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How rock samples can be representative of in situ condition: A case study of Callovo-Oxfordian claystones

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ABSTRACT

Within the framework of feasibility studies for a reversible, deep geological repository of high- and intermediate-level long-lived radioactive waste (HLW, IL-LLW), the French National Radioactive Waste Management Agency (Andra) is investigating the Callovo-Oxfordian (COx) formation near Bure (north-east part of France) as a potential host rock for the repository. The hydro-mechanical (HM) behaviour is an important issue to design and optimise different components of the disposal such as shaft, ramp, drift, and waste package disposal facilities. Over the past 20 years, a large number of laboratory experiments have been carried out to characterise and understand the HM behaviours of COx claystones. At the beginning, samples came from deep boreholes drilled at the ground surface with oil base mud. From 2000 onwards, with the launch of the construction of the Meuse/Haute-Marne Underground Research Laboratory (MHM URL), most samples have been extracted from a large number of air drilled boreholes in the URL. In parallel, various constitutive models have been developed for modelling. The thermo-hydro-mechanical (THM) behaviours of the COx claystones were investigated under different repository conditions. Core samples are subjected to a complex HM loading path before testing, due to drilling, conditioning and preparation. Various kinds of effects on the characteristics of the claystones are highlighted and discussed, and the procedures for core extraction and packaging as well as a systematic sample preparation protocol are proposed in order to minimise the uncertainties on test results. The representativeness of the test results is also addressed with regard to the in situ rock mass.

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1. Introduction

The French National Radioactive Waste Management Agency (Andra) has been performing a comprehensive scientific programme to acquire knowledge on Callovo-Oxfordian (COx) claystones through theoretical work, laboratory testing on samples, large-scale experiments and numerical modelling. The main objective of the studies is to characterise and understand the thermo-hydro-mechanical (THM) behaviours and associated mechanisms of the COx claystones. Such models could be used to

design the French Radioactive Waste Disposal Project (CIGEO), demonstrate the relevancy of the design, and optimise the design.

In the 1990s, a geological investigation area covering over 100 km² was carried out involving geophysical survey and drilling of boreholes near Bure village in eastern France. The surface reconnaissance programme helped to locate the site of the Meuse/Haute-Marne Underground Research Laboratory (MHM URL), and provided an impressive wealth of data covering a wide range of geosciences like hydrogeology (Distinguin and Lavanchy, 2007), structural geology (Vigneron et al., 2004), mineralogy (Gaucher et al., 2004), geochemistry (Vinsot et al., 2008; Dewonck et al., 2010), rock mechanics (Lebon and Ghoreychi, 2000; Armand et al., 2017), and so on.

In 2000, Andra began to build the MHM URL by shafts sinking to study the COx claystones lying between 420 m and 550 m below the ground surface. The claystone layer is overlain and underlain by poorly permeable carbonate formations (Dogger and Oxfordian

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limestone layers). The main objective of the first research (Delay et al., 2007) phase (2000–2005) was to characterise the containment properties of the clay through in situ hydrogeological tests, chemical measurements and diffusion experiments in order to demonstrate that the construction and operation of a geological repository would not introduce pathways for radionuclides.

Excavation monitoring works and associated geomechanical experiments at the MHM URL have been designed and carried out to provide information on the hydro-mechanical (HM) behaviours of COx claystones. Since 2004, data have been continuously acquired through several measurement campaigns mainly in galleries at the main level –490 m (clay unit level, i.e. UA level), but also in a niche at –445 m (silty carbonate unit level (USC)). These observations allow a better understanding of the HM behaviours of claystones and highlight various phenomena. Up to now, the research programme continues and is also dedicated to technology improvement and demonstration issue of the different disposal systems. Characterisation studies are still on-going.

In addition to in situ experimentations, numerous samples are regularly extracted for laboratory tests. These tests serve to improve the state of knowledge on the HM behaviours of the claystones and to determine the main parameters for numerical modelling. They can be then used in the constitutive laws in order to better represent the behaviours of structures by integrating numerous mechanisms at different scales of observations. However, one of the difficulties encountered in the laboratory tests carried out on samples is the representativeness of the material compared to the COx claystones in their natural environments and ultimately the relevance of the measurements.

In fact, the COx claystones are very sensitive to the external environments and in particular to changes in water content and stress, and particularly in the UA because of the greater proportion of clays.

Many studies have shown the effects of drying and wetting cycles on COx claystone samples. Samples are subjected to air with a controlled relative humidity (RH) and mass or/and strain changes are measured. For example, Guillon et al. (2012) performed static atmosphere drying tests which highlighted the HM coupling and mechanical anisotropy of the samples. Both wetting and drying paths were explored, allowing the potential hysteretic behaviours of the samples. During imbibition, the swelling of the sample is mainly achieved by the interaction of water with swelling clay sheets (illite/smectite interstratified). The opening of some cracks can also be responsible for the dilation of the sample.

