Self-Sealing of Fractures in Argillaceous Host Rocks for the Disposal of Radioactive Waste

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Abstract:

The self-sealing behaviour of argillaceous host rocks for radioactive waste repositories has been comprehensively investigated at the GRS laboratory. Various sealing experiments were performed on cracked samples of different sizes from the Callovo-Oxfordian argillite and the Opalinus clay under relevant repository conditions. The fractured samples were compacted and flowed through with gas or synthetic porewater under confining stresses up to 15 MPa and elevated temperatures from 20 °C to 90 °C. Fracture sealing was quantified by measurements of deformation, permeability, and qualitatively by elastic wave velocity measurements. Under the applied thermo-hydro-mechanical conditions, significant fracture closure and permeability decrease to very low levels of 10^{-19} to 10^{-21} m² were observed within time periods of months to years. All test results provide strong evidence for the high self-sealing capacity of the studied claystones.

1 INTRODUCTION

Excavation of an underground repository for the disposal of radioactive waste in an argillaceous formation generates fractures around the openings and may induce pathways for fluid transport and radionulides migration. Because of the favourable properties of claystone such as the rheological deformability and swelling capability, a recovery process of the excavation damaged zone (EDZ) can be expected due to the combined impact of rock compression, backfill resistance, and clay swelling during the post-closure phase. For the assessment of the long-term safety of a repository, various kinds of evidence as for instance adequate quantitative characterisation of the host rock for a reliable prediction of the selfsealing process of the EDZ are indispensable. Within the last decade, the self-sealing behaviour of fractures in damaged claystone has been comprehensively investigated by GRS on the Callovo-Oxfordian argillite (COX) at the Bure Underground Research Laboratory (URL) in France and the Opalinus clay (OPA) at the Mont Terri URL in Switzerland under relevant repository conditions [1-5]. In various kinds of laboratory experiments, stronglyfractured samples were compacted and flowed through with gas or synthetic porewater under triaxial stresses and elevated temperatures. Fracture closure and permeability changes were determined over long testing durations of months to years. The most important results are presented in this paper.

2 SEALING OF FRACTURES UNDER COMPRESSION

The damaged zone will be gradually compressed by the progressive rock deformation and the increasing resistance of the backfill during the repository post-closure phase. The response of the damaged claystone to the mechanical compression was investigated on pre-fractured samples under various load conditions.

Fig. 1 shows pictures of strongly-fractured COX and OPA samples and the schematic of the triaxial compression tests. The samples were isolated in jackets and compacted by increasing axial and radial stresses. Closure of fractures parallel to the sample axis was



measured by a circumferential extensioneter mounted at the sample mid-height. Additionally, fracture closure was also detected by monitoring changes in elastic wave velocity using an ultrasonic device. Changes in permeability to gas along the fractures were measured by injecting nitrogen gas to the sample bottom and by recording the outflow at the opposite side.



Fig. 1 Pictures of fractured clay samples and principle of triaxial sealing test

The measured data of fracture closure (Δb) at some COX samples are presented in Fig. 2a as a function of the applied normal stress (σ_n). All the σ_n - Δb curves express the non-linear behaviour, involving the hysteresis cycle by loading/unloading and the permanent plastic deformation. Fracture closure evolves faster at large apertures in the initial loading stage and then closure rates decrease with increasing fracture stiffness at reduced apertures. The relationship of fracture closure with normal stress can be approached by an exponential equation [4-5]

$$\Delta b = b_m \left[1 - \exp\left(-\alpha \sigma_n^{\beta}\right) \right] \tag{1}$$

where Δb is the aperture closure, b_m is the possible maximum aperture closure (equal to the initial aperture), σ_n is the normal stress, α and β are constants. If the stress tends to infinity, $\sigma_n \rightarrow \infty$, the fractures will be fully closed, $\Delta b \rightarrow b_m$. Fitting the data derives a unique set of the parameters $\alpha = 0.3$ and $\beta = 0.5$ for the samples with different initial apertures of $b_m = 0.38 - 2.30$ mm. A reasonable agreement between the model and the data is achieved.

