EXPECTATIONS, OPEN QUESTIONS TO BE ADDRESSED IN THE WORKSHOP WITHIN THE CONTEXT OF A DEEP GEOLOGICAL REPOSITORY IN CLAY FORMATIONS

(Summary of the introductory presentation given by Patrick Landais, ANDRA;
Chair of the NEA Clay Club)

Concerning radioactive waste disposal in geological formations a growing consensus in the world can be observed that deep geological disposal is the most appropriate solution for the long-term management of spent fuel and long-lived wastes (especially high level wastes). This consensus is based on the scientific work over several decades to develop technical solutions for deep geological repositories and to assess their long term safety over long time periods (up to 1 Million year). In many countries (such as i.a. Belgium, Germany, France, Japan, Switzerland, United Kingdom) deep argillaceous formations are considered as potential host rock for radioactive waste disposal. In other countries favoring crystalline host rock formations (such as e.g. Sweden, Finland) clay-based materials (e.g. bentonite) are proposed as the geotechnical barrier of the multi-barrier system for a deep geological repository.

Precise knowledge of the clay properties in the various domains concerned by the construction feasibility, the exploitation phase of the repository facilities, as well as the long term evolution of the waste and of its environment is therefore of crucial importance in assessing the performance and the safety of the various radioactive waste disposal concepts. The knowledge to be acquired on clays as such goes well beyond solely the field of disposal of radioactive waste. For both, clay formations or bentonites in engineered barriers, the characterization in a continuous way from the nanometer to the micrometer, of their internal structure and the study of the associated physico-chemical phenomena is a fundamental issue. It aims for explaining:

- The “Initial state” of the clays, in particular for the clay formations: the nature of the mechanical, hydraulic and geochemical processes, in a broad sense, and the way these processes were involved during the geological history of these formations,
- The fundamental processes involved by physico-chemical or hydraulic stresses, related to the evolution of the repository at the macroscopic scale.

Some major (macroscopic) objectives of Research & Development (R&D) work can be pointed out. For an appropriate characterization of the 3D poral and mineral organization of the clay rocks at the infra-millimeter scale (a) the characterization and the modeling of the elementary HMC (hydrological-mechanical-chemical) mechanisms and the transfer of gas involved at very small scales and (b) the establishment of a link between the elementary HMC mechanisms and the fluid transfer is needed. Here, examples on the atomistic scale looking in detail on the structuring of water molecules at the clay mineral surface or the cation complexation at the clay mineral surface have improved clearly our knowledge.

Clay Rocks are a complex material in terms of *mineralogical composition* constituting of variable proportions of clay minerals and mainly carbonates/ tectosilicates and *microstructural organization* with variations of pore sizes and minerals from some nanometers to several tens of microns. The spatial arrangement of these minerals controls the distribution and the nature of the water (free, bound, interlayer) and of the solutes within the clay-rich rocks and consequently the reactivity under repository conditions. Therefore, the nano- to mesoscopic microstructure given by the mineralogy, the porosity and the pore size distribution and also microstructural variations on the formation scale due to sedimentary processes will determine the transport processes (anion exclusion, surface diffusion and water structuration) and transport properties (permeability, diffusion coefficients, ion concentrations) on the small and large scale (basin). Similarly, mechanical and gas transport properties are closely related to this microstructure and can be described with this approach (see Figure 1).

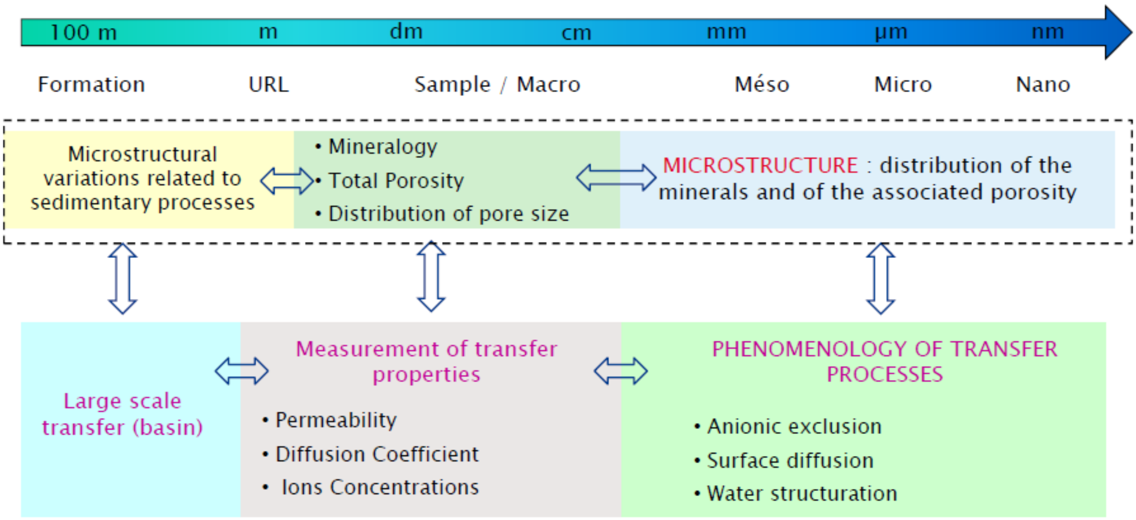

Figure 1. The multi-scale approach connecting mineralogy, porosity and transport properties.

