Physical interaction model and EPSP materials characterisation

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Summary
As the EPSP ("Experimental Pressure and Sealing Plug") experiment will not been dismantled during the course of the DOPAS project ("Full-Scale Demonstration Of Plugs And Seals"), the construction of the laboratory scale model was proposed to simulate and describe saturation and interaction processes which can occur in the EPSP plug. Physical Interaction Model (PIM) was focused on changes in physical and chemical properties during the interaction of bentonite, concrete and synthetic granitic water, the identical materials as those used in EPSP construction. Studies and analyses were mainly focused on characterisation and evaluation of concrete leachate pH, material saturation, porosity determination, mineralogy, bentonite cation exchange capacity (CEC), bentonite specific surface area (SSA), water chemical composition after the interaction with concrete and bentonite. The results show that interaction processes occur to some extent, on the other hand duration of the interaction experiment was not sufficient to observe and confirm anticipated changes (e.g. in mineralogy, physical properties etc.). Details of laboratory testing are reported in Deliverable D3.21 (Vašíček et al. 2016).

1. Introduction background

The Czech deep geological repository concept assumes that waste packages containing spent nuclear fuel assemblies will be enclosed in steel-based canisters placed in vertical or horizontal boreholes at a depth of ~ 500m below the surface. Several types of sealing plugs will be required in the repository. The function of which will be to provide for the sealing and closure of individual waste packages not only throughout the period of repository operation, but also following the permanent closure of the facility. Such plugs will have to provide a high level of resistance to the considerable pressure which will be exerted by hydrostatic forces and volumetric changes within the engineered barriers (Dvořáková et al., 2014).
The Czech contribution to the DOPAS project (Demonstration of Plugs and Seals) was focused namely on the EPSP experiment, being built in Underground Laboratory Josef (Svoboda et al., 2016). The objectives of the EPSP experiment were following: to develop, monitor and verify the functionality of constructed experimental plug to determine and describe in detail the materials which are used for experimental plug construction. Shotcrete with low-pH leachate and bentonite pellets were selected for EPSP construction. The local Czech materials were used, including local bentonite B75_2013 (based on bentonite Rokle) and low-pH concrete based on local recipe. According to the plan, all materials used for plug construction have to be characterised prior, during and after the experiments. However, the EPSP experiment will not be dismantled during the course of the DOPAS project, the construction of the laboratory scale models was proposed.

2. Scope and objectives

Laboratory test were focused mainly on two materials, low-pH concrete and bentonite. The main goal was to determine processes ongoing on the bentonite-concrete interface.
Studies and analyses were mainly focused on characterisation and evaluation their physical and chemical properties and also on the interaction with synthetic granitic water (SGW). Concerning concrete, following tests were performed: leachate pH measurement, porosity determination, hydraulic conductivity measurement. Concerning bentonite material, following tests were performed: porosity determination, water content determination, hydraulic conductivity measurement, cation exchange capacity (CEC) determination, specific surface area (SSA) determination and mineral composition analyses. Concerning water phase, the composition and physical and chemical properties were tested.

3. Materials and laboratory tests

Physical interaction model (PIM)

Physical interaction model (PIM) was designed and started in 2014. The schematic geometry of PIM is shown in Figure . The identical materials as those in the EPSP experimental plug were used. Synthetic granitic water (SGW; Havlová et al., 2010) was used as a liquid phase for saturation and interaction processes in the interaction model. PIM consisted of three stainless steel cylinders with a diameter of 8cm and a length of 5cm each. The total length of the model is 15 cm. Such an arrangement reflexes the real in-situ EPSP plug geometry in the best way. Into the middle part of model, bentonite powder was pressed to reach the dry density 1400 kg/m^3. The central bentonite part was surrounded by two blocks of low-pH concrete. One side of PIM was connected to the source of synthetic granitic water, infiltrating under pressure 2MPa. By the end of 2015 year, the PIM was dismantled and material analyses (chemical composition, mineralogy, physical properties, etc.) were performed in order to compare the physical and chemical properties of the materials prior to and following to the termination of the experiment.

