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Laboratory Setups for Core Flooding and CT Scanning Experiments at In-Situ HP/HT Conditions

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ABSTRACT

To investigate porous media and to get better insights in geophysical rock properties experiments at in-situ high pressure and high temperature conditions will be performed. As a main part of the project a specially developed computer tomography- scanner (CT scanner) will be established in the lab. The experimental setup of the CT scanner will be extended by a core flooding unit to investigate the behavior of reactive fluids in flow through experiments. To simulate reservoir conditions the laboratory device is designed for high fluid pressures up to 200 bar and high temperatures up to 200 °C. In addition, it will be possible to use corrosive and volatile fluids such as supercritical CO_2 to investigate physical and chemical processes in different rock types. Experiments will be performed on cylindrical rock samples with lengths between 10 and 600 mm and diameters of 10 up to a maximum of 100 mm. Time resolved fluid sampling allows the analysis of changing rock properties and fluid composition during experiments. This will give new information of the interactions between fluids and rocks as well as factors like temperature, pressure, time etc. Additional measurements will be performed with newly configured laboratory devices. Thermal conductivity and thermal diffusivity will be performed measured with a device based on the optical scanning method. Hydraulic conductivity will be dtermined by so called oscillatory pore pressure method and fluid measurements be conducted by ICP-OES. The measurements will be used to get additional information on changing rock properties and to verify results.

1. INTRODUCTION

The key objective of this research project is to investigate experimentally the physico-chemical interactions of fluids, rocks and the cementation of geothermal wells. The influence of hydrothermal alteration on rock properties like permeability, thermal conductivity and thermal diffusivity is under investigation for typical rock samples like the Westerly Granite (USA) as well as different carboniferous silt- and sandstones of Germany, especially from the Ruhr Area. Some granites and volcanic rocks from the Upper Rhine Valley are also taken into account. The changing properties of well grouting materials and interactions with the often highly saline brines are also under research in a second step.

Dissolution experiments at different temperatures, pressures and with different fluids will be performed to get more insights in reaction kinetics and processes within the geothermal reservoir. For the analysis newly configured, laboratory devices will be used. The direction-dependent permeability is measured with the so called oscillatory pore pressure method. Simultaneously measurements of the upstream and downstream pressures and the upstream fluid flow permit the calculation of permeability and specific storage by two methods, the conventional upstream – downstream pressure analysis and the upstream pressure-flow analysis. The thermal conductivity and thermal diffusivity are measured with a device based on the optical scanning method.

The first laboratory experiments will be expanded step by step to get a greater understanding of the physico-chemical interactions of fluids, rocks and the cementation of geothermal wells.

2. ROCK SAMPLING AND PREPARATION

For the proposed testing procedures sandstone varieties were sampled. The so called Ruhr- (RSS), the Kaisberg- (KBS) and the Bentheim- (GBS) sandstone were used for first measurements. For the realization of the experiments a good mineralogical description within the geological context is of high importance.

2.1 Local Geology

For upcoming deep geothermal projects in the Ruhr-Area the geology of purposed target horizons is of special interest. The geology in this area is dominated by alternating sequences of sand-, silt and mudstones with interstratified coal layers in the Upper Carboniferous. The whole Ruhr-Area, especially the area of Bochum including the drilling zone Future Energy is characterized by mainly carboniferous rocks that are reaching in a depth up to four kilometers. The rocks were sedimented around 300 Ma ago in the subvariscian foredeep. Early sediments are characterized by marine sedimentation, while later sediments (Upper Carboniferous) were affected by more fluvial conditions. The eustatic sea level fluctuations have led to the typical reoccurring sequences of sandstone, siltstone, and mudstone with root clay, mudstone with high organic compound, siltstone and finally sandstone. During the variscan orogeny (late Permian to Triassic) the sediments were folded and buried and coal developed according to depth and temperature and the amount of organics. In this case stone coal was formed. The main folding structure is characterized by amplitude of several kilometers and show a general dive down of the layers in northern direction. As a primary target of geothermal exploration the sandstone layers of the deeper parts were identified. These sandstone layers are fine to coarse grained or even conglomeratic. Some quartzite's and greywacke's occur mainly in the deeper parts. The layering can be in the range of a few centimeters up to more than 50 meters. The lateral sedimentation can fluctuate on short distance because of the fluvial milieu, which is characterized by distribution channels. Correlations on greater distances are not possible in this case.

