) celestite scaling.

3.4 Scaling During Operations

For the cyclic operation phase the observation points are separated into cold and warm side of the reservoir as well as cold and warm side of the surface installations. For the surface installations the processes of cooling/heating (heat exchanger) and relaxation are distinguished. Only calcite, barite and celestite are the relevant minerals causing scaling over the course of long term cyclic production at base case conditions with a storage temperatures of 120 °C at the warm side and 60 °C at the cold side. Silica scaling as well as anhydrite/gypsum scaling can be excluded (see section 3.5.2 and 3.5.3). Iron hydroxides are not precipitating when the contact of the fluid to the atmosphere is avoided.

After the fluid has once been produced and stored at the cold and the warm side of the reservoir, a stable behavior in terms of the amounts of mineral dissolution and precipitation can be observed from the second cycle (Fig. 4). Calcite (~65 mg/L) is mainly precipitated at the warm side of the surface installations caused by the heating up of the fluid before being stored, only minor amounts are forming due to relaxation at the warm and cold side. Nearly the entire amounts of precipitated calcite have priorly been dissolved at the cold side of the reservoir. Cooling in the heat exchanger cause ~6 mg/L of barite scaling, only subordinate amounts precipitate due to fluid relaxation. The amounts which are precipitating are priorly dissolved at the warm side of the reservoir. Celestite precipitates at the warm side of the surface installations (~43 mg/L due to heating and ~51 mg/L due to relaxation) and at the cold side (~60 mg/L due to relaxation). In the reservoir ~100 mg/L are dissolved at the cold side and ~50 mg/L at the warm side.
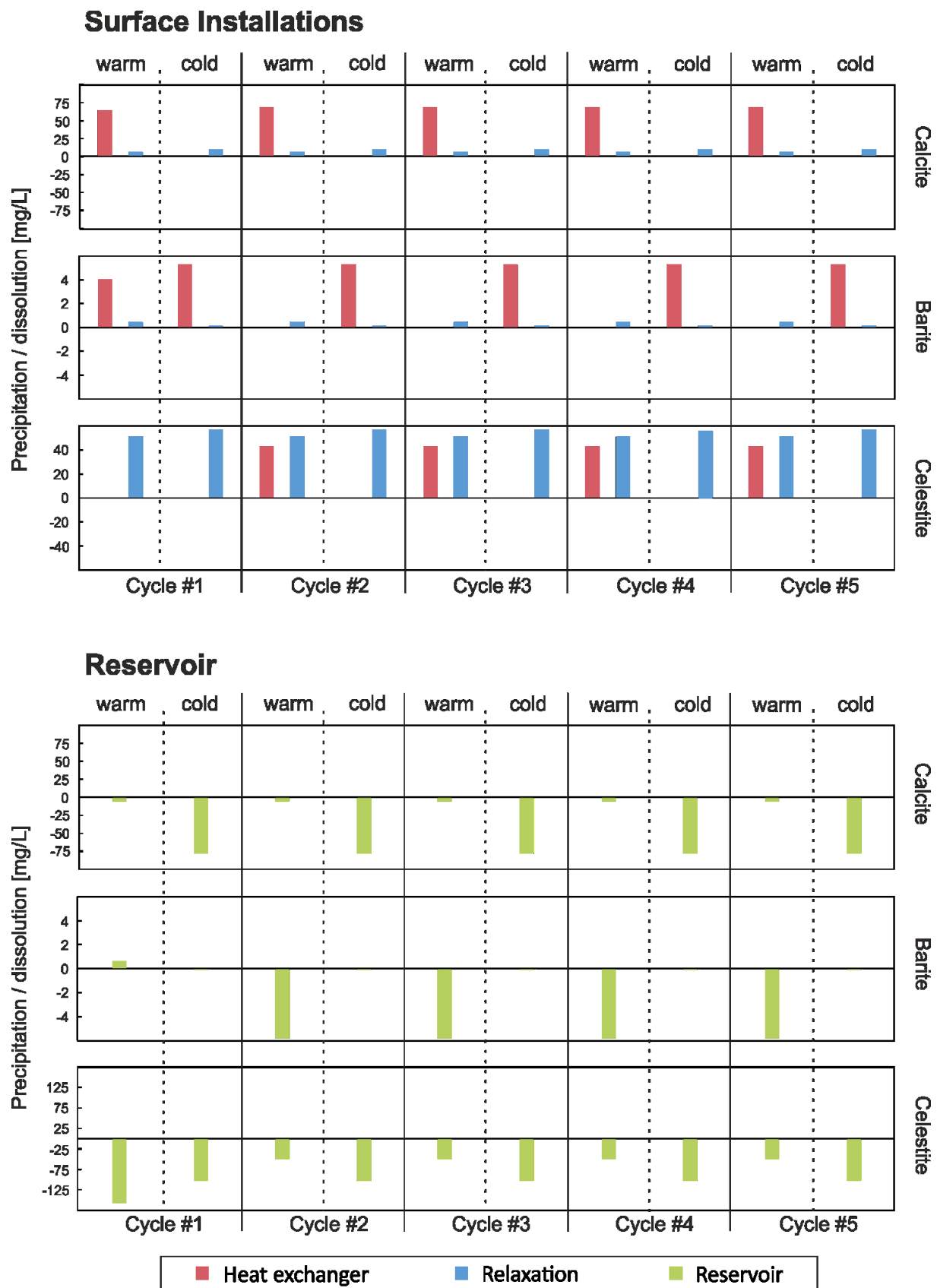
Figure 4: Results of the amounts of scaling (calcite, barite, and celestite) that are expected during five cycles (i.e. five years) operation phase. Red colors indicate scaling induced by heating or cooling the fluid, blue by fluid relaxation and green indicates precipitation and dissolution in the reservoir.