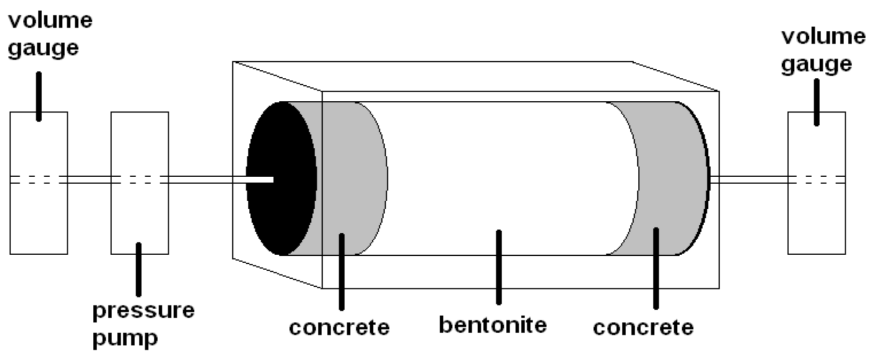

Figure 3-1: The geometry of the physical interaction model (PIM)

Concrete

Low pH concrete was developed under cooperation of ÚJV and subcontractor, however, precise recipe is the internal know-how of the subcontractor. One of required limits was to reach the pH of leachate < 11.7, in optimal case ≤ 11.5, pH measurements methodology followed the one described in SKB report R-12-02 (Alonso et al., 2012). Porosity measurements on both concrete samples from dismantled PIM and on the unaffected concrete samples were performed by water submersion method (Melnyk and Skeet, 1986).

Bentonite

A Czech Ca-Mg industrially milled and sifted non-activated bentonite, laboratory labelled as B75, produced by Keramost, Plc., was used in purposes of DOPAS project (B75_2013). The laboratory tests were performed with two forms of bentonite material (powder, pellets) in order to verify their properties. Prior to the experiments, bentonite material was compacted up to 1400kg/m^3 unless otherwise stated. This compaction value represents the (lower) dry density limit of the material which it is supposed to be able to be achieved by both spray technology and by the compaction of the bentonite pellets and powder for the in situ plug construction. Hydraulic conductivity tests were conducted in two types of experimental cells: small cell (diameter 3cm, length 1.5cm) and large cell (diameter 8cm, length 5cm). After the sample saturation or after performed conductivity tests, the water content and porosity were evaluated.

After PIM dismantling bentonite was sampled from points where some changes were visible (especially if the color (gray and white) spots appeared) compared to solid mass of bentonite block. Mineralogical XRD (X-ray diffraction) analysis on these samples was performed. During the dismantling of the model, the bentonite block was divided into 6 different slices. Each slice was then divided into five parts for the water content analysis. Furthermore, samples of bentonite were taken also from selected slices and those were analyzed for their cation exchange capacity (CEC), exchangeable cations and for their specific surface area (SSA) using EGME method (Carter et al., 1986).

4. Results and discussion

Used concrete material fulfils all required parameters, namely the limit for the leachate pH, having been determined as varying between 11.4 and 11.5 for both concrete parts of EPSP. Leachate pH value was not affected by interaction with bentonite and SGW in laboratory PIM model. In all cases the porosity of concrete evaluated by water submersion method ranged around 20-25% value. The permeability value in the concrete reached $7.9 \cdot 10^{-11}$m/s value with an uncertainty range of 2.7%. In addition, a material dry density value of 2046kg/m^3 was determined for the tested sample.

The processing technology has been identified as the main factor affecting the properties of B75 produced in recent years. Furthermore, it was found that partial activation and/or contamination caused by the presence of an activation reagent affect the composition of the water suspension or water leachate, cation exchange capacity and bentonite pore water composition.

Concerning property determination, the quantity of water flow through the sample was recorded throughout the duration of the experiment (**Figure 4-1**) and coefficients of hydraulic conductivity were calculated from the linear parts of the curves. The values of hydraulic coefficients varies in range $3.1\text{-}5.7 \cdot 10^{-13}$m/s.

For PIM saturation the SGW was taken from the reservoir and pumped to the PIM under the constant pressure 2 MPa using gas-hydraulic pump (as shown in Figure 3-1). First sample collected at the output of PIM was taken longer than 4 months after the start of the PIM experiment. Until this time only saturation of model components was observed. The outflowing PIM water was significantly enriched in all analyzed ions with exception of nitrates and fluorides (see Table 4-1) in comparison with original SGW. Dismantling of PIM it was found that water content in bentonite part slightly decrease (from 40.4 to 35.9 wt%) in the direction of water flow. Bentonite material does not reach the full saturation along the whole mass of bentonite sealing part. Example of bentonite slice distribution from the PIM dismantling, altogether with detemined water content is shown in Figure 4-3. As EGME method provides the value of total specific surface area for expanding clay minerals (sum of external and internal surface), it can point to mineralogical changes in these minerals caused by the alteration processes that results in changes of expandability. As can be seen from

Table Table 4-2**Fel! Hittar inte referenskälla.**, although some minor difference in specific surface area among PIM bentonite samples, generally their values of SSA (specific surface area) are close to the value of the original bentonite B75 pellet. The same situation is observed in case of CEC (cation exchange capacity) of all selected samples. It is clear that the property practical unchanged in comparison with B75 pellet sample, with respect to the analytical determination uncertainties (Table 4-3).