On the drying path, water is removed from the clay minerals that shrink. As the material progressively dehydrates, water menisci form in the porous space. These menisci can strengthen the cohesion between grains and improve the apparent stiffness of the rock (Pham et al., 2007). This issue is highlighted by triaxial tests performed at the end of the drying tests. Zhang et al. (2012) performed micro-indentation and mini-compression tests on the small samples of COx claystones under different RH values. They reported that the mechanical properties of the claystones are strongly influenced by water saturation and structural anisotropy. At a smaller scale, Wang et al. (2013) also studied the evolution of the COx claystones under hydric cycles using techniques based on the combination of environmental scanning electron microscope (ESEM) and digital image correlation (DIC), in order to quantify local strain field. The observation is carried out on zones of several hundred micrometres, thus the evolution of such material under hydric loadings can be studied at the scale of inclusion-matrix composite. A heterogeneous strain field is evidenced at micro-scale during wetting. Nonlinear deformation is observed at high RH value which is related not only to damage, but also to the nonlinear swelling of the clay mineral itself, controlled by different local

mechanisms depending on RH value. This sensitivity to variations in water content leads to enhanced variations of mechanical parameters in response to water saturation.

The extraction and conditioning procedures and the sample preparation phase are essential. If they are not properly controlled, desaturation and deconfinement of the material will be induced and the sample will be damaged subsequently. This damage can have a significant impact on some parameters such as the Young's modulus and gas permeability. Consequently, it is difficult to distinguish the real variability of parameter due to slight mineralogical change in a same litho-stratigraphic unit from variability induced by the sample state subjected to preparation conditions. In order to better assess the variability of mechanical parameters, Andra and associated academic laboratories try to improve procedure and better assess the effective variability of mechanical parameters induced by mineralogical changes.

Andra has also implemented procedures for core extraction and conditioning, in particular by developing a cell that allows to maintain a state of stress on the core and to preserve water saturation. A sampling protocol was proposed by its scientific partners, in order to preserve a healthy initial state among others, and an initial systematic characterisation which allows controlling the impact of the upstream phases on the core sample.

Despite all these precautions, slight desaturation of the sample is often observed. It is therefore necessary to study the impact of the initial or modified water saturation on the HM characteristics of the claystones. Several studies have been carried out using different resaturation techniques either through percolation or using a controlled RH chamber. As expected, a reinforcement of the material is generally observed when the sample is desaturated. On the other hand, a drastic drop in mechanical properties on the fully resaturated samples was observed, suggesting a possible degradation of the material during the resaturation phase. As suggested by some authors (e.g. Ewy, 2015), when necessary, the protocol of resaturation must be particularly controlled and it is imperative to resaturate the sample under in situ stress in order to avoid damaging to the material.

The first part of this paper is devoted to the presentation of COx claystones: microstructure, mineralogy, and physical properties (porosity, water content, etc.). Then, an optimised procedure for core extraction and packaging is proposed as well as a systematic sample preparation protocol in order to minimise the uncertainties. In this protocol, some methods such as tomography are used to eliminate microcracked samples. Next, analysis of mechanical behaviour of a rock sample with respect to the initial conditions is presented. A comparison of mechanical test results obtained on artificially resaturated samples and samples whose saturation was preserved during the sampling process shows the negative impact of the resaturation process on the mechanical parameters. Finally, the representativeness of the pre-testing resaturation process with regard to field material properties is discussed.

2. Physical properties

This section is devoted to description of the main physical properties of COx claystones (mineralogy, water content and porosity). The experimental protocols used to determine these physical properties are presented with their reference values and associated uncertainties.

Sediments of the COx formation consist of a dominant clay fraction associated with carbonate, quartz with minor feldspars, and accessory minerals (Lerouge et al., 2011). Variations in the sea level at the time of the deposit result in variations in the sedimentary layers: the higher the sea level is (referred to as the maximum flooding), the greater the argillaceous fraction is. In

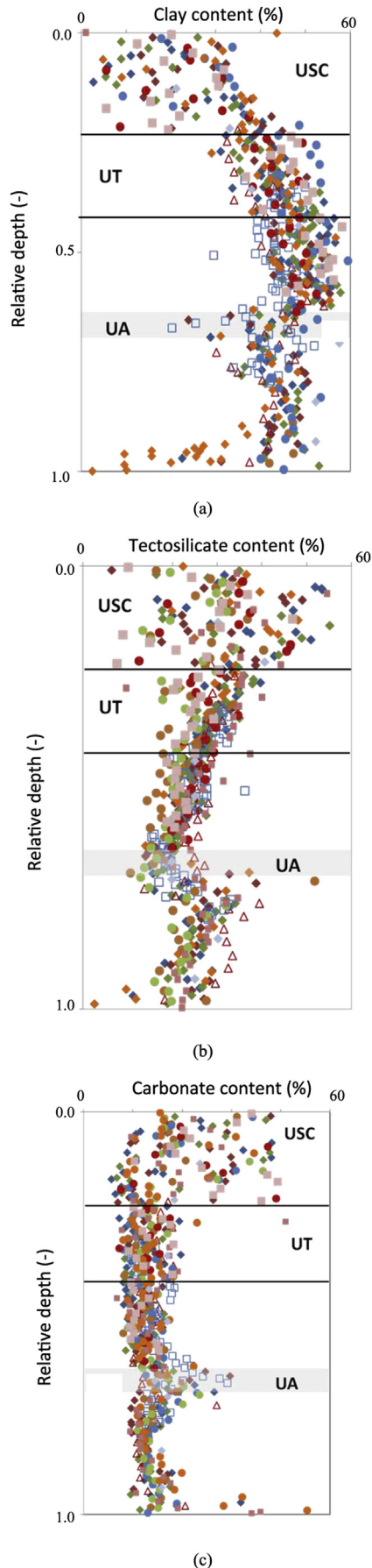


Table 1
Average mineralogical composition per COx geological unit.