The closure of fractures is related to the intrinsic permeability in the fractured clay rock. The permeability variation with fracture aperture may be approached by the "cubic law" for a set of parallel fractures of equal aperture which are oriented parallel to flow direction. The permeability of a fractured rock is contributed by the fracture conductivity (K_f) and also increasingly with compaction by that of the porous matrix (K_m) [4-5]:

$$K_{g} = K_{m} + K_{f} = K_{m} + \frac{F}{12} (b - b_{c})^{3}$$
⁽²⁾

where b_c denotes a critical aperture, b is the average fracture aperture, and F represents an integrated character of the set of fractures. $K_m = 1 \cdot 10^{-21} m^2$ is determined for the intact rock. Fig. 2b compares the cubic model by application of various F-values with the measured permeability data as function of the fracture aperture decreased by loading. Whereas the parameter F is differing from one sample to another due to the different fracture features, a unique critical aperture $b_c = 2 \ \mu m$ determined is adequate for all the samples. As the geometric fracture aperture b decreases to a very low magnitude, $b \leq b_c$, the connection of

the fractures tends to be disconnected and the hydraulic conductivity in the dead-end fractures is ineffective.



Fig. 2a Fracture closure - normal stress relationship



Fig. 2b Fracture permeability - aperture relationship

3 SEALING OF FRACTURES UNDER WATER FLOW

As well known, the studied claystones have high water adsorption potentials and swelling capabilities [5-6]. They can take up great amounts of water much higher than the water content in the naturally saturated and loaded state. The increase in water content results in a large volume expansion of up to 10 % in unconstrained conditions. The wetting-induced swelling, weakening and slaking will contribute to the sealing of fractures. This issue was examined by flowing synthetic porewater through cracked samples under confining stresses.

Fig. 3a illustrates the combined impact of compression and water flow on the sealing of fractures in a COX sample. The recorded radial strain normal to the fracture planes shows that each load increase led to an immediate closure of the fractures. Because of the high swelling capability of the claystone, a gradual expansion of the fractured sample followed at each constant load below 10 MPa rather than a gradual closure. The externally observed expansion indicates high local swelling pressures acting in contacting areas between fracture walls, where the material must expand more into the fracture voids than elsewhere.



Additionally, the clay particles in the fracture walls contacting with water become weakening, slaking and filling the voids. The sealing of the fractures leads to a decrease in permeability. After stepwise increasing the load to 13 MPa over seven months, a very low permeability of $K_w = 2 \cdot 10^{-19} \text{ m}^2$ was reached, which is close to that of the intact clay rock. The permanent sealing of fractures by swelling and slaking of clay matrix is clearly visible on the pictures in Fig. 3b comparing the fractures on the sample surface before and after testing. The fractures with sharp wall edges disappeared due to the wetting-effects.



a. normal strain and permeability parallel to fractures during water flowing



after water flowing

b. fracture patterns before and after water flowing

Fig. 3 Sealing of fractures in claystone during water flowing under various confining stresses

After the disposal of HLW in repository cells, the damaged host rock will be heated up and dried. The clay minerals may change, affecting their swelling capacity and correspondingly the recovery of the heated EDZ. Another test aimed at examining the sealing capacity of preheated claystone. Three COX samples were artificially cracked along their length and then pre-heated up to temperatures of 50, 100 and 150 °C, respectively. The intensity of the