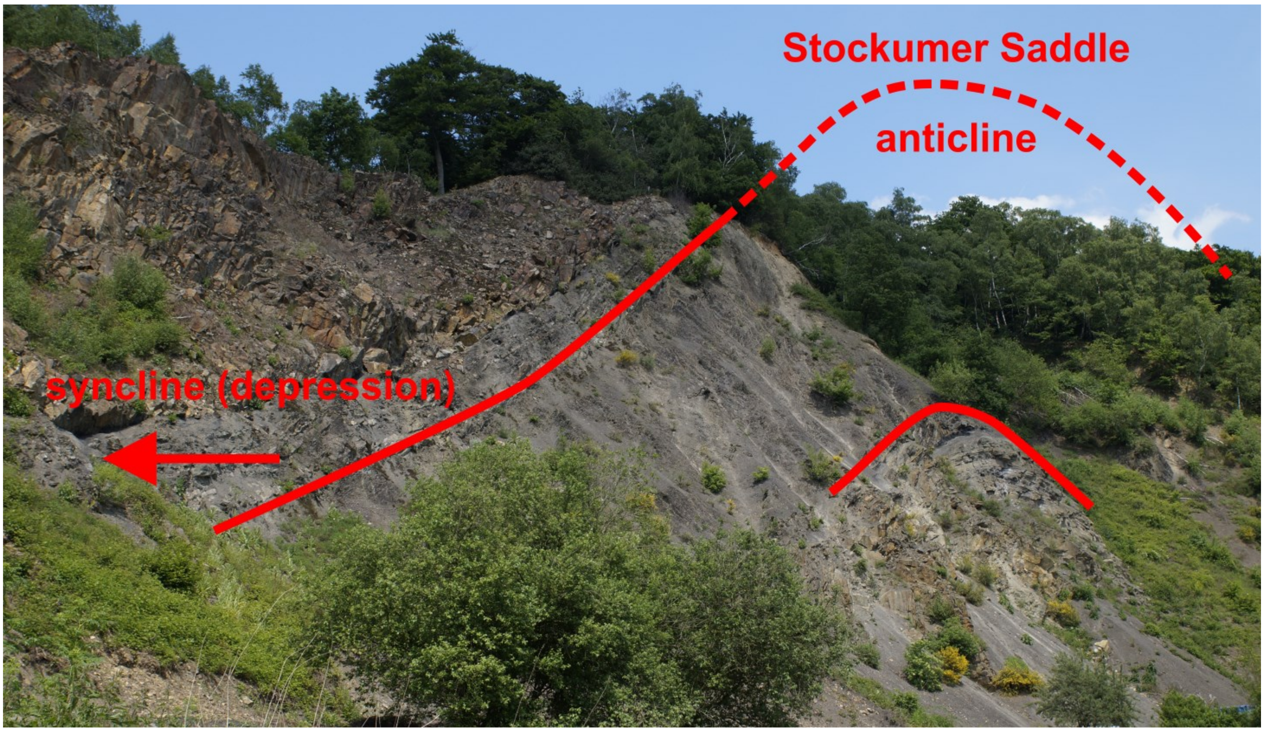

Figure 1: The photo shows the Stockumer saddle with a typical folding sequence of anticline and syncline. Thick-layered sandstones can be seen in the upper and lower part of the photo, while the thin layered sediments in the middle are composed of mud- and siltstones (photo: Lars Knutzen).

The Lower Carboniferous rock formations of the Namur A to Namur C with high amount of quartz-sandstone were chosen as primary sampling targets. The Lower Carboniferous is limited by the Devonian Alaunshists to the bottom. These formations are situated in 3 to 5 km depth in the area of Bochum. In the saddle core of the Stockumer saddle (see Figure 1) the border between the Carboniferous and Devonian can be expected to be in a depth of ca. 3500 m. Therefor outcrops in the south of Bochum in the area of Hagen were sampled. The Kaisberg sandstone (KBS) was sampled at an outcrop at the type locality of the Kaisberg itself next to the lake Harkort, while the Ruhr sandstone (RSS) was sampled in a still active quarry called "Imberg GmbH Natursteinwerke" north of lake Hengstey.

2.2 Mineralogical Description

Mineral determination was done with a standard polarization microscope and the VHX-2000 digital microscope by keyence including semiautomatic calculations with the VHX-2000 software by the means of threshold analysis. But due to the complexity of textures and the mineral appearance artifacts occurred. Unavoidable errors were corrected manually after the segmentation process. A lot of work was already done on this topic e.g. by Köse (2012))or Barraud (2006))when using the watershed segmentation method for example.