COx	Phyllosilicates		Carbonates		Tectosilicates	
	Average (%)	SD (%)	Average (%)	SD (%)	Average (%)	SD (%)
UA	46	7	27	8	24	6
UT	43	8	26	8	28	8
USC	26	11	42	18	30	10
Total	42	11	30	12	25	8

Note: SD is the standard deviation.

contrary, the lower the sea level is, the greater the carbonate deposits are. The COx formation is divided vertically into three lithostratigraphic units from the base: the UA rich in clay content, the transition unit (UT), and the USC with higher carbonate content.

The sedimentation processes and the morphology of clay minerals are the origin causing anisotropic behaviours of COx formation. It is noticed that the in situ stress state is also anisotropic. The major horizontal stress is higher than both the vertical one and the minor horizontal stress that are roughly equal (Wileveau et al., 2007). The major horizontal stress σ_H (oriented at N155°) is about 16 MPa, while the vertical stress σ_v and the minor horizontal stress σ_h (oriented at N65°) are both about 12 MPa.

2.1. Mineralogy of COx claystones

Andra has performed numerous mineralogical tests on core samples to study the distribution of mineralogical composition of the COx claystones. The samples taken from the cores are usually packaged in heat-sealable bags to limit alterations of the highly-reactive minor phases, such as pyrite. The COx claystones consist mainly of phyllosilicates, carbonates and tectosilicates, which represent around 95% of the constituents. These minerals are typical of sedimentary clay-rich rock mass. The mean content of the various mineral groups and their associated standard deviations (phyllosilicates, tectosilicates, carbonates and ancillary minerals) were calculated at the scale of the studied area over the entire thickness of the layer (see Table 1). The average distribution of mineral groups in the claystones throughout the whole formation is clay fraction (phyllosilicates) of $42\% \pm 11\%$ of the rock, carbonates of $30\% \pm 12\%$ of minerals, tectosilicates of $25\% \pm 8\%$, and ancillary minerals constitute less than 4%. Mineralogical analyses show low variability in mineral content in the bed plane and significant vertical variations (see Fig. 1). These mineralogical variations can be correlated with the microstructure of the claystones and directly influence the measured HM values. For example, uniaxial compressive strength (UCS) measured in areas with high carbonate content as encountered in the USC unit is much higher than those in clay-rich area (UA and UT units).

2.2. Water content

The water content (w) of the COx claystones is determined by the mass of the sample in its wet state (m_w) and its dry mass (m_d) (CFMR, 2000):

$$w = \frac{m_w - m_d}{m_d} \tag{1}$$

The dry mass is obtained by drying at temperature 105 °C and/or 150 °C for 24 h (m_d). The so-called “natural water content” refers to

Fig. 1. Variation in mineralogical composition across the thickness of the COx formation. Data come from different boreholes and the relative depth is between the top of the USC unit and the bottom of the UA unit.

as the water content corresponding to the in situ condition, i.e. unaffected by any engineered activities (excavation, ventilation, etc.). For the measurement to be representative of the natural conditions, it should not be influenced by the samples conservation and transport (see Section 4.2).

A vertical distribution of water content is given in Fig. 2 across the thickness of the COx formation. Water contents are measured between 5% and 8% in the UA level. At –490 m, the reference value used is 7.9% ± 0.6% (see Table 2). The density of the grains (ρ_s) of the COx claystones is about 2.69 g/cm³. A clear correlation has been established between the clay content (Fig. 1) and water content (Fig. 2b).

2.3. Porosity

The total porosity (ϕ) of COx claystones depends on their mineralogical composition, the nature of the clays, the arrangement of the minerals, the rock depth, and their geological history (Robinet et al., 2015). Two methods are used by Andra to measure the porosity of the COx claystones: the nuclear magnetic resonance (NMR) log and the water content measurement. The NMR method provides a direct measurement of porosity in the borehole with an accuracy of 1%. The measurement tool uses the nuclear magnetisation of the protons inside liquids (thus hydrogen atoms) to saturate the porous medium. The second method allows determining the porosity by measuring the water content at 150 °C. The porosity is expressed as

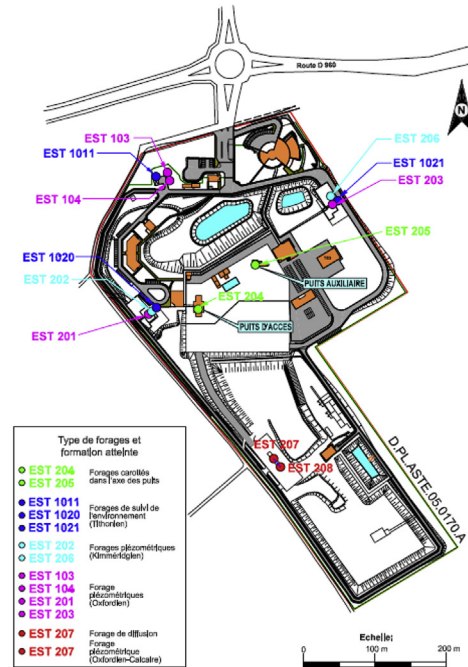
$$\phi = \frac{\rho_s W}{1 + \rho_s W} \tag{2}$$

The porosity values measured by Andra are given in Table 2.

