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Mechanisms of transformation of bentonite barriers – testing a new experimental concept

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1 Introduction

Bentonites are foreseen as a construction material for at least one of the engineered barriers buffer, backfill, sealing elements of boreholes, drifts and shafts in the currently developed or proposed geological repository concepts for high-level radioactive waste (HLW) and spent nuclear fuel (SF) worldwide. These barriers should either (i) retard the movement of external solutions into – and of internal pore solutions within – the repository area in general and most importantly in the near field of the emplaced HLW/SF, or (ii) exert support pressure on the surrounding rock masses or (iii) retard the migration of radionuclides from the repository area into the biosphere.

The clay mineral montmorillonite constituting the most of bentonites is able to absorb water in between its aluminosilicate layers, which can thereby be pushed up to four times farther apart as compared to the dehydrated state despite the external pressures of several megapascals at the repository relevant depths /MAD 89/. This swelling ability is responsible for the first two functions of the engineered barriers listed above, as the swelling montmorillonite particles, the most of which are characterized by the lateral dimensions below 2 µm, effectively (i) fill up the meso- and macropores in the bentonite material – reducing thus its permeability for solution flow – and (ii) exert a swelling pressure on the surrounding solid material upon the up to twofold increase of their thickness /LIK 06/. The knowledge of the swelling pressure and the permeability of bentonites is thus of high importance for their application in a repository system.

The primary aim of the present work was to prove whether a new experimental concept for measuring swelling pressures and permeabilities of compacted bentonites after reaction with concentrated solutions at high temperatures allows to overcome previous experimental difficulties and to provide reliable results. Besides, the quantification of the expenditure of time necessary to carry out such measurements was a further expected project outcome. This work was therefore a technical project aimed at a testing of a new experimental approach to be used in the subsequent main project (UMB, FKZ 02E11344A).

In previous experiments, a studied bentonite was loaded as a dried powder into the measurement cell, compacted to the density of 1.6 g/cm³, and saturated with a model solution /HER 11/. The swelling pressure developing in the cell as a result of the saturation and the permeability were measured online. The swelling pressure reaches its maximum value upon the accomplishment of the bentonite saturation and remains then unchanged for a certain period of time. As a result of chemical reactions between clay minerals in the bentonite and the solution within the measurement cell (Fig. 1.1), a transformation of clay minerals occurs, which on the timescale of months or years leads to declining swelling pressure and to a raising permeability. The rate of such clay mineral transformation depends on the structure and chemical composition of the minerals constituting the clay /HER 11/.

This experimental approach provided reproducible and as such reliable results for experiments at room temperatures. However, this was rather not the case for experiments at 60°C and 90°C /HER 11/. The reason for this is of a technical nature and is due to the design of the measurement cell (Fig. 1.1), which cannot prevent the evaporation of the solution from the cell at temperatures beyond the room temperature. Such evaporation will not occur at repository conditions in a bentonite barrier, which after its saturation with the claystone pore solution will represent an equilibrated and with respect to water loss intrinsically closed system. As a result of this evaporation in the experiment, on the contrary, salts are precipitating from the pore solution upon the increase of their concentration and exceeding the solubility limit. This precipitation leads to a filling out and sealing of the pore space. Accordingly, the measured changes in swelling pressure and permeability were no longer because of the mineralogical transformations of clay minerals, but mainly because of the unintended modifications of the pore space and the mineral surfaces.

