2D and 3D high-resolution imaging to reconstruct the microstructure of clay media

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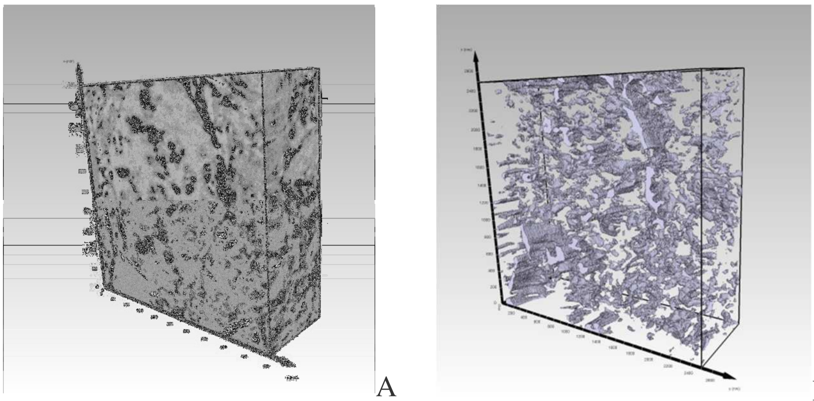
Clayrock formations are under consideration to serve as host formation and geological barriers for radioactive waste repositories due to their favorable properties (low permeability, low diffusion coefficient, high retention capacity for radionuclides…). The currently on-going European CATCLAY project (EURATOM FP7) (www.catclay.org) is focused on understanding the fundamental processes governing the diffusion-driven transport of cationic species in natural clayrocks. The project combines modeling with experimental studies of migration of three cations (Sr^{+2}, Zn^{+2} and Eu^{+3}) in a monophasic compacted clay (illite) system, considered to be an analogy for the clay matrix constituting clay-rocks, and three different clayrocks (Callovo-Oxfordian argilites (FR), Opalinus Clay (CH), Boom Clay (BE)). Part of the experimental studies are focused on the small scale structural (μm to nm) properties of the different materials in order to determine how the spatial distribution of mineral and pores at small scales can influence diffusion driven transport of cations. The present study focuses on compacted illite properties in hopes of extending this study to the natural clayrocks. Clay-rocks are more complex and include other mineral phases such carbonates or quartz. Our approach is mainly based on imaging the small scale structural organization of compacted illite material and analyzing the resulting images in order to extract information on pore space geometry/morphology and mineral spatial distribution, in order to use them as input data for transport modelling.

Small-scale investigations were performed using two high-resolution imaging techniques: Transmission Electron Microscopy TEM (2D imaging) and Focused Ion Beam/ Scanning Electron Microscopy (3D imaging). Both of these techniques need to work under vacuum conditions and therefore require removing water from pore space without disturbing its geometry. Consequently, techniques for imaging the texture of an illite material representative of its characteristics in a water-saturated and compacted state, were first developed. The first step was to improve on classic resin impregnation methods in order to preserve the texture without losing the clay confinement and modifying the pore space geometry. This has been done by taking into account the molecular size of the monomer used, its low viscosity and dipole moment (adapted for a clayrock with swelling clay minerals) and the controlled time polymerization. The MMA monomer proved to be the most suitable resin in our study.

A set of serial images (2048 ×1536 pixels) with a pixel resolution of 5×5 nm^2 were acquired using FIB/SEM and the "slice and view" method described in Holzer et al., (2004). Several image analysis tools were applied to the raw images using the free image processing software ImageJ (Abramoff et al., 2004) to remove classical FIB/SEM artifacts (vertical stripes were removed using Fast Fourier Transform images, alignment of images according to z axis…) and improve image quality (noise reduction, shadowing effect correction…). Analyses of the pore space and the construction of a 3D representation were then done with Avizo software. Segmentation tools provided by Avizo software were used to extract the pore network from the 3D volume (Figure 1). A set of tools were then applied to the binary (pore/solid) volume to quantity geometric and morphologic features (correlation functions, pore size distribution, pore orientation frequencies, tortuosity of the pore network…). Diffusion process modelling was also performed directly on the 3D volume using a random walk method (Robinet et al., 2012) to compute transport parameters such as the diffusion tortuosity.