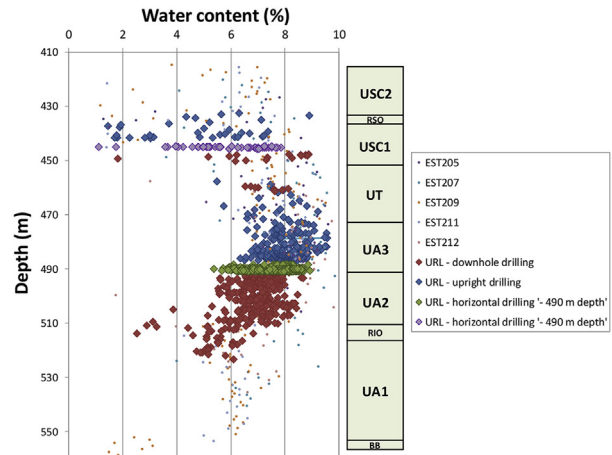
3. Core extraction and conditioning

The COx claystones exhibit a natural variability due to the variation in their mineralogical composition, which must be taken into account in the analyses of the mechanical tests. However, it is important to distinguish this variability from that related to the state of the samples. For several years, Andra has been working to optimise and improve sampling methods from drilling to laboratory tests. The aim is to have the most representative samples of the natural state of the claystones.

Numerous measurements on samples have been made since the URL was built in 2000. Nearly 1500 cores have been extracted and delivered to laboratories for testing, but the multitude of methods for both sampling and test protocols often makes it very difficult to interpret the results and compare them.



(a) The MHM URL.



(b) Water content with depth.

Fig. 2. Vertical distribution of the measured water content at the MHM URL.

Table 2 Water content (w) and porosity (ϕ) of the Callovo-Oxfordian claystones.

Water content (w)		USC		UT + UA3 + UA2		UA1		–490 m depth	
Totality	SD (%)	Average (%)	SD (%)	Average (%)	SD (%)	Average (%)	SD (%)	Average (%)	SD (%)
7.1	1.7	6.7	2	8	1.2	6.2	1.3	7.9	0.6
Porosity (ϕ)		USC		UT + UA3 + UA2		UA1 + RIO		–490 m depth	
Totality	SD (%)	Average (%)	SD (%)	Average (%)	SD (%)	Average (%)	SD (%)	Average (%)	SD (%)
16	3.3	15.1	4.2	17.7	1.9	14.2	2.3	17.5	1.2

Note: RIO is the Repère Inférieur Oolithique for petroleum geologist.

The quality of clay/shale cores that are extracted from great depths and tested in the laboratory to determine their mechanical properties is a major concern, as discussed by Ewy (2015). Materials are subjected to the successive effects of coring, deconfinement, transport, conditioning, and laboratory preparation, which often lead to desaturation and damage. Ewy (2015) recommended wrapping cores for a few minutes after extraction to avoid prolonged exposure to air and applying confining stress within one hour. To optimise the quality and conservation of COx cores prior to preparation and testing, Andra developed a special packaging cell called T1 cell. The T1 cells are intended to maintain a state of stress on the core and to avoid core desaturation and microcracking. At the URL, most cores are extracted at the main level (−490 m depth) and for logistical reasons, packaging cannot be carried out at that depth. The time taken to raise/treat the cores must therefore be kept to a minimum. To check whether the time-scale recommended by Andra is short enough to maintain a satisfactory saturation level at the specimen scale, a new water-content measurement campaign has been carried out on cores immediately being wrapped under a leak-proof film and on cores rose using the usual procedure. Additional measurements were also carried out on cores already packaged in T1 cells to check the permeability of the latex sheath around the material. The purpose of this sheath is to avoid water exchange and thus to limit claystones desaturation.

3.1. Special confining T1 cell

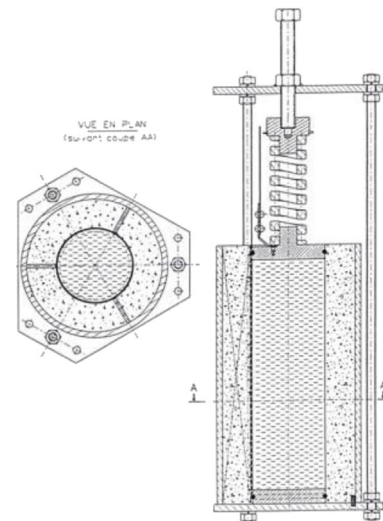
The T1 cells (see Fig. 3) developed by Andra are used to condition the COx claystone samples. The design of these cells allows a vertical stress up to about 6 bar (1 bar = 0.1 MPa) to be applied to the sample by means of a compressed spring and radial confinement using a non-shrinking expansive mortar, in order to keep the decompression of the material to the minimum. The sample is preliminarily placed in a thick latex membrane to avoid desaturation.

The insertion of cores into T1 cells is highly controlled in a systematic way. At the exit of the corer, a segment of 1.5 m long is protected with a plastic box to limit its desaturation and raised to the sample conditioning workshop. The drilling is carried out using a process that avoids vibrations. The top and the side walls of the sample to be taken (nominally 32 cm long) are immediately identified by the geologist team who performs coupling analysis of the cores. Samples are cut using a diamond circular saw bench. The cutting is performed in dry state and must be done smoothly without vibrations. It is achieved by rotating the core on a roller device. The upper and lower faces of the sample must be completely flat, parallel and perpendicular to the core axis. Immediately after the sample is cut, it is brought to the sample conditioning workshop and is placed in a T1 cell as quickly as possible.