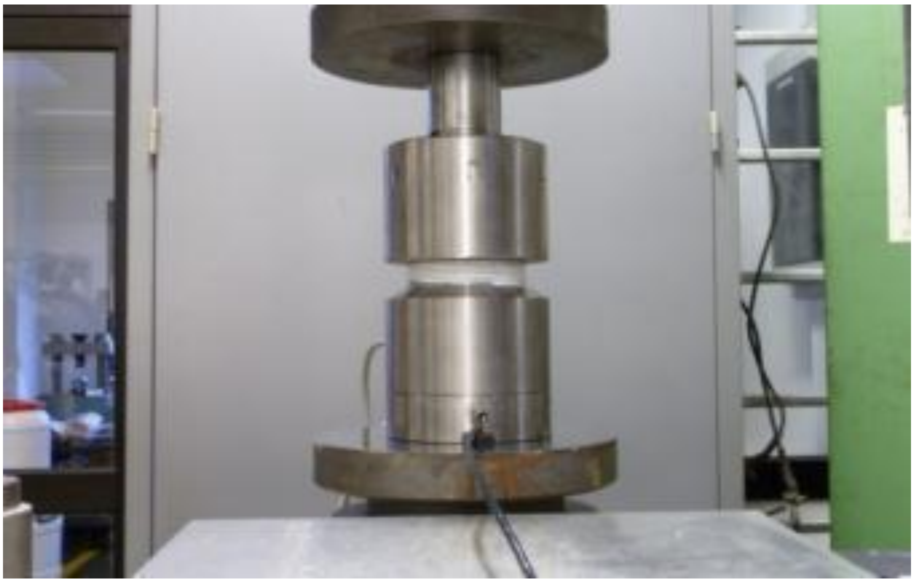

Fig. 1.1 A measurement cell for determination of swelling pressure and permeability of a compacted bentonites reacting with a model solution in the preceding project /HER 11/.

Thus, by virtue of the evaporation, no reliable measurement could be made at repository relevant temperatures beyond 25°C. As a result, considerable knowledge gaps remained with regard to critical issues of bentonite stability in the near field of the heat-generating radioactive waste.

Therefore, a new experimental concept must be developed, according to which bentonite powders are to react with solutions at the required high temperatures and the supernatant solution to be decanted after the specified reaction time in order to be able to compact the dried reacted bentonite powder to the target dry density of 1.6 g/cm^3 for measuring swelling pressure and permeability. For the purpose of comparison, swelling pressure and permeability are to be measured also for the unreacted bentonite powder compacted to the density of 1.6 g/cm^3. The comparison of the obtained results for reacted and unreacted bentonites would allow a quantification of chemical-mineralogical transformation in the bentonites due to the reaction with applied solutions at different temperatures in the main project. To facilitate this comparison, both reacted and unreacted bentonites are to be dialyzed in advance of the measurements.

The primary aim of the pre-project was accordingly to test the applicability of the new experimental concept, its ability to provide reproducible results, and the expenditure of time necessary for measurement of the swelling pressure and the permeability. The test was to be done for a comparatively short time period of several months, which was the time constraint for the carrying out the pre-project.

2 Materials and Methods

In the pre-project, Bavarian B11 bentonite (consisting to 89% of smectite /UFE 08/ with Na, Ca, and Mg constituting 55%, 30%, and 15% of smectite's interlayer cations, respectively /KAU 08/) was applied. The bentonite material was provided by the Federal Institute for Geosciences and Natural Resources. Prior to the start of the experiments, the bentonite B11 was dialyzed using a semi-permeable membrane with a regular change of water carried out during a period of several months until the residual conductivity of the water did not exceed 10 µS/cm (Step 1 in Fig. 2.1). Based on the experience of the mineralogical laboratory of the University of Greifswald, which participated in the previous experiments /HER 11/, attaining this conductivity was judged to facilitate carrying out reliable transmission electron microscopy analyses of the mineral phases of bentonites reacted with saturated salt solutions.

