Low Perm: Development of a calibration standard for evaluating permeability measurements of tight rocks

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Introduction

Permeability of tight rocks is an important subject in the site selection process for a deep geological repository for high-level radioactive waste. The permeability of the host rock needs to be very low, because the containment-providing rock zone shall isolate the waste and prevent or limit transport of radionuclides from the repository into the biosphere for 1 Ma.

However, permeability measurements of tight rock are a great challenge for rock physics laboratories. Data from different laboratories – applying various testing methods – are generally hard to compare. For a plausible decision-making in the site selection process the development of a trustworthy, comprehensive and reproducible permeability data base is a key aspect. In addition, methodology recommendations for permeability measurements of tight samples of the three host rocks (claystone, crystalline rock, rock salt) would be beneficial. In this regard it is desirable to:

1. develop a calibration standard which is not affected during testing by changes in confining or pore pressure, temperature, or the type of fluid used.
2. test this standard at various laboratories which apply comparable but variegated permeability testing methods and experimental setups.

The research project *Low Perm* is geared towards both these objectives and aims to improve the comparability of measured permeability values of tight rocks. In a first step (part 1 of the project) a calibration standard is being developed that is chemically inert, has high mechanical strength, high temperature stability and low thermal expansion. In a second step (part 2 of the project), specimens of the standard material will be sent to five laboratories for testing.

The current development status is described below.

Manufacturing of the calibration standard material – an Al_2O_3 ceramic

1. Molding

Fig. 1a: Plug after molding at 10 MPa.

Molding of the plugs is done with a hand-lever press at low pressure; air bump is at ~50 % of the peak pressure. Subsequently, the plugs are treated with CIP (cold isostatic pressing) at 100, 200 and 300 MPa, respectively.

2. Debinding

Fig. 1b: Sample after debinding. The plugs have a diameter of ca. 33 mm, a height of about 9 mm and weigh roughly 28 g.

Debinding is done as a four-stage process at a slow cooling rate and at peak temperatures between 300 and 600°C.

3. Sintering

Fig 1c: Specimen after sintering.

Five-stage sintering process for the 100, 200 and 300 MPa samples at five different final temperatures, namely 1300°C, 1350°C, 1400°C, 1500°C and 1600°C with varying heating and cooling rates.

Pore space within the 1400°C/300 MPa material investigated by BIB-SEM:

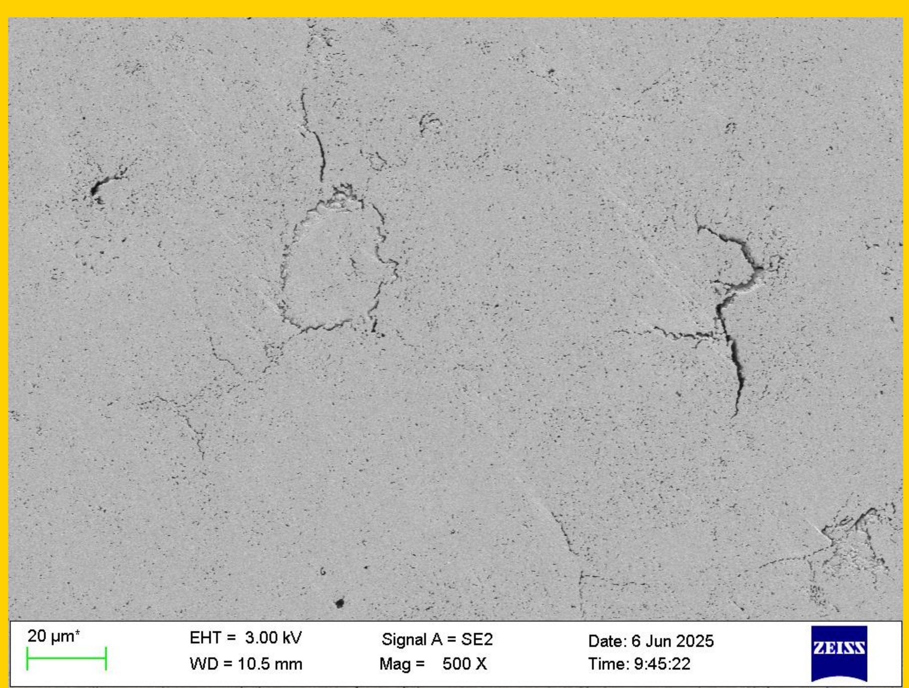

Fig. 2: Overview on the pore inventory of the ceramic with large pores resembling the former Al_2O_3 granules and very fine matrix pores.