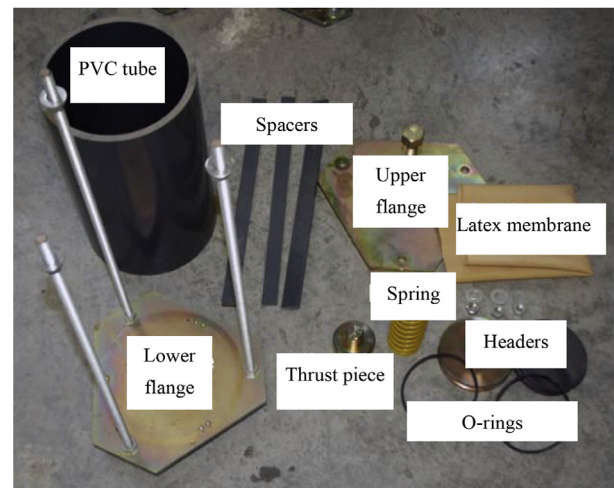
From boreholes of 101 mm in diameter, one can obtain cores in 79 mm diameter. The core is inserted into a latex membrane and placed in a PVC tube (Fig. 3). Two bases are inserted on each side of the membrane and blocked with two O-rings placed over the membrane. A non-shrinkage mortar is poured between the core and the tube to ensure radial confinement. The upper face is then constrained by the spring. All these operations must be ideally carried out within one hour after the core is extracted from the borehole. COx claystone samples should be handled as carefully as possible, in order to avoid any vibration, impacts and/or rotation.

3.2. Time effect on the water content

In order to illustrate the evolution of water content of cores with elapsed time, two measurement campaigns on water contents were



(a)



(b)

Fig. 3. (a) Schematic representation of a T1 cell; and (b) Photograph showing T1 cell components.

carried out on core samples from boreholes OHZ3004 and OHZ5015 horizontally drilled in the MHM URL. The objectives of both campaigns were to:

- (1) Assess the impact of the time taken to raise the core from the bottom (drilling site at −490 m) to the surface (sample conditioning workshop) on the water content,
- (2) Find out whether it is necessary to immediately wrap the core at the borehole outlet, and
- (3) Assess the impact of drilling on the homogeneity of the water content in the core by measuring the centre and the periphery of the core.

In addition to these objectives, the second campaign aimed to study the reliability of T1 cells by measuring the:

- (1) Change in water content over time on T1 cells from the same borehole, and
- (2) Water contents in T1 cells taken several years ago and stored in a core sample library to watch their evolution.

3.2.1. Measurement protocols at borehole outlet

The two campaigns were launched by following the same protocol. Two core samples were taken at depths of more than 10 m in horizontal boreholes, i.e. outside the influence area of the gallery, which is approximately one-diameter distance. Each core was 1 m long. At borehole outlet, one of the cores was immediately wrapped in plastic film before being placed in the plastic box and then raised to the sample conditioning workshop. The second core was unwrapped. In both cases, the time taken to raise the sample was minimised so that the measurements could be immediately conducted as possible.

On each of these cores, three cylindrical patties of 5 cm thick were sampled at regular interval along the 1 m sample length (to represent the average of 1 m of the material). A water content measurement was performed on pieces taken at the centre and periphery of each patty. The cutting of the wrapped core was carried out without removing the film. The remaining pieces (for subsequent measurements) were then rewrapped as soon as possible with plastic film and aluminium foil. Concerning the unwrapped core, the remaining pieces were stored uncovered at ambient temperature pending further measurements. Subsequently, 4 measurements were carried out on the remainder of each core with a periodicity ranging from a few hours to 10 d for the last measurement. The initial measurements are given in Tables 3 and 4.

3.2.2. Protocol of measurements of evolution of water content with time in T1 cells

During the first campaign, samples were also collected in three T1 cells (EST53495, EST53498, and EST53500) from borehole OHZ3004, at depths between 12 m and 15 m. These cells were prepared using conventional procedure and stored for further testing. A water content measurement was carried out at the centre

Table 3
Initial measurements of water content (%) in OHZ3004 borehole.

Unwrapped		
Section	Centre	Periphery
M1	8.32	7.88
M2	8.22	7.86
M3	8.53	8.21
Average	7.98	8.36
Wrapped		
Section	Centre	Periphery
M1	8.08	7.91
M2	7.97	8.02
M3	8.17	7.97
Average	8.07	7.97

Table 4
Initial measurements of water content (%) in OHZ5015 borehole.

Unwrapped		
Section	Centre	Periphery
M1	7.48	7.37
M2	7.54	7.15
M3	7.45	7.12
Average	7.41	7.21
Wrapped		
Section	Centre	Periphery
M1	7.49	7.43
M2	7.54	7.51
M3		
Average	7.52	7.47

and periphery on each sample. These measurements were considered as the initial water content measurements of these T1 cells. Three subsequent measurements were then carried out on the same T1 cells following the same procedure as previously discussed on 3 pieces taken along the T1 cell. The first measurement was carried out after 15 d, the second after 6 months and the last about one year after conditioning in T1 cell.

