Qualitative and quantitative characterization of porosity in geomaterials by combining broad ion (argon) beam and scanning electron microscopy (BIB-SEM)

Introduction

We present here the results of a combined semi-automated approach to map and characterize porosity in argillaceous materials (clays and shales) in terms of size and local distribution of pores, related to mineralogy as well as orientation of pores and minerals. We use a broad ion (argon) beam polisher combined with a conventional high resolution SEM to study pores down to the nanometer scale. Detailed information on pore morphologies and distribution are key points in understanding porosity evolution during burial and to be able to later model permeability. The characterization of pore morphologies at mm-scale is important to improve the understanding of diffusion controlled transport processes of radionuclides in argillaceous materials. Furthermore to understand sealing capacities, coupled flow, capillary processes and mechanical properties of a rock, knowledge on its porosity is indispensable.

Method

Cross-sections are made perpendicular to the bedding of the samples, using a JEOL-SM 09010 cross-section polisher (Figure 2). By this means the polished surface is enlarged by a factor of thousand, compared to a focused ion beam (FIB) approach. The resulting area is ~ 1 mm² large, with a flat, undamaged surface and a relief of less than 5 nm (Figure 3). These high quality cross-sections are predestined to be investigated with a state of the art scanning electron microscope (SEM, ZEIS - supra 55, Figure 1). Before imaging, samples are coated with a thin layer of gold to prevent them from charging. Identification and mapping of mineral phases is performed on a SEM-EDX detector, as well a segmentation of pores is performed on representative areas, stitched together from several hundreds of high resolution SEM-images. The creation of mosaics is necessary to get statistically significant results, but at the same time be able to investigate pores at the highest suitable magnification (~ 30kX).

Opalinus Clay

Opalinus clay has been investigated thoroughly over the last forty years at the Mont Terri site (Switzerland), to analyze its hydrogeological, geochemical and rock mechanical properties. The research has aimed to provide important input for assessing the feasibility and safety of deep geological disposal of radioactive waste in argillaceous formations. The research work has its focus on the determination of the in-situ porosity of the Opalinus clay by using BIB/FIB milling techniques in combination with SEM and cryo-SEM microscopy.

Organic rich shale

Two adjacent organic rich core samples from a shale interval in the Middle East were investigated. Based on more than 13,000 manually traced pores, porosity could be classified into two major pore-size classes. Relative large pores (> 0.5 μm², Figure 4), typically found in the organic matter-rich intervals, contributing strongly to the overall porosity of the shale. However, the majority of the pores found had an equivalent radius of less than 400 nm, with pore areas below 0.5 μm² (Figure 5), including these pores, porosities of both samples give similar results in the order of less than 1 % (Figure 7).

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Figure 1: BIB-cryo-SEM Zeiss supra 55 (@ RWTH Aachen University).

Figure 2: Picture of the JEOL BIB cross-section polisher with schematic drawing of polished cross-section of a sample.

Figure 3: AFM image of a scanned BIB-polished surface, with a relief of +/- 4.5 nm in this area (Aretz, GFE).

Figure 4: Typical pores in different mineral phases present in Opalinus clay, imaged at a magnification of 40kX.

Figure 5: BIB-SEM Zeiss supra 55 of Opalinus Clay at 10kX magnification.

Figure 6: Picture of the JEOL BIB cross-section polisher with schematic drawing of polished cross-section of a sample.

Figure 7: Fractal distribution of pores in matrix phase at all magnifications used in both samples.

Figure 8: Typical pores in different mineral phases present in Opalinus clay, imaged at a magnification of 400X.

Figure 9: Image of typical pores in Boom Clay at 6kX magnification.

Figure 10-12: Typical porosity in Boom Clay after manual segmentation; red and blue are pores inside clay matrix of fine-grained, clay-rich samples, green = pores within clay matrix of more coarsely grained, quartz-rich sample; other colours = porosity in non-clay minerals, e.g. mica or quartz.

Figure 13: Typical intragranular pores in framboidal pyrite imaged with Inlens detector.

Figure 14: Small intragranular pores in matrix.