To prepare bentonite suspensions, the saturated NaCl-$CaSO_4$ solution (cap rock solution) characterized by a salt content of 325 g/L was chosen as a model solution to be expected at the interface between neighboring claystones and salt formations in the North Germany (Step 2 in Fig. 2.1). The bentonite suspensions were prepared at a solid/liquid ratio of 1:4 and reacted for seven months in gas-tight vessels at temperatures of 60°C and 90°C (Step 3 in Fig. 2.1, Fig. 2.2). The reacted bentonite powder was dialyzed as described above (Step 4 in Fig. 2.1), loaded into the measurement cell, and compacted to the density of 1.6 g/cm^3 (Fig. 2.3) for measurements of swelling pressure and, subsequently, permeability (Step 5 in Fig. 2.1). For the measurement of swelling pressure, the compacted bentonite was saturated in the measurement cell with the cap rock solution until the constant swelling pressure was reached (Fig. 2.4). Afterwards, the cells were put under the stepwise increasing pressure load, and the weights of solution entering and exiting the cell were measured online. Reaching the constant solution throughput at the maximum pressure load of 10 MPa on the entering solution, for which according to the laboratory experience no channel formation occurs in the compacted bentonite pellets, marked the accomplishment of the permeability measurement (Fig. 2.5).

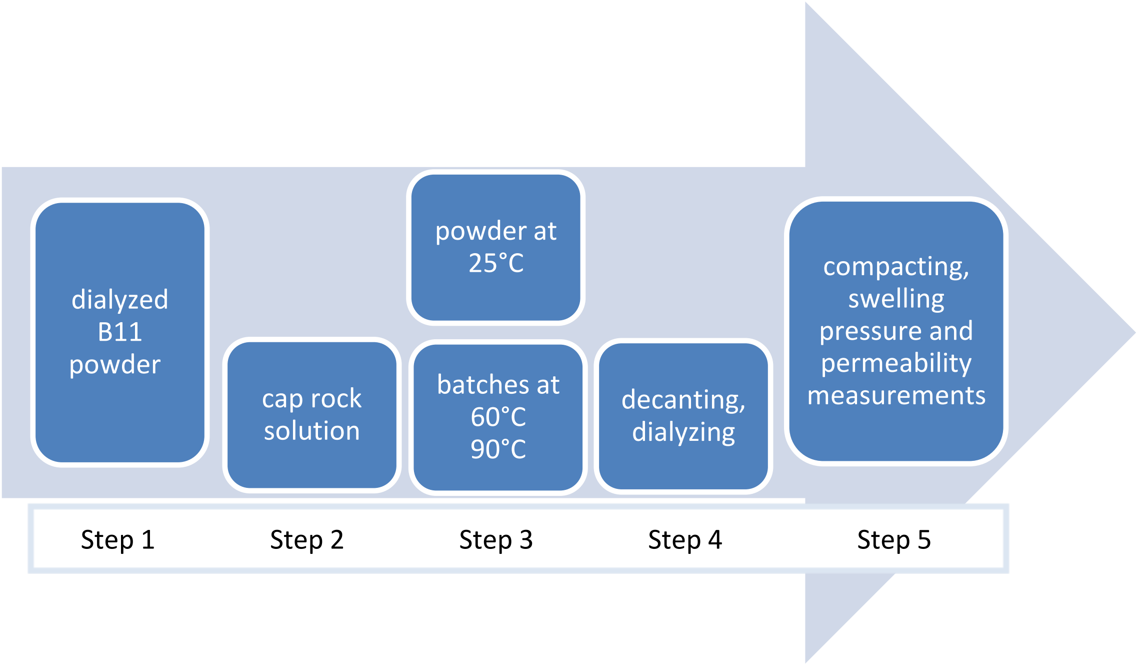

Fig. 2.1 Schematic diagram of the proposed experimental concept.

Fig. 2.2 Bentonite B11 powder after 7 months of reaction with a saturated NaCl-$CaSO_4$ cap rock solution at a temperature of 90°C.