Seven old cores (more than seven years old) confined in T1 cells were also selected to measure their water contents. Three measurements were performed as per previous cells as these cells had the common feature of more than 7-year history. Three of these cells were derived from deep boreholes EST423, EST433 and EST441 (Table 5) drilled from the surface but located in the same geological unit as the MHM URL. The remaining four samples were from the MHM URL, TED1001 and PGZ1201 (Table 5).

3.2.3. Results

For both campaigns, the average time to bring the core from –490 m depth to the sample conditioning workshop was one hour. Fig. 4 shows the change in water content over time for unwrapped and wrapped cores.

These measurements show that the initial water content is the same for the wrapped and unwrapped cores. This shows that if the core is brought to the surface quickly enough, it is not necessary to protect the core with a plastic film, suggesting that the closed plastic box is sufficient to avoid its desaturation at that time. During the first campaign, the measured values are different between the core centre and the periphery. This indicates that the drilling slightly desaturates the periphery of the core. This difference is nevertheless lower than the precision of the measurements and was not highlighted during the second campaign. It should be noted that there is a re-homogenisation of the water content over time in the wrapped core, and the difference between the outside and the inside of the core seems to diminish. Although the successive measurements remain almost constant in the wrapped core in comparison with the unwrapped core, there is an almost immediate decrease in water content from the first hour after the sample is raised, confirming the need to perform conditioning operations as quickly as possible to avoid exposure to air. Ten days later, the water loss can reach 40%.

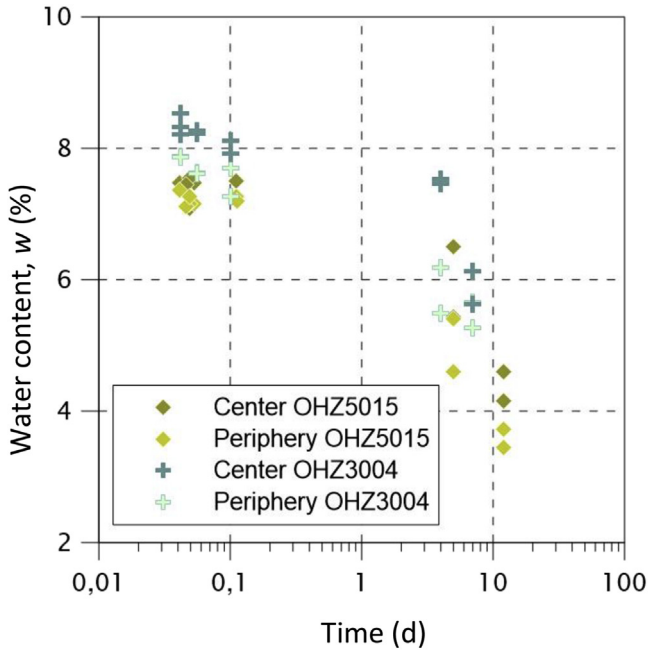
An interesting point to be noted is approximately 1% difference in water content value of the two campaigns. This probably reflects the natural variability of the environment. The main consequence is that, in order to accurately assess the degree of saturation of core, a porosity measurement has to be taken in parallel. Assuming that the core is close to water saturation, the porosity is 16.8% in one case and 17.8% in the other. This porosity range is consistent with the values obtained for the UA unit (Table 5).

The mean water contents measured on the T1 cells (EST53495, EST53498, and EST53500) from borehole OHZ3004 are shown in Fig. 5. A slight decrease is observed as early as the 6th month but appears to be less on the sample tested at one year later. Despite of

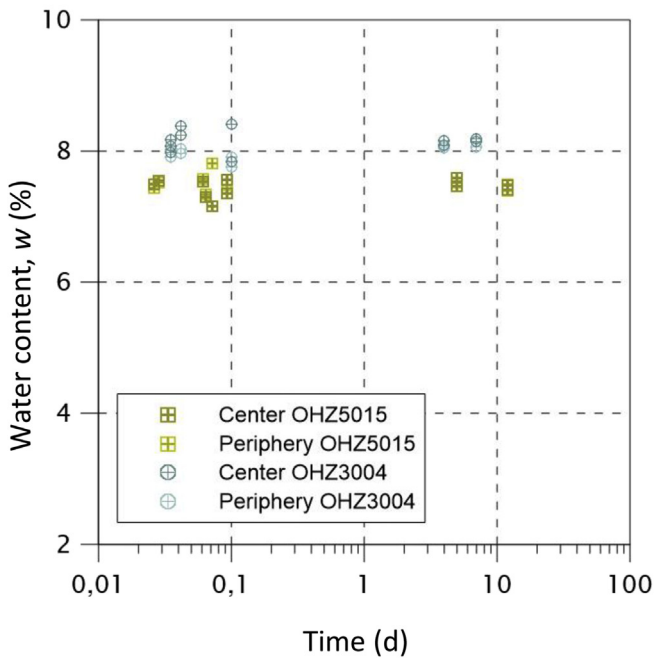
Table 5
Water content measured on old T1 cells.

Borehole ID	Drilling date	Core ID	Depth (m)	w (%) (measured)	w (%) (initial)
TED1001	2008	EST30130	13.8	9.12	n.c
TED1001	2008	EST30132	14.34	8.96	n.c
PGZ1201	2009	EST33556	26.03	8.19	6.7
PGZ1201	2009	EST33553	25.1	7.96	6.5
EST441	2008	EST28921	640.15	5.2	7.27
EST433	2008	EST29290	624.03	5.03	7
EST423	2008	EST28137	579.25	6.05	7.7

Note: n.c means no measured value.