A B

Figure 1 A 3D volume reconstructed from 2D FIB/SEM images. B-Pore network segmented from the 3D volume

As a result of the resolution limitations of the FIB/SEM technique, the 3D reconstructed volume represents a porosity which is characterized by pore radii larger than 15nm, which means that the smaller pores which account for about 75 % of the total pore space in such compacted clay materials could not be investigated in this study. A 3D structural analysis of these smaller pores is needed to conclude whether the larger pores are interconnected or if they form isolated pore objects.

For this purpose, a high-resolution TEM approach was applied to image the pore space and minerals at resolutions of several nanometers. For TEM observations, a set of ultra-thin serial sections (50 - 100 nm) were cut perpendicularly and parallel to the compaction axis using a microtome. A set of 2D images were then acquired using a spatial resolution ranging between 100 nm and 1 Å. From these 2D sections, various textural features were quantified using plugins implemented in ImageJ software. Automatic local orientation maps (Rezakhaniha et al., 2012) were computed to analyze the distribution of clay particles. Variogram techniques were also employed to discriminate the representative elementary volume (REV) based on the size and periodicity of the objects in the image. These techniques were applied at different magnifications, from the aggregate to the particle size to extract the main textural features of the clay media.

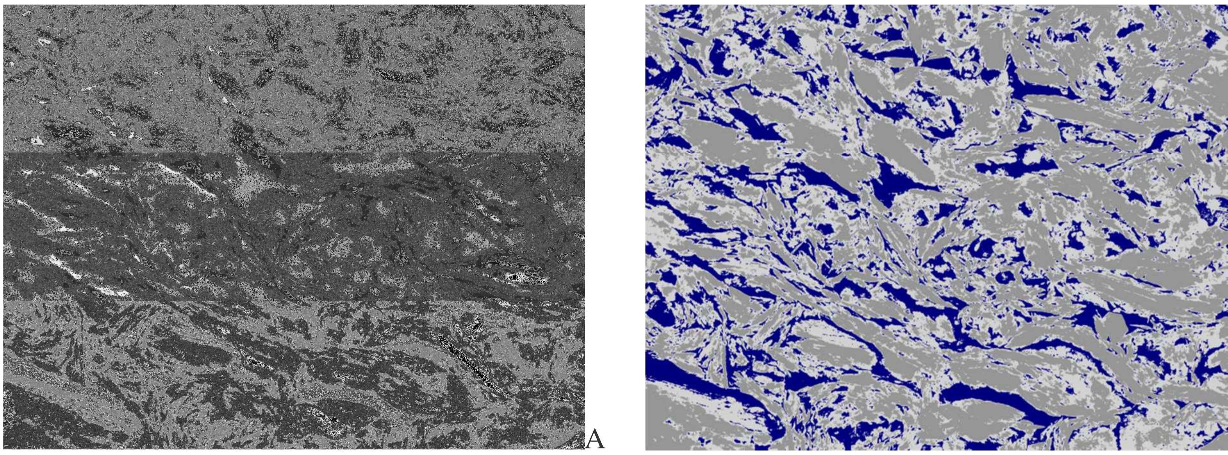


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Figure 2 TEM image of compacted clay material (A) and associated segmented image (B) (bleu: pores – grey: solid)

Imaging clay-rich porous media using high resolution techniques and performing a qualitative description of the images in terms of pore and mineral arrangement constitutes a first major result towards a better understanding of how transfer pathways are organized in the porous media. Thanks to a set image analysis methods including image segmentation, the main features characterizing pore and mineral geometries/morphologies can be quantified and directly used as input data in modelling approaches. Both methodologies (FIB-tomography and TEM techniques) can therefore be used in a complementary fashion for up-scaling the structural organization of compacted clayey materials. TEM images analysis allows down-scaling the resolution size since only a part of the pore space could be imaged with FIB/SEM method. Applying electron tomography techniques undoubtedly constitutes the next step to image such porous media in 3D with a sub-nanometer resolution. This technique could help us to extract the whole porosity domain and to resolve the connectivity of the pore space at the scale of a few nanometers.

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