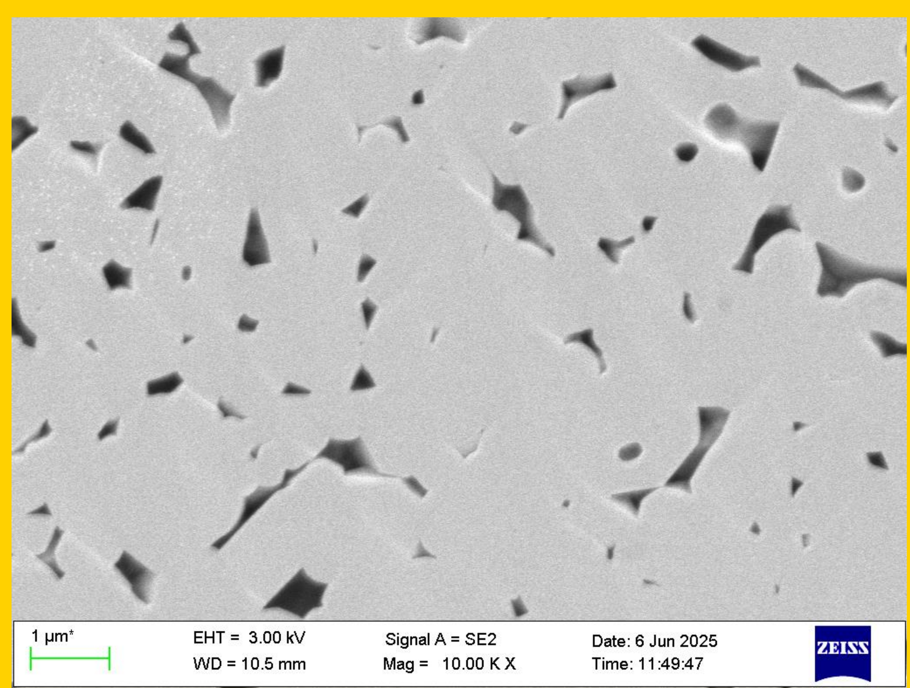

Fig. 3: Angular, euhedral matrix pores viewed at small scale.

BIB-SEM (broad ion beam – scanning electron microscopy) investigations were done with samples molded between 100 and 300 MPa and sintered at temperatures between 1300 and 1600°C. Here we focus on the corundum ceramic molded at 300 MPa and sintered at 1400°C, which will most probably be used for production of the calibration standard. Although the 1400°C/300 MPa specimen matrix is dense, remnants of the granular Al_2O_3 material can be found (Fig. 2). The dense ceramic shows a bimodal pore size distribution with very small matrix pores of roughly 1 µm length and large pores of up to 50 µm length. They seem to represent the margins of the former corundum granules. The smallest matrix pores have frequently sharp edges, resembling euhedral grain boundaries (Fig. 3). Interconnection of the matrix pores will be tested.

Radial diffusion data:

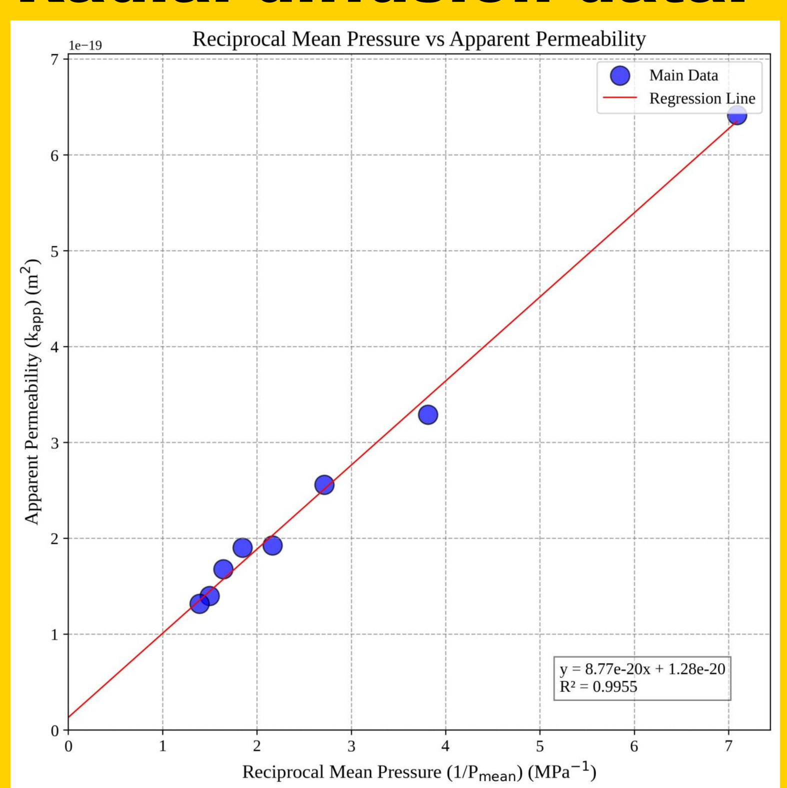

Fig. 6: Radial diffusion measurements of the 1400°C/300 MPa sample resulted in a porosity of 6 % and a permeability of 1.28 x 10^{-20} m².