(a) Unwrapped cores.



(b) Wrapped cores.

Fig. 4. Changes in water content over time for (a) unwrapped cores and (b) wrapped cores from the first (OHZ3004) and second (OHZ5015) campaigns.

the slight water loss being observed, it seems that in the short term, the latex sheath correctly ensures its function of sealing.

Finally, the measurements made on the old cores show that in the long term, the latex deteriorates (Table 5). This point is confirmed by the observations as illustrated in Fig. 6.

Two results emerge from these measurements, in which the boreholes were drilled from the URL, and the water content appears to have increased. This is confirmed by the measurements taken at the time on the core as they were extracted from borehole PGZ1201

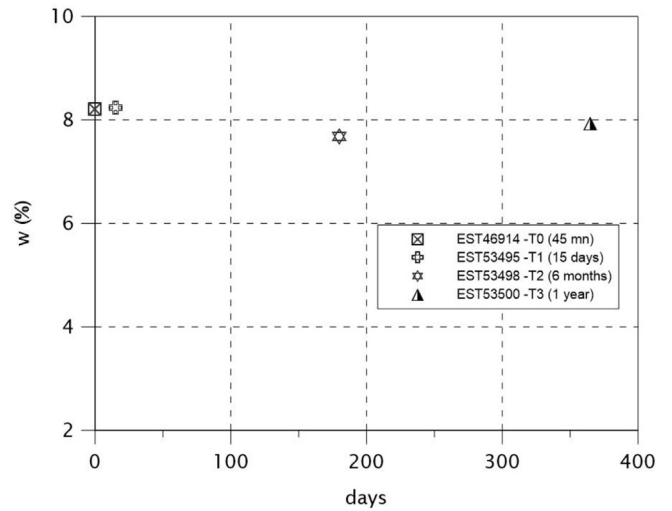


Fig. 5. Change in water content in a T1 cell.



(a)



(b)

Fig. 6. (a) Core from borehole EST423 after opening a T1 cell, and (b) Core from borehole TED1001 carried out 8 years ago.

which indicated an average water content of 6.6% (see Table 5). Conversely, for the boreholes drilled from the surface, the water content appears to have decreased. This is confirmed by the measured values close to the cell when the borehole was drilled. This difference in physical behaviour is probably due to the change in latex sheath that was operated between the boreholes from the

surface and those from the URL. These results do not allow conclusions to be drawn about the date when the desaturation or increase in water content occurred. However, given the results for the more recent cells (see Table 5), it likely occurred after the first year.

These campaigns verified that the recommended time of less than one hour for removing cores and placing them in T1 cells is sufficient to avoid core desaturation even if the core is unwrapped. They have also led to improvements in both T1 cell conditioning and core characterisation protocols. In order to preserve as much as possible the initial water content, the decision was made to add an extra protection consisting of a self-adhesive aluminium foil prior to the installation of the sheath. When the initial condition of cores is known, a water content measurement can be conducted systematically as possible to the core placed in T1 cells. This scenario is critically important, in particular for laboratories performing HM tests to determine the initial state of material. Other measurements such as porosity should also be introduced in future.

4. Laboratory experimental protocol

Precautions have to be taken when preparing COx claystone samples to minimise the risk of damage. The main difficulty lies in the fact that these materials are very sensitive to variations in confinement and desaturation. Moreover, when in contact with oxygen, the pyrite inclusions can oxidise and lead, for example, to cracking expansion of the corrosion products. It is generally difficult to detect this damage, which is characterised by submillimetric microcracks. The introduction of sampling protocols (machining, cutting and initial characterisation) to the laboratories using T1 cells and the homogenisation of these protocols can reduce the risks of damage and give better comparability of the results presented by several teams. During the sample preparation phase, a precise initial characterisation (water content, porosity, carbonate content, density, initial damage, etc.) is systematically carried out to provide macroscopic indicators of the sample condition and to assist in the interpretation of test results by integrating the potential natural variability of the rock mass. This section presents the protocols used to process the COx claystone samples.

4.1. Sample machining/cutting

Air coring, lathing and polishing can be used to machine, cut, and eliminate roughness or rectify the surface of the sample. On the other side, oil coring, drying of the sample and the use of humid air during coring are prohibited as they may cause a disturbance in the initial condition of the sample. The maximum shelf life of prepared samples is set at 1 year.

To preserve the samples before use, the following protocol has been defined: (1) wrap the sample under plastic film (preserving water content), (2) apply a low vacuum to remove the oxygen, (3) place the sample in an environment with a minimum pressure of at least 2 bar under inert N₂ or Ar (no CO₂) atmosphere or under water, (4) label with the sample production date, the core ID, and the sample orientation within the core to indicate its position within the formation, and (5) automatically report the sample preparation date and its date of use.

4.2. Characterisation of initial condition

During the samples preparation, a few parameters are measured almost systematically.

4.2.1. Water content (by mass)

The initial water content must be measured by weighing a sample as soon as the T1 cell is opened, and then it is dried at

105 °C. The sample mass is considered to be stable when the variation between two successive weighs at a 24-h interval is less than 0.1%. When the sample conservation time is longer than 1 month, a water content measurement is required before each test. Control samples (samples damaged or dropped in preparation, etc.) are kept under the same conditions as the specimens used in the tests.