Consequently, the choice of the characterization scale and relevant modeling is of first importance in the approaches leading to the establishment of the models of representation. Various research works pointed to experimental difficulties in quantifying the microstructure of the clay rocks at scales smaller than a micrometer, because of technical/instrumental limitations. This lack of knowledge at small scales does not allow to fully connect all the Thermal-hydrological-mechanical-chemical (THMC) mechanisms and to integrate them into an up-scaling approach.
There already exist conceptual models and experimental approaches to describe the microstructure of argillaceous formations in terms of porosity and texture (see Figure 2). Here, examples on the undisturbed Callovo-Oxfordian (COx) argillite are given:

- **Sample scale.** Variability investigations on the decimetre to centimetre scale on COx rock slices typically with a thickness of approx. 1cm by radiography and thin sections by optical microscopy have revealed a clear positive correlation of clay content and sample homogeneity showing highest heterogeneity in samples with carbonate rich zones and calcified zones having up to 65% carbonate content.
- **Mineral scale.** Investigating samples on the mineral scale by SEM- BSE/EDX with a resolution of 0.3µm and by X-Ray Microtomography under a voxel resolution of 0.7µm revealed a simplified 2D and 3D distribution of carbonates, tectosilicates, heavy minerals and the not further resolvable matrix. This information can furthermore resolved by microprobe (EPMA) analysis to obtain data beside the argillaceous matrix on calcite, quartz, dolomite, feldspar, pyrite, siderite and muscovite.
- **Pore scale.** As shown schematically in Figure 2 different types of pores have to be considered in argillaceous rocks with a) intergranular pores ranging from 50-100nm as observed in SEM and FIB-tomography and b) clay sheet stacks with interlayer pores considerably smaller that can be resolved by TEM having a resolution of 1-10nm.

Differences in bulk material properties (mass transfer) observed for anion and cation diffusion can be directly explained by molecular scale interactions (anion exclusion, surface diffusion) and/or directly linked to the geometry and poral network of the rock as in the case of the difference between horizontal effective diffusion coefficient ($D_{e,h}$) and vertical effective diffusion coefficient ($D_{e,v}$) with $D_{e,h}/D_{e,v} = 2$.

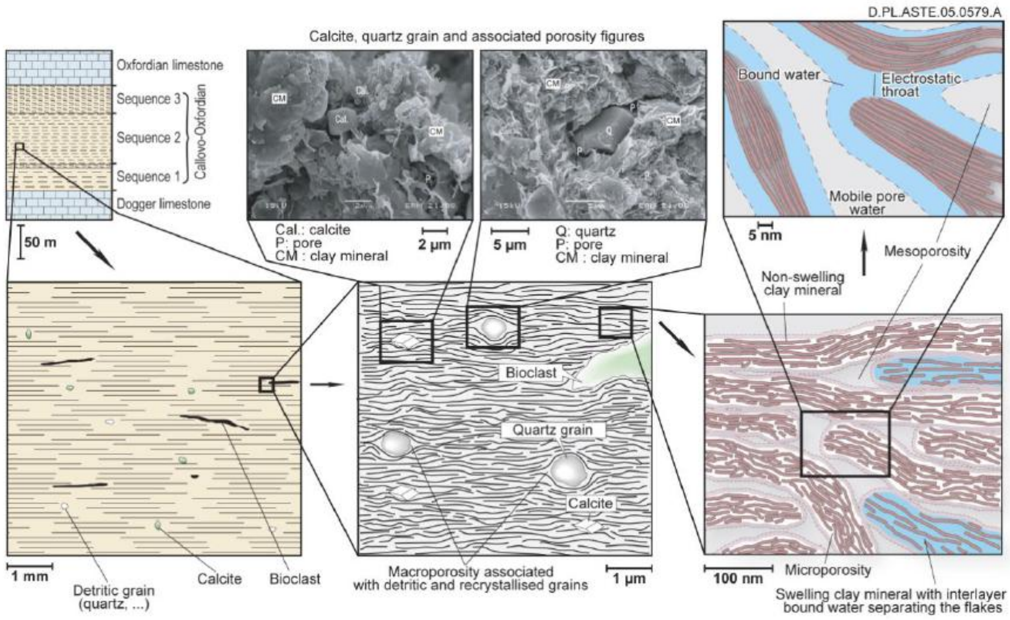

Figure 2. Conceptual model of the porosity scales to be considered within the Callovo-Oxfordian argillite (COX).

Another example is the issue of heat transfer properties, which has shown to be strongly influenced by the petrofabric and specifically by the distribution and content of heat conducting minerals (quartz, calcite). Here, in the bedding plane the thermal resistivity is weak, whereas perpendicular to the bedding plane the resistivity shows significant variability due to the clay mineral content and the porosity of the material.