3.5 Sensitivity Analysis and Design Calculation

3.5.1 Surface Installation Pressure and CO_2 Degassing

The degassing pressure of CO_2, the so-called bubbling point, is a key parameter for the design of a geothermal power plant or the operation scheme of an HT-ATES. Relaxing the pressure of a fluid during production, the bubbling point is the pressure when a gas phase starts to form by degassing from the fluid. With degassing of CO_2, the activity of carbonic acid decreases and the pH increases. As a consequence, the saturation index of calcite starts to increase (Fig. 5). Further relaxation increases the slope of the saturation index over pressure function and leads to increasing amounts of calcite scaling. The bubbling point at the DeepStor site for the initial reservoir fluid (57 °C) and the fluid produced from the cold side of the reservoir (60 °C) is 5.8 bar. For the same fluid at 120 °C (warm reservoir side conditions) the bubbling point is at 6.5 bar. The fluid for the initial conditions and at the cold side of the reservoir is in equilibrium with calcite (log(SI) = 0). The initial fluid heated up to 120 °C will be oversaturated (log(SI) = 0.8) driven by temperature. In this case, calcite scaling will form also for pressures higher than the bubbling point.
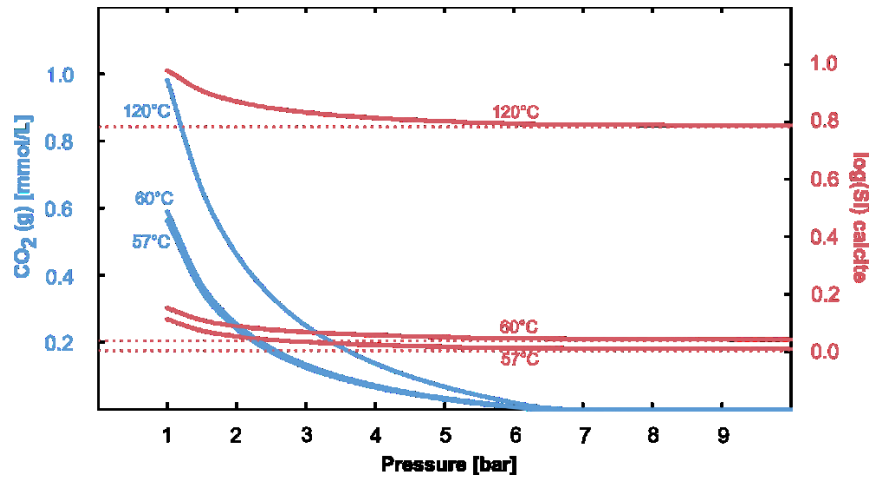

Figure 4: Results of the design calculation specifying the installation pressure to avoid CO_2 degassing for the initial reservoir fluid composition for unperturbed reservoir temperature (57 °C), for the cold reservoir fluid (60 °C) and for the warm reservoir fluid (120 °C). It is shown that from the CO_2-bubbling point (5.8 bar and 6.5 bar) the saturation index of calcite is increasing.