Diffraction patterns were recorded in reflection geometry with an Empyrean Theta-Theta diffractometer (Panalytical, Almelo) equipped with a copper tube, 0.25° divergent slit, 0.5° antiscatter slit (incident beam), 7.5 mm high antiscatter slit (diffracted beam), incident and diffracted beam 0.04 rad soller slits, and a position sensitive PIXcel-1d detector. The K-beta emission line is suppressed by a Ni Filter. For qualitative phase analysis the specimens were scanned in the 5-80° 2θ range with a step width of 0.0131° and 250 s collection time at an ambient temperature of T = 300 K. The ICDD powder diffraction file (PDF2) in conjunction with the HighScore Plus software (Panalytical, Almelo) was used for qualitative phase analysis.

The KBS is part of the Kaisberg-Formation, which is characterized by interlayering of sandstone- silt- and mudstones. The thickness of the layers ranges from decimeter to meter. The sandstone consists of 50 to 60 % quartz and approximately 40 % feldspar. The feldspar is highly altered. Sericitization and feldspar alteration led to a higher amount of Chlorite of around 5 %. The grains are usually subangular to angular and no preferred orientation or layering is visible. The grain size is around 250 µm. Some more conglomeratic parts can occur with quartz grains up to a maximum of 1 cm size.

The RSS is a widely used (historical) synonym for sandstones of the Namur-C up to the Westfal-A in the area of the Ruhr used by local companies, but is not anymore an official classification. The right classification can be sometimes very difficult because of the similarity between the different sandstone formations of the Carboniferous. The sampled RSS also consists of mainly quartz of around 60 % and feldspar (~ 40 %) with minor chlorite. The feldspar is heavily altered to sericite, which can be distinguished by cloudy appearance. This process is called sericitization. The quartz is characterized by undulose extinction Some parts of the formation contain a high amount of organic material such as lycopodiophytes called sigillaria and calamites. These parts were not chosen for sampling. Liesegang rings indicate a higher amount of iron in dependency of fluid circulations next to fractures.

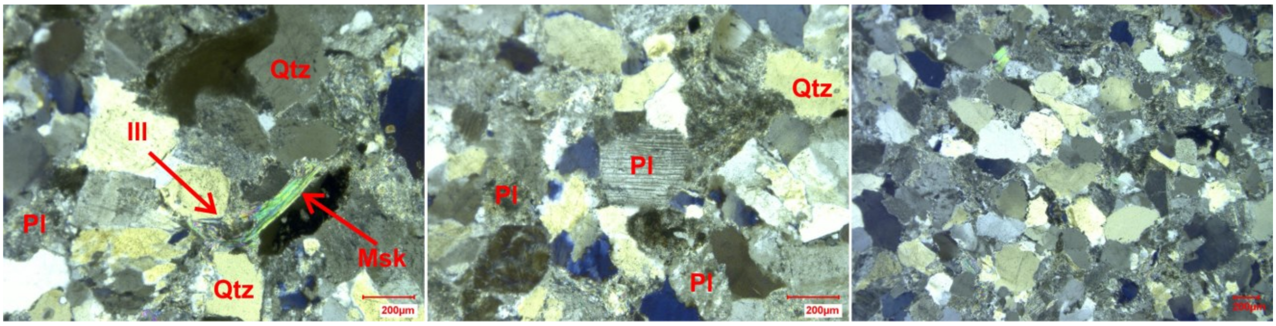

Figure 2: The cross-polarized photos shows the typical Ruhr sandstone with sub-angular quartz grains. Typical is partial replacement of the muscovite by illite (left) and sericitic alteration of plagioclase, which can be recognized by brownish colors and cloudy appearance. In addition polysynthetic twinning can be seen (middle).

XRD results of both sandstones, the KBS and RSS, strongly indicate that both sandstones are very similar regarding their mineralogical composition (compare to Figure 3). The main difference is the occurrence of orthoclase (K-feldspar) at the RSS. The thin section analysis confirms these results. Therefor for further analysis the fresh rocks of the quarry will be used. Anyhow the sandstones of the Carboniferous possess a high amount of clay minerals, which developed by alteration of the feldspars. In both sandstones chlorite and illite represent typical alteration products. Muscovite is commonly replaced by illite. The plagioclase is replaced by sericite. In addition the porosity is very low. Altogether the properties of these kind of sandstone is not ideal to perform first calibration measurements within the CT-Scanner and for the usage in dissolution experiments and subsequent water analysis at the newly configured ICP-OES. Because of these facts the Bentheim sandstone (GBS) was taken as a perfect, very homogenous sandstone example.