fractures was different from one sample to another, as shown in Fig. 4. They were compressed at relatively low confining stresses of 2 to 3.5 MPa and flowed with synthetic porewater through over more than 3 years. Before injecting the synthetic porewater, a high intrinsic permeability of $K_g = 3 \cdot 10^{-12}$ m² was determined on all samples by gas flowing. As soon as the water was supplied, the intrinsic permeability dropped immediately by five to seven orders of magnitude down to $K_w = 10^{-17} - 10^{-19}$ m², depending on the fracture intensity of each sample. As discussed above, this drastic drop in permeability is mainly attributed to the water-induced swelling, slaking, and clogging of the fractures. At each load level, the permeability variation was not significant in the testing range. Interesting is that the preheating up to 150 °C did not hinder the sealing process of the fractures in the claystone. Another important observation is that after reduction of the injection pressure from 1 to 0.1 MPa (corresponding to a pressure gradient of 200 m/m), no water outflow was detected over 8 months. This means that the Darcy's law might be not applicable for the highly re-sealed clay rock under certain low hydraulic gradients. The final permeability values determined after 3 years are very low at $3 \cdot 10^{-20} - 7 \cdot 10^{-21}$ m², being the same as that of the intact clay rock.



Fig. 4 Long-term evolution of the water permeability of fractured and pre-heated claystone under different confining stresses

4 THERMAL IMPACT ON SEALING OF FRACTURES

Heat released from high-level radioactive waste is dissipated through the buffer and the surrounding rock. The resulting temperature changes may affect the development of the EDZ. The thermal impact was investigated by heating cracked claystone during water flow under confining stresses.

Fig. 5 illustrates the evolution of water permeability measured on two cracked COX and OPA samples during heating and cooling between 20 °C and 90 °C over more than 3 years. The temperature-dependent viscosities of the synthetic water are taken for the calculation of the intrinsic permeabilities. Again, the water-induced swelling effect on the sealing of fractures is evident during the first stage at 20 °C. The permeability decreased from $1 \cdot 10^{-15}$ to $3 \cdot 10^{-18}$ m² at the OPA and from $1 \cdot 10^{-17}$ to $5 \cdot 10^{-19}$ m² at the COX sample. Subsequently, the rates of permeability reduction, however, are less affected by the temperature increase up to 60 °C. Further heating up to 90 °C and also cooling down to 60 °C had no or only little effect on the permeability reached before. Further cooling down to 20 °C, however, induced a reduction of the permeability to $3 \cdot 10^{-19}$ and $1 \cdot 10^{-19}$ m² at both the samples. Again, under the injection

pressure reduced to 0.1 MPa (corresponding to a pressure gradient of 200 m/m), no outflow of water could be detected within a time period of 8 months. Obviously, the water permeability values of 10^{-18} to 10^{-19} m² determined during the heating/cooling cycle are very close to that of the intact rock.



Fig. 5 Long-term measurements of water permeability on fractured COX and OPA claystones during heating and cooling

5 SIMULATIONS OF EDZ-DEVELOPMENT AROUND BORHOLES

In order to simulate fracturing and sealing processes in the surrounding host rock after the disposal of HLW in boreholes, a series of large-scale tests have been carried out on big hollow clay cylinders. They were prepared to an outer diameter of 280 mm and lengths between 460 and 530 mm with axially-drilled central boreholes of 100 mm diameter. Fig. 6 illustrates the test layout and shows a photo of a large hollow cylinder before testing.



Fig. 6 Principle of large hollow cylinder tests for investigation of fracturing and sealing of the damaged zone around boreholes under relevant repository conditions



The relevant processes during borehole excavation, backfilling, water transport, heating, and cooling were simulated by applying coupled THM-conditions to the large hollow samples. The main results are summarized as follows.

The <u>borehole excavation</u> was simulated by reducing the inner borehole pressure from a rock stress of 15 MPa down to 1 MPa. Consequently, no increase but a slight decrease in permeability was observed (see Fig. 7). This means that no interconnected pathways had been developed along the sample length of half a meter. This observation suggests that in the much longer boreholes and drifts of a repository, excavation-induced fractures in the EDZ may not be connected in a direction parallel to the boreholes and drifts at depths not deeper than 500 - 600 m. A drastic increase in gas permeability from 10^{-21} to 10^{-15} m² was observed only at high confining stresses beyond 20 to 24 MPa (corresponding to the depths of 800 – 1000 m).