3.5.2 Anhydrite Saturation Temperature

For the base case conditions neither gypsum nor anhydrite scaling will form. Both minerals are undersaturated for the operational temperature range (Fig. 6). The reason is that both minerals do not occur quantitatively in the reservoir. The fluid will not equilibrate with these minerals at reservoir conditions. This is also underlined by the composition of the initial reservoir fluid, which is strongly undersaturated with respect to these minerals at unperturbed conditions.
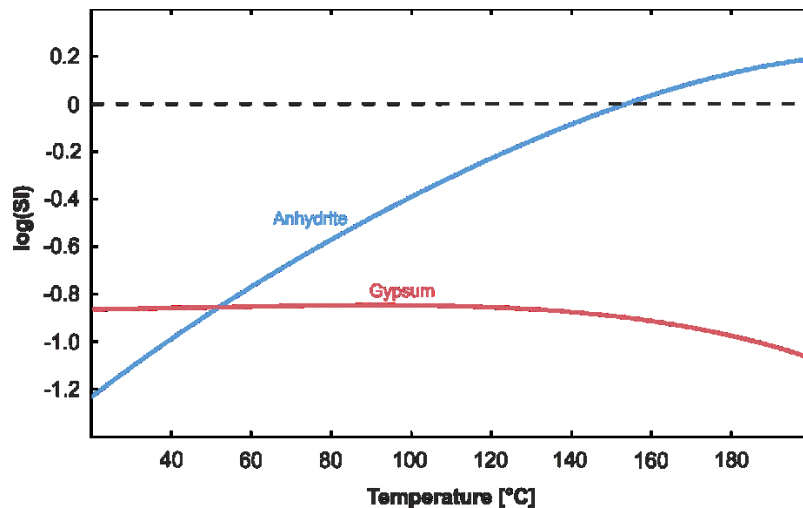

Figure 5: Sensitivity analysis designing the maximum storage temperatures in order to avoid anhydrite scaling/reservoir clogging. From ~150 °C the initial reservoir fluid composition is oversaturated with respect to anhydrite.

For the design of future operation schemes the retrograde solubility behavior of anhydrite needs to be taken into account. For the initial fluid composition at the DeepStor site, ~150 °C would be the temperature limit for which the anhydrite saturation is exceeded (Fig. 6). The retrograde solubility behavior is a particular challenge for this kind of operational scheme (permanent cold and warm side of the system), since it can potentially lead to reservoir damage/clogging at the warm reservoir.

3.5.3 Amorphous Silica Saturation Temperature

Silica scaling is well known to be a major challenge to geothermal operations. The evaluation of the scaling risk is a bit more complex than for other mineral systems since the two SiO_2 polymorphs quartz and amorphous SiO_2 with very differing dissolution and precipitation properties must be considered. A good assumption is that a geothermal fluid at reservoir conditions is in equilibrium with quartz. Quartz has a prograde solubility. The cooling of the fluid leads to oversaturation, however without causing quartz precipitation due to very slow kinetics. Further cooling can potentially lead to an oversaturation with respect to amorphous silica with a rather fast precipitation. Therefore, every reservoir fluid which is in equilibrium with quartz has a lower cooling limit, which is controlled by its silica saturation.
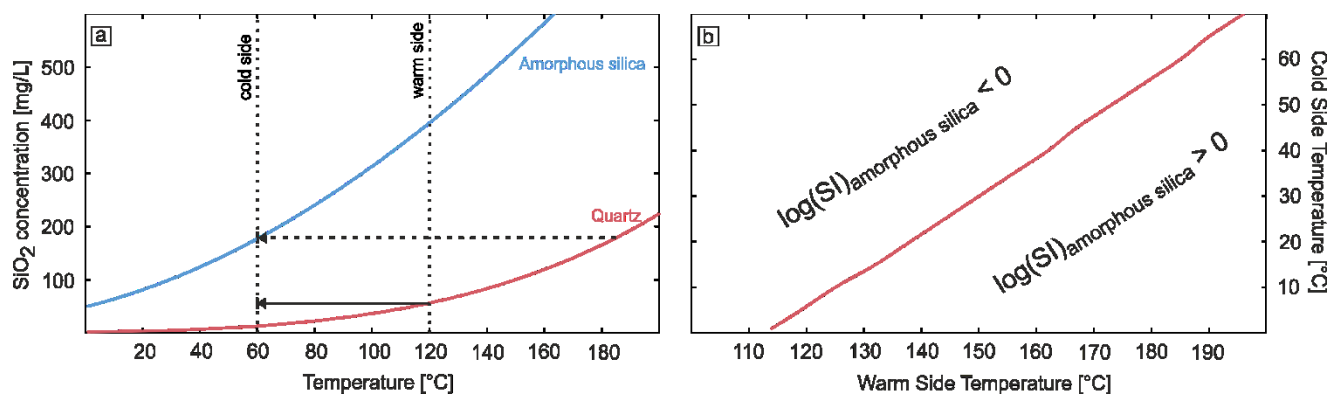