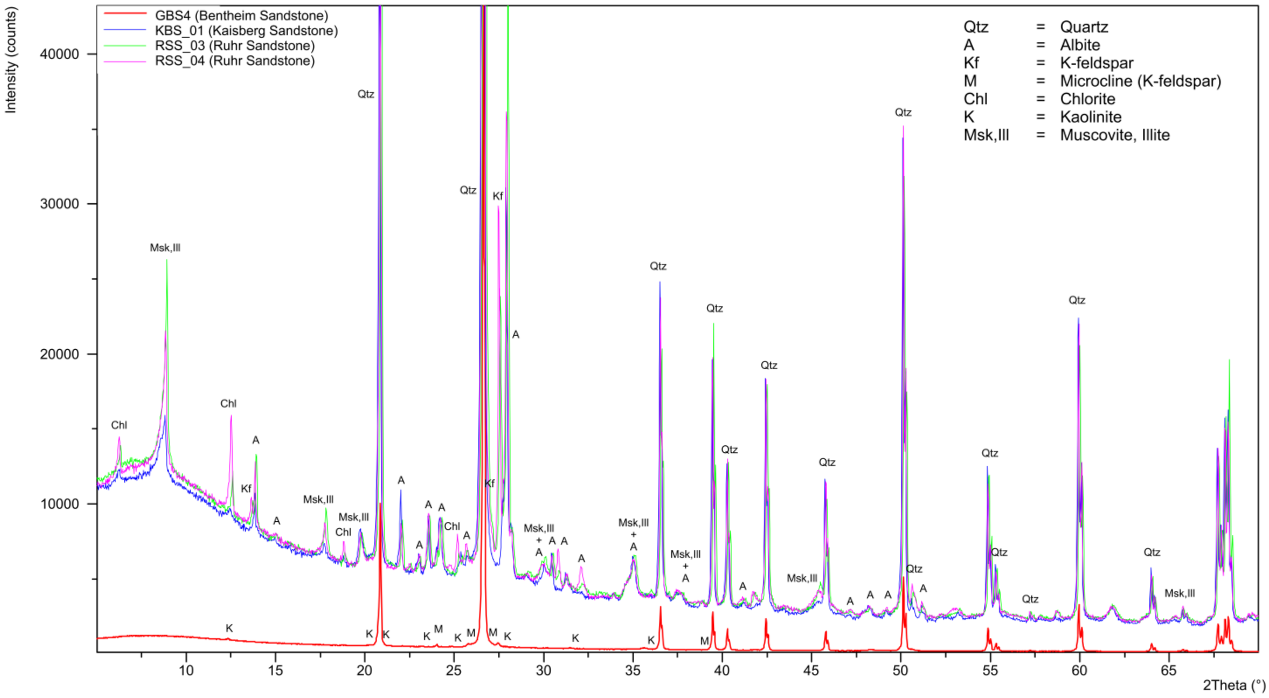

Figure 3: The figure shows XRD results of the different sandstones (RSS, KBS, GBS). The KBS and RSS are very similar in intensity and mineral composition. The only difference is the existence of K-feldspar in both RSS samples. The intensity of the GBS4 sample was downscaled by a factor of 10, to see better the differences in mineral composition.

The GBS is characterized by a mineral composition of around 90 to 95 % of quartz, and 2 to 5 % of K-feldspar as well as kaolinite (Klein and Reuschlé, (2003)). The feldspar is mainly microcline, which can be easily determined by the cross-hatched twin patterns in cross polarized light. The sandstone is well sorted with a uniform grain distribution. The GBS is medium grained with a grain size of ca. 300 µm. Grains are subangular. The grain supported sandstone has a silicic matrix. The GBS4 sample used for experiments consists of ≥ 90 % quartz, 7 – 8 % feldspar including microcline and 2 – 3 % kaolinite. The amount was determined by semi quantitative XRD analysis (see Figure 3). Previous work of Kemper (1963)) and Klein et al. (2001)) assume a porosity of between 20 and 25 %. The maturity of the sediment is indicated by the subangular grains of one grain size and the abundance of plagioclase.

2.3 Sample Preparation

Cylindrical samples with a diameter of 3 mm, 10 mm, 30 mm and 40 mm and varying length were drilled with a diamond bit. These cylindrical samples will be used for the CT scans. Different sizes allow different solutions at the scans. Furthermore slices were cut off with the rock saw for thin section preparation. One chunk about 3 to 4 cm^3 was taken from every sample to grind powder with the ball mill. Afterwards the powder was sieved through a 125 µm sieve. The powder was used for the XRD analysis as well as for the dissolution experiments.

3. DISSOLUTION EXPERIMENTS

Dissolution experiments at different temperatures, pressures and with different fluids are in progress to get more insights in reaction kinetics and processes within the geothermal reservoir. This is important to see the greatest effect between non-altered and hydrothermal altered specimens in the CT-Scans by least time as possible. A lot of literature e.g. (Dove, (1999)),(Gong et al., (2012)),(Schlabach, (2000)),(Yadav et al., (2000)) already exists on this topic concerning different questions.