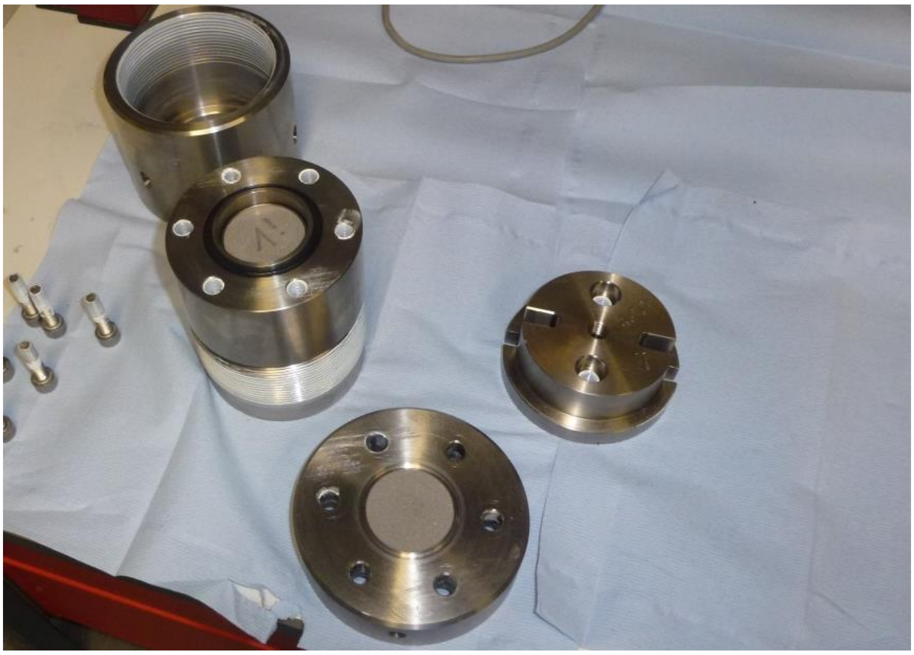

Fig. 2.3 Measurement cell with compacted bentonite B11.