Fig. 7 Fracturing-induced permeability changes in a large hollow cylinder of COX claystone

The <u>impact of backfill support</u> was simulated by increasing the borehole pressure from 1 MPa to the in situ prevailing rock stress of 15 MPa. Fig. 8 shows that the permeability was decreased by the increasing borehole pressure by several orders of magnitude from $10^{-14} - 10^{-18} \text{ m}^2$ to $10^{-18} - 10^{-21} \text{ m}^2$, depending on the initial fracture intensity in the EDZ around the boreholes.

The <u>water flow</u> in the EDZ was simulated by injecting synthetic porewater into the damaged cylinders. The intrinsic permeability determined by water flowing is about 10^{-18} m² which is three orders of magnitude lower than the permeability value of 10^{-15} m² obtained before by gas flowing. This reflects the effect of swelling and slaking of claystone on the sealing of fractures.

The <u>thermal load</u> was simulated by heating the damaged hollow cylinder from 29 °C to 74 °C and then cooling down to 29 °C again with each stage lasting about three weeks, respectively. The synthetic water was injected into the sample at a pressure of 0.3 MPa at a confining stress of 15 MPa and an inner borehole pressure of 1 MPa. Fig. 9 illustrates the recorded inflow and outflow of water. Before heating, as well as during and after heating, the variations in flow rate are insignificant. The water permeabilities which were determined before and after heating vary in a small range from $2.0 \cdot 10^{-18}$ m² to $3.5 \cdot 10^{-18}$ m².

All the observations on the large hollow cylinders during the EDZ-simulation tests, i.e., that the permeability decreases by compression and water flowing and that thermal effects are limited, are consistent with the results obtained on the normally-sized samples.

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Fig. 8 Compaction-induced permeability decrease in the EDZ around boreholes



Fig. 9 Water flow through the EDZ in a large hollow cylinder of COX claystone before, during, and after heating

6 CONCLUSIONS

From the various kinds of sealing experiments on strongly-cracked claystones, the following conclusions can be drawn:

- The fracture aperture decreases exponentially with increasing the normal confining stress. The resulting decrease in gas permeability is related to the aperture by a cubic law. Fracture closure and permeability decrease are also time dependent and accelerated by moisture increase in the environment.
- 2. Wetting by water flow through fractures induces swelling and slaking of the clay matrix, filling and clogging of the interstices, and a drastic decrease in permeability by three to seven orders of magnitude compared with the data obtained by gas flow. At hydraulic

gradients of smaller than 200 m/m, no outflow of water could be detected through the resealed claystone over 8 months. It seemed that Darcy's flow might not take place in the re-compacted clay rock.

- 3. Heating from 20 °C to 90 °C has no remarkable impact on the water permeability of fractures, while cooling down again decreases the permeability slightly. The thermally-induced changes in hydraulic conductivity are mainly attributed to the variations of water viscosity and density.
- 4. Under the applied THM conditions (confining stresses of 1 MPa to 13 MPa, temperatures of 20 °C to 90 °C, gas and water flow) to be expected in repositories, the permeability of the fractured claystones decreases significantly to very low levels of 10⁻¹⁹ to 10⁻²¹ m², which corresponds to intact clay rock, within very short periods of months to years compared with the long repository post-closure phase of tens of thousands of years.

All the laboratory observations suggest the high self-sealing potentials of the studied argillaceous rocks. Prediction and assessment of the long-term sealing performance of the damaged zone around the repository openings need adequate constitutive models and computing codes. Respective model development and modelling work will shortly be started at GRS in the frame of a new research project.

7 ACKNOWLEDGEMENTS

This work has been co-funded by the European Commission (EC) as part of the sixth Euratom research and training Framework Programme (FP6) on nuclear energy (2002-2006) under contract FP6-036449 and by the German Federal Ministry of Economics and Technology (BMWi) under contracts 02E10045 & 02E10377. The support from ANDRA by providing the samples is gratefully acknowledged.

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