A first series on time dependent dissolution experiments in small self-made capsules of approximately 1 ml was performed with sandstone powder. The capsules are consisting of an outer mantle made of stainless steel and an inner PTFE inlay. Sandstone powder of the Bentheim sandstone (GBS 4) was sieved through a 125 µm sieve. 100 mg of powder was placed into each capsule and filled up with 1 ml of distilled water. Then the capsules were closed tight and put in the dry oven. Samples were heated up to 150°C for the series. Fluids were analyzed after 6, 13 and 18 days. For comparison the same procedure was repeated with the GBS4 sandstone powder and 1 ml of fluid at room temperature ± 25°C. Determination of cation species within the produced fluids was done by measuring with an Optima 8300 ICP-OES of PerkinElmer. A single element standard was used for Si, Na and K, while the other elements were measured with a multielement standard. The expected error of measurement is around 1 ppm.

In general an increase of the dissolved species with time can be measured, like to be expected. In the diagram (Figure 4) it is clearly visible that saturation is reached after 13 days and no further quartz and K-feldspar of the sandstone powder can be dissolved. The dissolved silicium of 233 mg/l is one magnitude greater than the other elements. This is in analogy to the mineral composition with abundant quartz and minor orthoclase. Values below one ppm have to be treated with caution because of the measurement error.

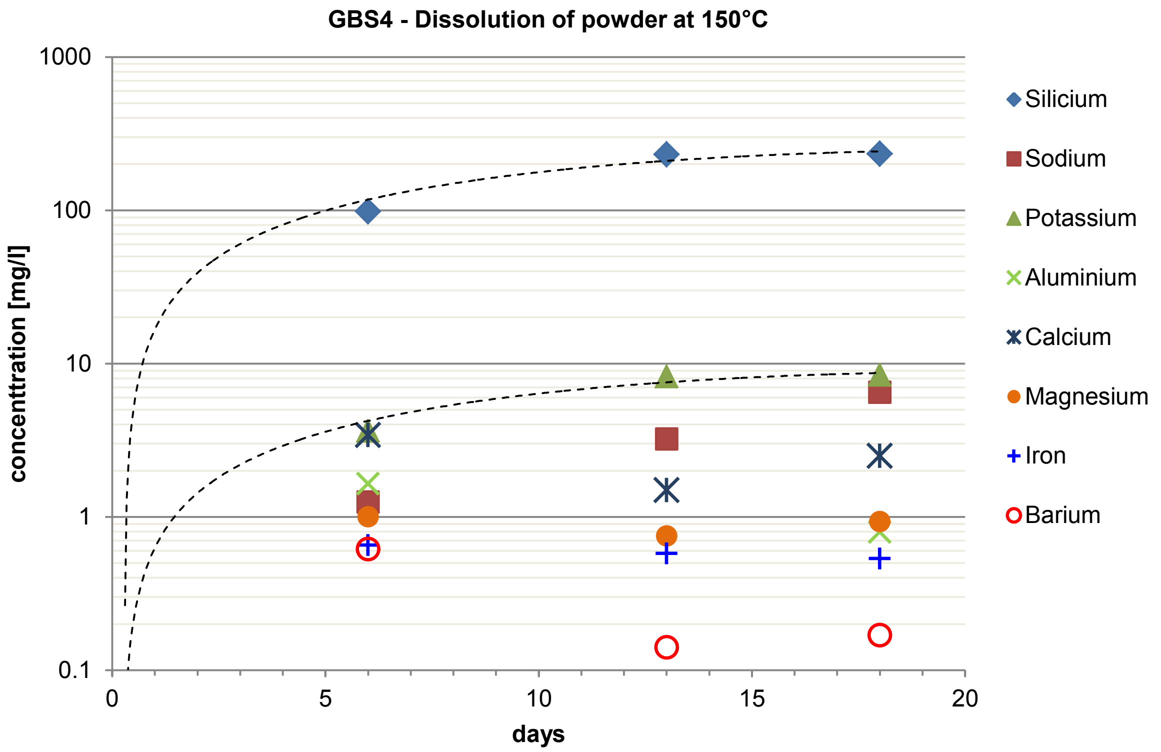

Figure 4: The diagram shows the results from dissolution experiments of GBS4 at 150°C. Time is on the x-axis and concentration in mg/l on the y-axis. Equilibrium of the aqueous solution is reached after 13 days.

A simple modelling was done by using the phreeqc software (Appelo, C. A. J and Postma, (2005); Parkhurst and Appelo, C. A. J, (1999)) with the phreeqc database for 150°C and 25°C. The results are in the same range as measured in the first experiments, but with less silica in solution (70 mg/l). Anyway the results seem to indicate a quartz precipitation rather than quartz dissolution.