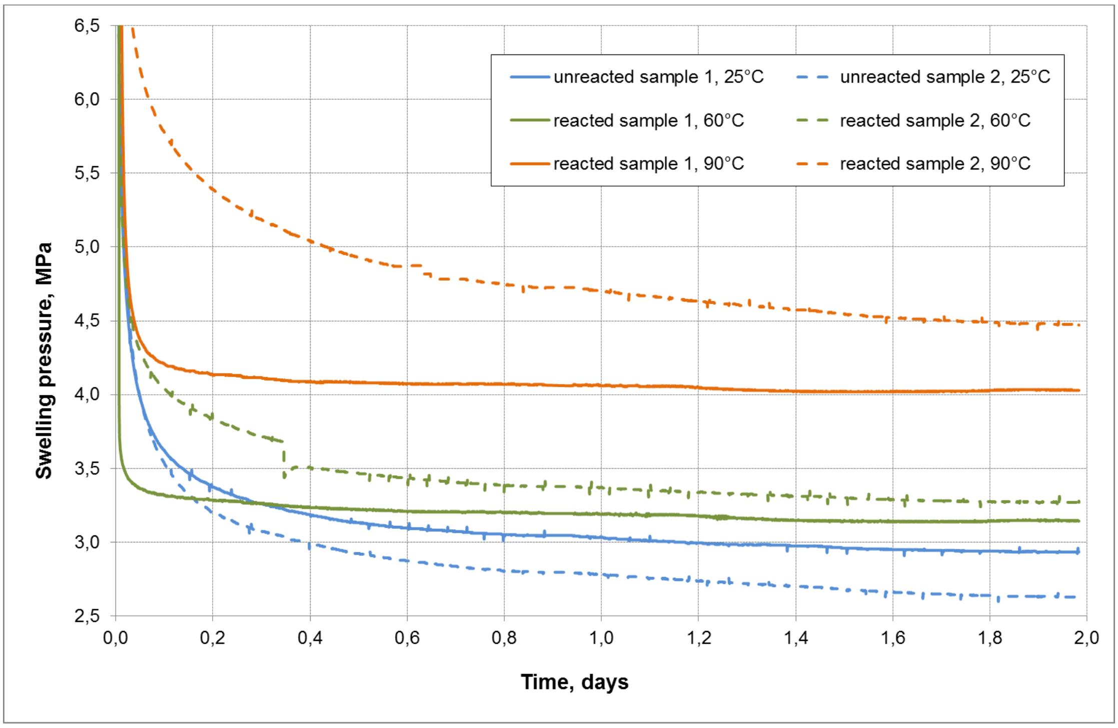

Fig. 2.4 Development of the swelling pressure with the measurement time for compacted bentonite B11 before and after reactions at 60°C and 90°C.

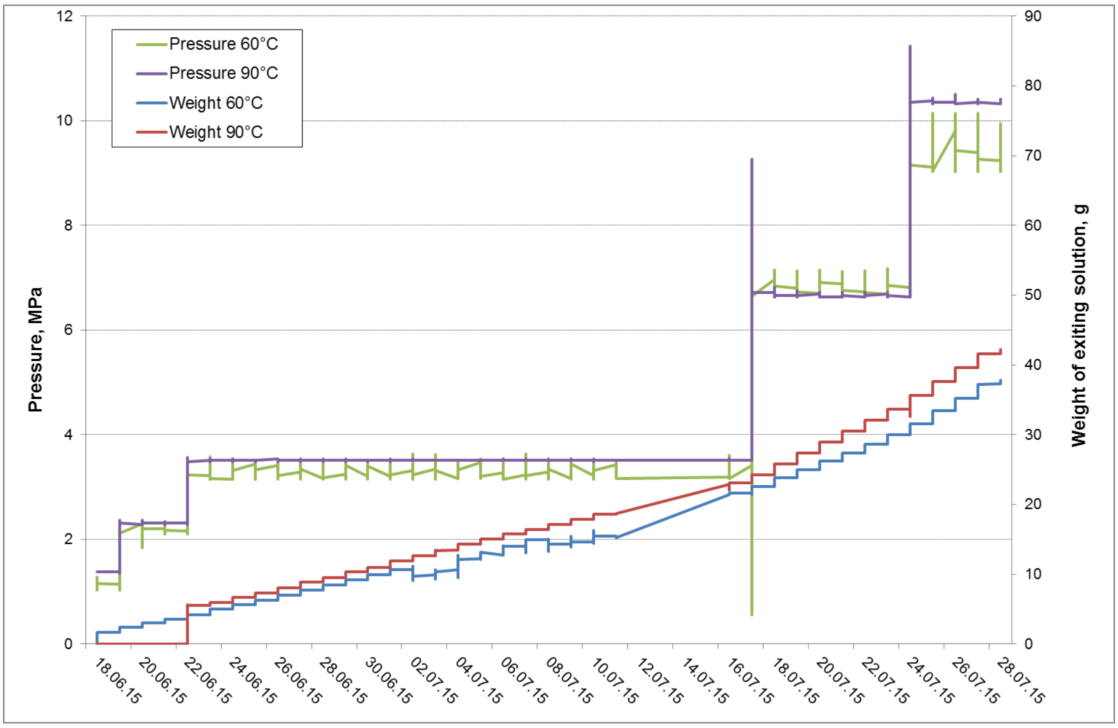

Fig. 2.5 Pressure load (MPa) and the weight of the exiting solution during the permeability measurements with compacted bentonite B11 after reactions at 60°C and 90°C.

3 Results and Discussion

The progress of the batch experiments confirmed the robustness of the chosen approach with respect to the application of high temperatures since no evaporation loss occurred within the experiment duration.

In total, six measurements of the swelling pressure and of the permeability were accomplished with the cap rock solution – two samples of unreacted bentonite powders and two samples of the each bentonite reacted at 60°C and at 90°C. The results of the measurements are summarized in Tab. 3.1 and Fig. 3.1.