Figure 6: a) The modelled saturation functions for quartz and for amorphous silica for the initial reservoir fluid reveal that the fluid in the base case equilibrating at 120 °C with quartz is far from precipitating amorphous silica at the cold side (60 °C). b) Value couples of the system's maximum warm side temperatures (quartz saturation temperature) are plotted versus the system's minimum cold side temperatures (amorphous silica saturation temperature).

In figure (7a) the modelled solubilities for quartz and amorphous silica for the DeepStor fluid are displayed. It is shown that for the base case, the fluid equilibrated with respect to quartz at 120 °C is far from being oversaturated for amorphous silica at 60 °C. The amorphous silica saturation concentration is equivalent to a quartz saturation temperature of 185 °C, which would be the maximum temperature of the warm reservoir for a later cooling to 60 °C. For designing the operations with regard to the amorphous SiO_2 saturation, the temperature value couples of the warm (quartz saturation) and cold (silica saturation) reservoir are given in figure (7b).

4. CONCLUSION

The chemical system of an HT-ATES and the associated fluid-solid interactions are key processes for the successful operations of an HT-ATES. Scaling and reservoir clogging assessment are therefore the major exploration goals, even more so when further typical exploration-related information are already existing from previous nearby production activities from the same reservoir (oil production). This study reveals the information which are contained in an old fluid analysis of rather low quality and it demonstrates of how to make use of it to design the future storage operation scheme with regard to hydro-chemical boundaries prior to drilling.

In this study such a hydro-chemical exploration procedure is demonstrated as a case study assessing a Cenozoic sandstone reservoir in the Upper Rhine Graben in South-Western Germany by analyzing an old formation fluid sample from the time of oil production. As the basis for the chemical analysis the initial unperturbed chemical system of the reservoir must be defined. Therefore, the reservoir temperature is the key parameter. It was determined to be 57 °C, using the multicomponent geothermometer MulT_predict. It represents the reservoir equilibrium temperature at which neither dissolution nor precipitation happen. Based on that, operational induced perturbation of the system are calculated. During the test phase when the DeepStor reservoir will be operated from a single well by using an open pond as a buffer, four minerals are expected to produce scaling. In the first cycle producing the initial formation fluid to the pond, calcite (~400 kg/cycle), barite (~48 kg/cycle) and iron hydroxide (800 kg/cycle) form. After reinjection and re-equilibration at storage temperature of 120 °C celestite (nearly 240 kg/cycle), calcite (~240 kg/cycle) and barite (~40 kg/cycle) precipitate. During regular operation at the herein defined base case conditions ~65 mg/L calcite (warm side), ~6 mg/L barite (cold side), ~95 mg/L celestite (warm side) and ~60 mg/L (cold side) precipitate in the surface installations. These amounts have priorly been dissolved in the reservoir and will precipitate recurrently each cycle. The presented sensitivity analyses reveal the limits of an optimization of the base case conditions. The fluid at the DeepStor site requires a surface installation pressure of ~6.5 bar to avoid CO_2 degassing and consequently more abundant calcite precipitation. The anhydrite saturation temperature is at about 150 °C. For warmer temperatures anhydrite precipitates at the warm side of the system. The situation for silica scaling is very favorable. A cold side temperature of 60 °C would allow for a storage temperature of 185 °C. Operating with a storage temperature of 120 °C a cold side temperature of 10 °C would be possible.

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The situation that all the scaling forming minerals tend to dissolve under reservoir conditions is very positive. However, the model is based on equilibrium reactions. In future a kinetic approach could give better insights of the location of precipitation, when the factor of time is considered. In this context, the transfer of the herein attained information into a reactive transport model will add important spatial information of the chemical and mineralogical development of the reservoir.

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