A reactor of 1 l volume and up to 350 bars and 350°C can be used in the nearby future for long term experiments. These experiments will be performed at varying conditions including temperatures, pressures as well as different fluids such as distilled water or highly saline brines. Again the time dependent reaction kinetics will be of great interest. Therefore, it is possible to extract the fluids with a sample taking tube at a regularly base. Afterwards the sampled fluids can be analyzed. The properties of the prepared rock and cement samples will be measured before starting an experiment and afterwards.

4. CT SCANNING AT IN-SITU HP/HT CONDITIONS

All the previous experiments are necessary to define the conditions for the key objective of this research project, which is to investigate experimentally the physico-chemical interactions of fluids, rocks and the cementation of geothermal wells within a computed tomography scanner (CT scanner) under high-pressure, high temperature conditions.

4.1 Experimental Setup

A new prototype of a CT scanner by Procon X-ray is currently under development with multifocal x-ray tube of 225 kV for resolutions of around 1 µm (best case scenario). It will be possible to scan rock cores up to 600 mm in length and from less than 10 mm up to 100 mm in thickness by the helix CT scanning method. The computerized tomography is a non-destructive technique, which provides 3D reconstructed images of absorbing materials by reconstruction algorithms. Simulation software will enable the 3D visualization combined numerical simulation capabilities to compute physical properties of the analyzed materials such as absolute permeability and porosity, or thermal conductivity. The CT scanning unit will be extended with a setup for flow through experiments at high pressures up to 200 bar and temperatures up to more than 150°C to simulate reservoir conditions (Figure 5). Pressure gauges and mass flow meters allow the calculation of the effective permeability, which can be compared directly to the results calculated by the scanning software (Ott et al., (2012)).