Steady-state permeability measurments:

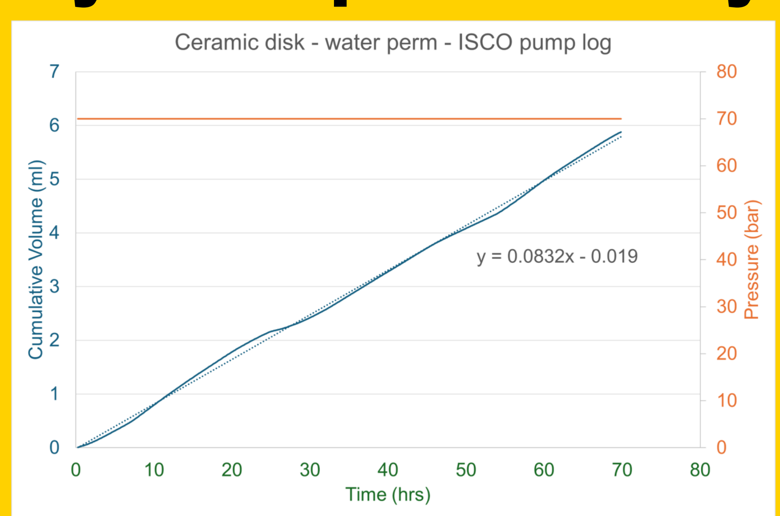

Fig. 7: Water permeability was measured under steady state conditions at a constant fluid pressure of 70 bars. The fluid flow rate was steady over three days at 0.08 ml/h.

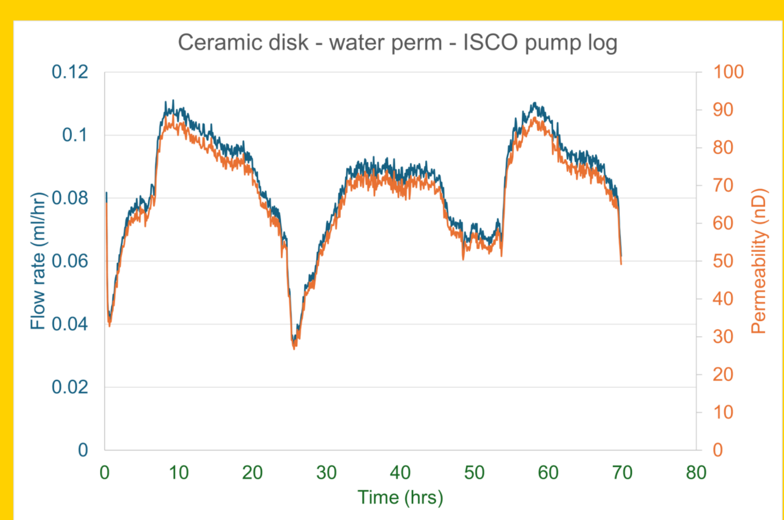

Fig. 8: The flow rate oscillates slightly between 0.06 and 0.11 ml/h leading to a permeability between 2.47 and 8.88 x 10^{-20} m² with a mean of 6.51 x 10^{-20} m².

Radial diffusion measurements were carried out at RWTH Aachen for the following specimen:
1300°C/300 MPa (k: 1.46 x 10^{-17} m²; ϕ: 0.25)
1350°C/300 MPa (k: 9.38 x 10^{-18} m²; ϕ: 0.38)
1400°C/300 MPa (k: 1.28 x 10^{-20} m²; ϕ: 0.06)

Here we focus on the specification which will result in the calibration standard material produced at a molding pressure of 300 MPa sintering temperature of 1400°C (see Fig. 6).

A 1400°C/300 MPa specimen was sent to Heriot Watt University (Edinburgh) for gas and water permeability measurements (steady-state). So far a first water permeability measurement (Figs. 7 & 8) confirms the radial diffusion data with a value of **6.51 x 10^{-20} m²**.

Results:

- Results of BIB-SEM and radial diffusion measurements are very promising
- BGR will perform additionally semiquantitative pore space analyses of the BIB-SEM images
- A first water permeability measurement confirms the radial diffusion data
- We expect further gas permeability (measured with N_2) and water permeability data
- We seem to have found the right P/T conditions (300 MPa / 1400°C) for manufacturing a ceramic with proper porosity (6 vol. %) and permeability (≤ 10-19 m²)
- The samples for part 2 of the project can most probably be produced

Acknowledgements:

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