Beside the characterization of the undisturbed clay rock an additional complexity is given by the processes involved during the evolution of a deep geological repository. Here, examples are given on a) the understanding of the long-term evolution of the EDZ, b) alteration processes at the interface between clay and engineered material and c) the complex interactions at the different interfaces in a high level waste cell.

a) The hydraulic properties of fractures created during the excavation of the repository infrastructure depend on their aperture (width). Those properties evolve as a function of the applied stress but also of the potential fractures self-sealing due to the swelling properties of the clay minerals. In the vicinity of seals and plugs the progressive re-saturation of the repository will be associated with the swelling of the clay minerals and the sealing of the fractures. If the bentonite plug is in contact with the geological formation its swelling pressure will increase the stress on the fractures.

b) In the framework of the iron (canister)-clay interaction the identification of corrosion rates, the corrosion reaction products and therefore the process understanding will lead to an adequate design (thickness) of the steel containers.

c) In a high level waste cell usually three type of materials have to be considered, namely the waste glass containing the radionuclides, the iron (steel) as canister material and the argillite/bentonite and it's contact with the steel canister. A temperature gradient will evolve from the glass to the argillite (host rock) whereas the penetration of water will progress from the far-field. Processes like glass dissolution and radionuclide release, the interaction with corrosion steel products and their interaction with mineralogical transformations of the argillite are of THMC (thermo-hydro-mechanical-chemical) type and occur on small to very small scales.

Overall, these three examples above highlight that a more precise and detailed description on the nano- to microscale is needed for the progressive optimization of the repository architecture and components.

In the following, questions and objectives to be addressed during the workshop on undisturbed argillaceous rocks concerning the characterization of the poral and mineral 3D network geometry at the infra-millimeter scales are highlighted.

- What is the spatial distribution and the morphology of pores?
- How are they organized with regard to minerals?
- What are the surfaces of interaction between minerals and pores?
- How get organized minerals at small scales?
- What is the morphology of the contacts between minerals?
- Are non-clay minerals in inclusion within a clay phase?
- Do carbonates constitute a skeleton?

Further objectives and questions in mechanical disturbed could include the following:

- Determination of the mechanical damage as a function e.g. of the type of mechanical discharge, deviatoric stress (in compression or in extension)
- In the case of a thermal load, in what pores is the dilatation of the water located? Is there a redistribution of the proportions of the various types of water? How is the pressure of the water on minerals reorganized? Where and how the porosity redistributed?
- Where are the deformations located? (Micro- fissuring of non-clay minerals and\or the clay phase, detachment between clay and non-clay minerals, sliding, dilatance of clay minerals)
- Is there an evolution of the nature of the water in the zones where deformations is located?

In chemical disturbed systems a focus is on precipitation and dissolution processes and their influence on transport/mass transfer parameters and the following questions inter alia can be raised:

- How and where in a 3D porous structure take the processes of dissolution-precipitation in saturated conditions and in non-saturated conditions to place?
- What pores are involved in the transport of solutions?
- Is there a partial or total accessibility of all the minerals to aggressive solutions?

  Beside the number of questions raised above the workshop was primarily intended to give an overview of currently available techniques as listed in the glossary using different incident beams, e.g. x-rays, neutrons, ions and electrons.

In the oral presentations by the invites speakers of the workshop the experts in the field/ technique are encouraged to give information on technical questions as e.g.:

Instrumental limitations? The Beam/probe technology specific resolution limits, incident energy and invasive/destructive or non-invasive/non-destructive method, sensor technology and detection limits.

How are images obtained? Addressing issues like sample collection and preparation/ preservation of samples (so called “Environmental” imaging by preservation of water content and textural information), image acquisition, and signal processing/signal mapping/ signal reconstruction as well as the elementary representative volume analysed.

What kind of information is collected? Here differentiation should be made between physical parameters (e.g. temperature, thermal conductivity, and magnetism), chemical parameters (e.g. composition, crystal structure), textural parameters (e.g. porosity, orientation, surface, relief) and the potential monitoring of property changes (spatial, temporal resolution, real time analysis). Furthermore, information on qualitative or quantitative results and the potential way to obtain quantitative data by combining different techniques on the same sample or the combination of microscopic and spectroscopic techniques should be given.

Patrick Landais ended his presentation with some concluding remarks/observations highlighting the importance of micro- to nano focusing techniques:

- Radioactive waste disposal offer a wide range of questions connected to various research areas
- Clay minerals and clay rocks still need to be studied in detail in order to provide robust models for the evaluation of the processes occurring during the evolution of an underground disposal
- Issues related to thermo-hydro-mechanical-chemical (THMC) processes are central when evaluating the safety of an underground repository
- Nano or microscopic resolution techniques provide new insights for the understanding of clay minerals reactivity
- Some phenomena, the description and understanding of which require micro-nano approaches, still constitutes major issues for the demonstration of the safety and the dimensioning of repository components.