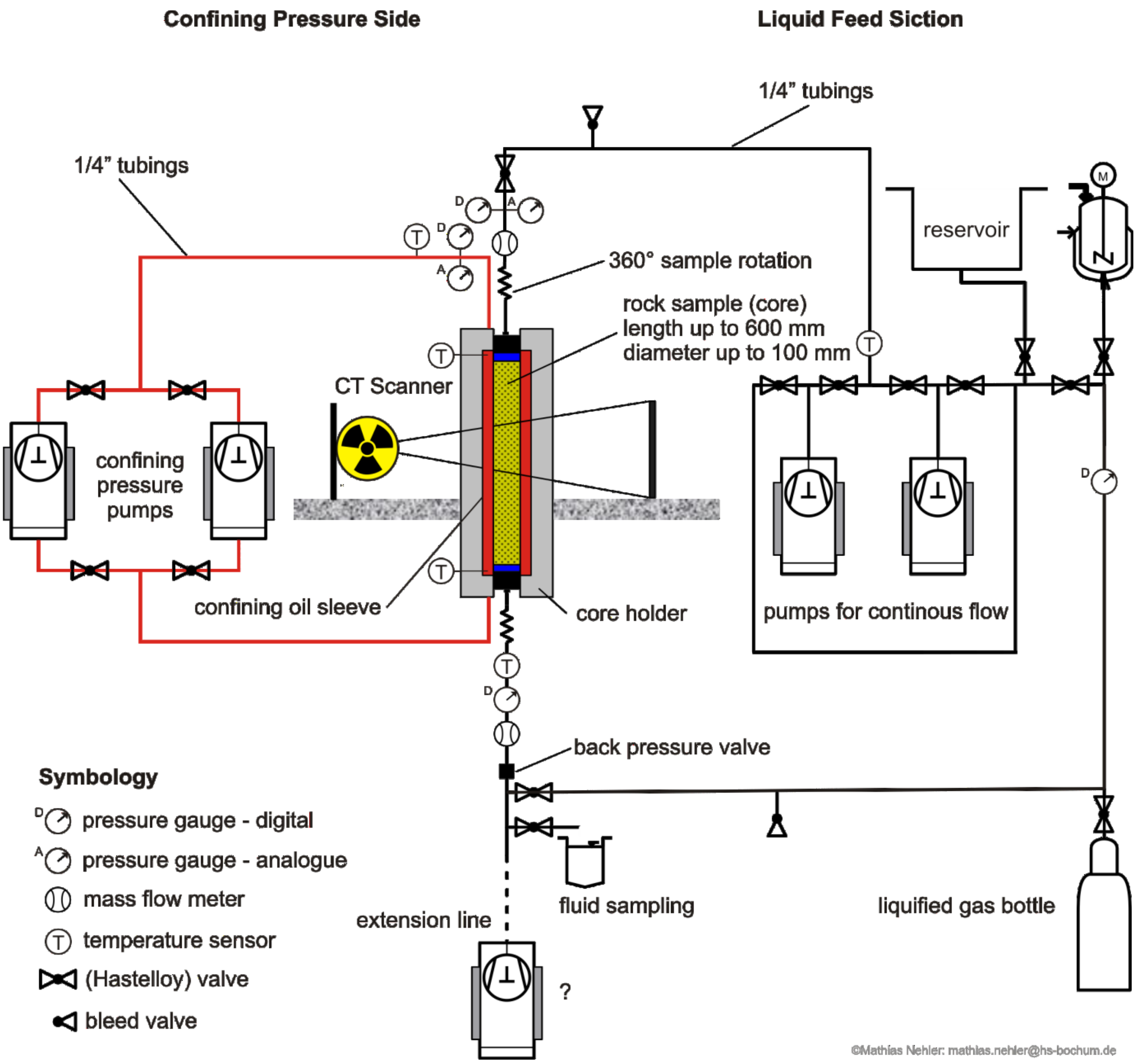

Figure 5: Experimental setup of the planned fluid-circulation unit, which is combined with a CT scanner.

A prototype of a core-holder consisting of layered aluminum and/or carbon fiber epoxy for optimal X-ray scans will be developed. In order to simulate the reactions between different miscible or immiscible fluids, the experiments can be conducted with gases such as CO_2 and liquids such as highly saline brines. The two-phase flow leads to changes in the effective permeability by mixing, as well as dissolution and precipitation processes. The couplings of chemistry and fluid flow in the porous rocks will be measured by spatial time resolved CT scans and time resolved sampling of the produced fluids.

These facts lead to some requirements of the intended materials. On the confining pressure side usual stainless steel can be used, whereas on the fluid circulation side highly corrosion resistant materials such as Hastelloy C-276 or Monel 400 will be used. To avoid mineral precipitation in the tubing's the temperature of the whole system will be kept constant as far as possible. That means the tubing's will be isolated. Besides heating is a matter of problem, because the sample cannot be heated directly at the core. This is due to the CT scanning unit. The X-rays must not be obstructed by obstacles such as ring heaters or heating jackets. The solution will be slowly circulating confining oil. So it will be possible to heat the oil in distant of the CT scanning unit.

4.2 CT scanning Results

CT-Scans of the Bentheim Sandstone with a diameter of 3 mm were already done. The measurements were done at a nanotem s by GE Healthcare with a resolution of 1.42 µm. The porosity was calculated by color thresholding with the software ImageJ (Lind, (2012)). The porosity was measured to 20.4 %. This is in accordance to porosities (20 – 25 %) calculated in earlier studies as already mentioned. In addition, the porosity was measured at a composite picture of the whole thin section of GBS4. The thin section was stained with a blue epoxy to identify porosity. The porosity was calculated to 22 % with this method. The porosity can be classified as interparticle pores.

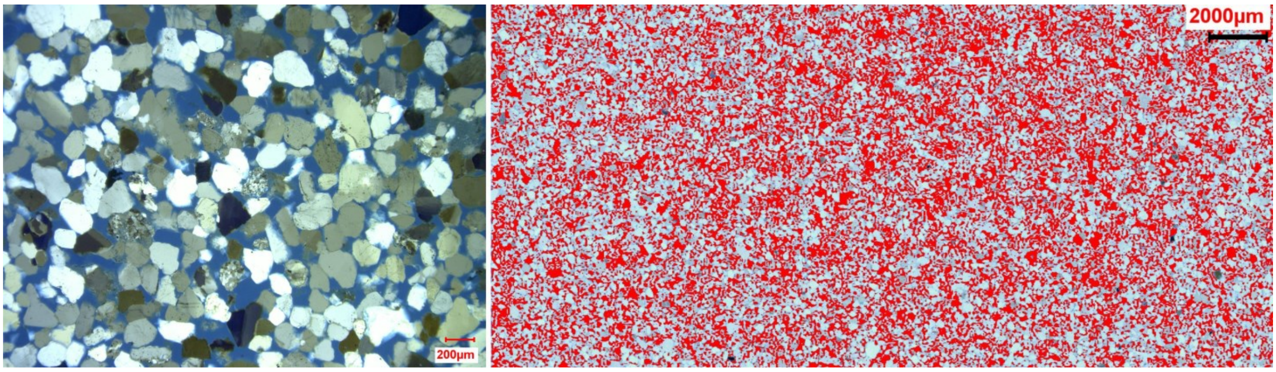

Figure 6: Thin section of the GBS4 sandstone. View of the blue stained porosity under cross polarized light (left) and calculated porosity (22 %) in red under normal polarized light (right).