Tab. 3.1 Swelling pressure and permeability for unreacted compacted bentonite B11 powders (25°C) and after reactions at 60°C and 90°C.

	25°C	60°C	90°C
Swelling pressure, MPa	2.78 ± 0.15	3.12 ± 0.16	4.22 ± 0.26
Permeability, × 10^{-18} m²	5.65 ± 0.22	5.71 ± 0.85	5.55 ± 0.04

It can be seen from this data that even after a relatively short reaction time of 7 months measurable differences in swelling pressure can be determined as a result of bentonite transformation at increased temperatures. Indeed, upon an increase of the temperature from 25°C to 60°C or 90°C, the swelling pressure shows significant increases by 0.34 or 1.44 MPa, respectively. These increases of swelling pressure with temperature are in agreement with the similar observation made in the preceding project for MX-80 bentonite in contact with a saturated NaCl solution characterized by a salt content of 350 g/L /HER 11/.

Differently than the swelling pressure, however, the permeability shows no significant changes with increasing temperature and equals to about 5.6 × 10^{-18} m² for the unreacted and the reacted bentonites (Tab. 3.1).

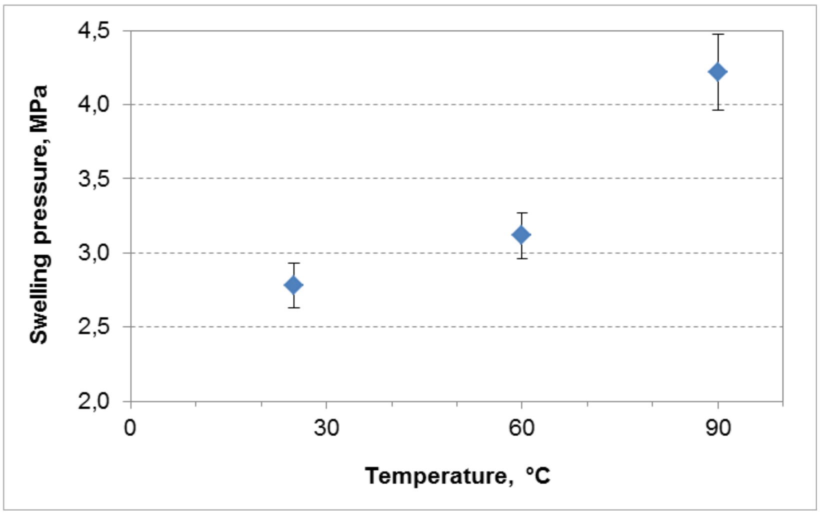

Fig. 3.1 Swelling pressure for unreacted compacted bentonite B11 powders (25°C) and after reactions at 60°C and 90°C.

Thus, the proposed experimental concept allows a reliable determination of the changes in the swelling pressure and the permeability of bentonite as a result of its interaction with cap rock solution at elevated temperatures for seven months. Therefore, the primary aim of this project can be considered to be successfully fulfilled. This experimental concept will be applied in the main project, in which the interaction between the bentonite and the diluted cap rock solution should last one and two years. This time should be sufficient for bentonite transformation to derive measurable changes in the targeted bentonite properties according to the results of the accomplished project. Furthermore, the expenditure of time for the carrying out the measurements of the swelling pressure and the permeability was quantified to be each about two weeks. This information should help in planning the workflow of the main project.

The structural reasons for the observed changes in the swelling pressure of the bentonite B11 with the treatment temperature and for the constancy of its permeability could not be derived in the current project due to the lack of mineralogical and structural analyses. It is the aim of the subsequent main project to combine the measurements of the swelling pressure and the permeability with the mineralogical and structural analyses at the mineralogical laboratory of the University of Greifswald.

4 Acknowledgement

Dr. S. Kaufhold (the Federal Institute for Geosciences and Natural Resources, Hannover) is gratefully thanked for providing bentonite B11.

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