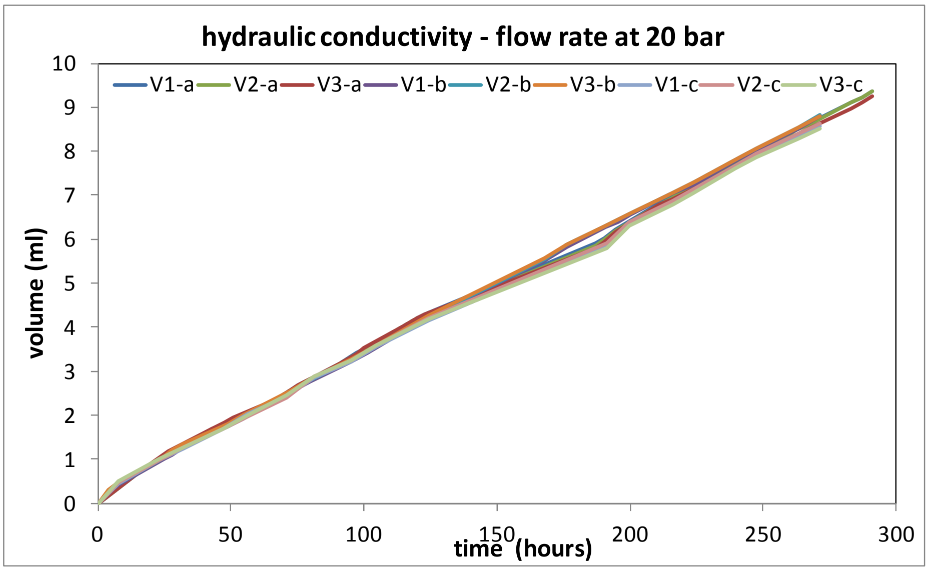

Figure 4-1: Hydraulic conductivity tests on B75 pellets in large cells, the water flow rates

Table 4-1: Comparison of SGW to PIM water composition (N.A. means not analyzed)

	SGW (mg/l)	**PIM water (1st sample) (mg/l)**
Na	10.6	1649
K	1.8	268
Mg	6.4	151
Ca	27.0	1695
Cl	42.4	426
SO_4	27.7	1845
NO_3	6.3	0.1
F	0.2	< 0.1
HCO_3	30.4	N.A.

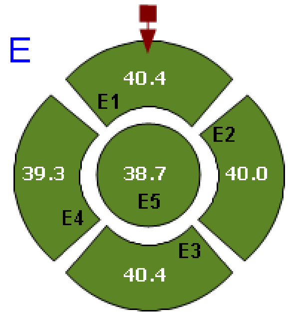

Figure 4-3: Example of a bentonite slice divided to parts for water content determination, with determined water content values (wt%)

Table 4-2: Specific surface of selected PIM bentonite samples

Sample	specific surface area (m^2/g)
2mm	430 ± 28
5A	491 ± 17
5C	476 ± 18
1E	482 ± 36
2E	466 ± 7
3E	465 ± 7
4E	478 ± 11
5E	491 ± 8
B75 pellet	453 ± 18

Table 4-3: CEC of selected PIM bentonite samples and B75 pellet

Sample	CEC (meq/100g)
2mm	55.1
5A	56.6
5C	57.3
1E	57.0
2E	54.9
3E	55.6
4E	56.9
5E	58.0
B75 pellet	56.4

Details of laboratory testing, results and discussion are reported in Deliverable D3.21 (Vašíček et al. 2016).

5. Conclusions

LPC concrete mixture was developed an applied in EPSP construction. The tests confirmed that this material fulfil all requirements stated at the beginning of the project. No changes in

the concrete leachate pH, porosity and mineralogy prior and after the interaction with bentonite and synthetic granitic water in physical interaction model were observed.
The properties of bentonite materials were studied in order to confirm the selection of the most suitable material. The analysis of chemical composition and mineralogy, the measurement of pH in bentonite suspensions and distilled water with different ratios, the analysis of leachates and the measurement of porosity were conducted with regard to B75_2013 bentonite which was selected as the construction material for the EPSP. Further, it was confirmed that the main properties of B75-2013 bentonite fulfil all the expectations, limits and requirements for the EPSP sealing part. It was observed no significant changes in mineralogical composition or presence of newly formed phases after the dismantling of physical interaction model. It is evident, that interaction processes between concrete and bentonite in performed physical interaction model did not reach to significant rate that can influence their properties, contributing to the material safety properties.

6. Acknowledgement

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7. References

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