The amount of quartz is around 77.1 %, while 2.5 are potassium feldspar. That means 96.8 % of the extracted picture is composed of quartz and 3.2 % are built up by other minerals. But a clear recognition and determination of the clay minerals, such as kaolinite in the GBS4, sample by CT scanning is impossible.

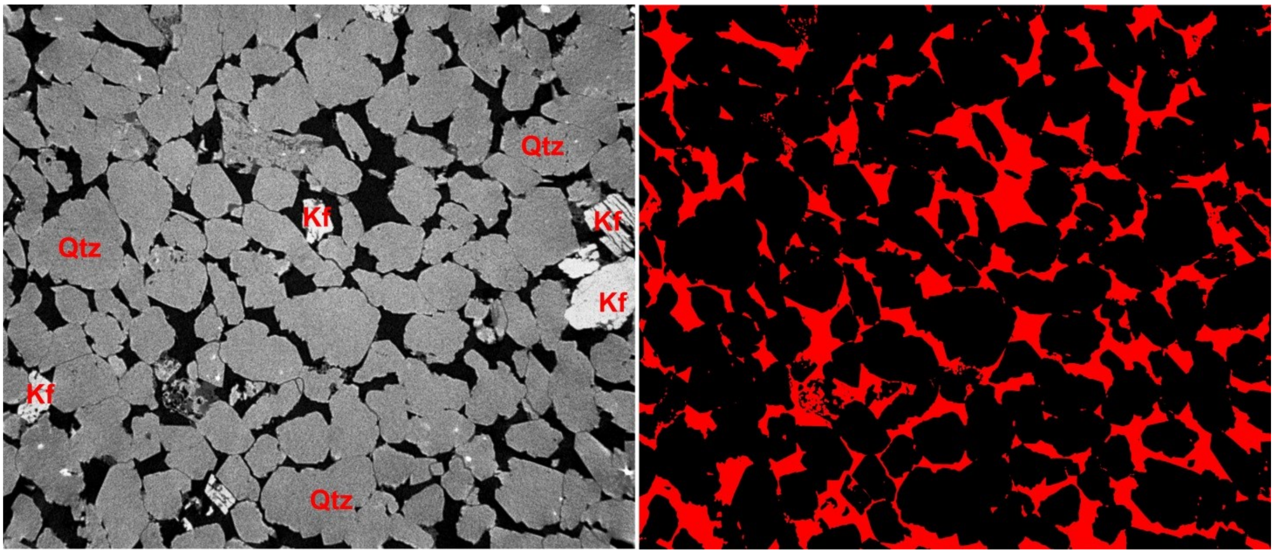

Figure 7: The figure shows a CT scan of the GBS1 (1.42 µm resolution). On the left side the minerals quartz (grey) and feldspar can be seen. The left side shows the image after thresholding for porosity (20.4 %) calculation by ImageJ software.

5. CONCLUSIONS

The first dissolution experiments indicate that equilibrium and total saturation at 150°C (reservoir conditions) is already reached after 13 days. This is due to the large surface of the sieved powder. This means following experiments at varying conditions with powder will be in the range of a maximum of 15 days. In contrast it will be expected that hydrothermal alteration experiments (precipitation and dissolution) of intact rock samples will take much longer until equilibrium is reached in these reactions. Anyway it should be possible to measure first effects in changing rock properties and fluid chemistry in reasonable time (weeks or month). Further experiments with different time lines and temperatures have to be conducted to verify the first results. Next experiments include different temperatures, pressure and saline waters e.g. sodium chloride water.

Also comparison of porosity calculations of CT scans, stained thin sections and literature values showed that the methods are in general applicable for quartz rich sandstones. Problematic will be the occurrence of clay, which is very difficult to determine by CT scanning due to the small grain size. Clays such as illite and smectite are the typical alteration products of the argillic alteration in the lower temperatures. At higher temperatures silification processes become more dominant, which will it make much easier for CT scanning.

The installation of the fluid circulation in combination with the CT scanner will still take some time and is matter of further discussions. Materials are very expensive due to corrosion resistance and comprises have to be done according to price and safety. Of biggest concerns are turning and moving parts, which needs to be sealed with O-rings. The pressure and temperature dependency of these parts together with the highly saline fluids places high demands. The valves of the pumping system are also limited to 250 bar and 160°C. Designing of the core holder is also a difficult task to improve CT imaging quality. Implementation of new measuring software for pressure transducers and thermo couples input signals is currently ongoing and first experiments will be conducted as soon as possible.

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