4.2.2. Porosity

A porosity estimate should be made for each core used in a test campaign, and the method used for this estimation must be given in the test report. Mercury porosimetry can also be carried out to complement the traditional method, if the laboratory considers this to be necessary.

4.2.3. Carbonate content, grain density and suction

If the laboratory is equipped with suitable equipment (e.g. Bernard calcimeter, a helium pycnometer or a dew point hygrometer), measurements of grain density, carbonate content or suction are recommended to provide extra information for analysing the results. Andra also has a geological model which allows estimating mineralogical composition of the sampled cores. This model is based on numerous measurements made since the start of research on COx claystones.

4.2.4. Initial damage

Tomography is highly recommended to understand the initial condition of the sample. Within the limits of the device precision, it can be used to eliminate damaged samples before the start of testing. In this case, the accuracy of the analysis and the size of cracks detected should be provided.

It is recommended that all necessary samples are taken immediately upon opening the T1 cell. For all THM tests, when the laboratory has suitable equipment, deformations and the interstitial pressure should be measured. Samples that are significantly desaturated ($S_r < 90\%$) should not be used for characterisation.

5. Representativeness of sample measurements in relation to their in situ conditions

One of the objectives of the study of COx claystones is to establish rheological behaviour models that are able to reproduce their HM evolution during and after the excavation under total or partial saturation. COx claystones are characterised in the laboratory (deformation modulus, compressive strength, tensile strength, etc.) using conventional triaxial or uniaxial compression tests and indirect tensile strength tests. These tests are complemented by the in situ tests carried out in undisturbed claystones at the MHM URL. Results for both deformation modulus and compressive strength are naturally widely dispersed. However, different trends are observed between laboratories results, which go beyond the natural dispersal and it is necessary for Andra to identify them. Previous sections of this paper have shown the importance of the precautions to be taken during sampling; however, despite these precautions, it is impossible to obtain completely saturated samples. Nevertheless, in order to characterise the claystones in the undisturbed environment, high degrees of water saturation remained in the samples are required. Their HM properties and behaviours can be directly transferred to the natural rock mass. The question arises whether samples should be resaturated before testing in order to get closer to the natural conditions. The impact of the degree of saturation on the mechanical characteristics of COx claystones is discussed below.

During the THM characterisation of the Teguline clays, Zeng et al. (2016) studied the effects of samples resaturation on their

microstructure. Monitoring swelling over time for several values of the water content shows the change in microstructure. The initial degree of saturation of sample can be estimated from its water content and porosity. According to Zeng et al. (2016), there is a 'desaturation threshold' (see Fig. 7) above which the mechanical parameters are not influenced by the variations in saturation. Above this threshold value, the measured mechanical parameters reflect only natural variability and can be considered representative of the undisturbed material.

Several authors have reported that resaturation of COx claystone samples could lead to degradation of their mechanical properties. Miehe (2004) showed that taking into account the HM coupling does not explain the decrease in the properties observed on some resaturated samples. Regardless of the confinement pressure applied to the sample, swelling is always observed during the resaturation phase, reflecting a modification of the structure of the material.

In 2008, Andra conducted a deep borehole drilling campaign (EST413, EST423, EST433 and EST441) to determine the vertical and lateral variability of HM properties, including permeability, deformation modulus and compressive strength. The samples were not resaturated. The measurements of water content and porosity indicate that the remaining water saturation in the samples varied between 80% and 100%. Authors such as Guery (2007) observed the influence of certain physical parameters on the THM behaviours of COx claystones. Analysis of the results using a multi-parameter linear correlation confirms this. In Fig. 8a, the Young's modulus values obtained from samples in borehole EST423 are plotted as a function of depth. These values are compared to the Young's modulus values estimated using a linear regression based on the sample's mineral composition and porosity. There is a fairly good correlation, which is also demonstrated on the compressive strengths.

In Fig. 8b, the same measurements obtained in the set of deep boreholes are represented as a function of the degree of saturation of the samples; no direct correlation is observed in this range of degrees of saturation. This is consistent with Zeng's observations (Zeng et al., 2016), which shows that at this saturation level, natural variability has a much greater impact. Fig. 8b also shows the Young's modulus values obtained by Zhang (2011) by uniaxial compression tests on some artificially resaturated samples. These

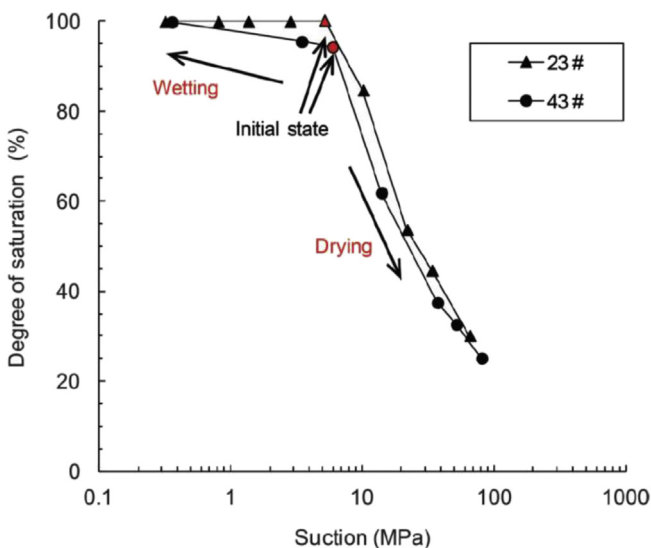
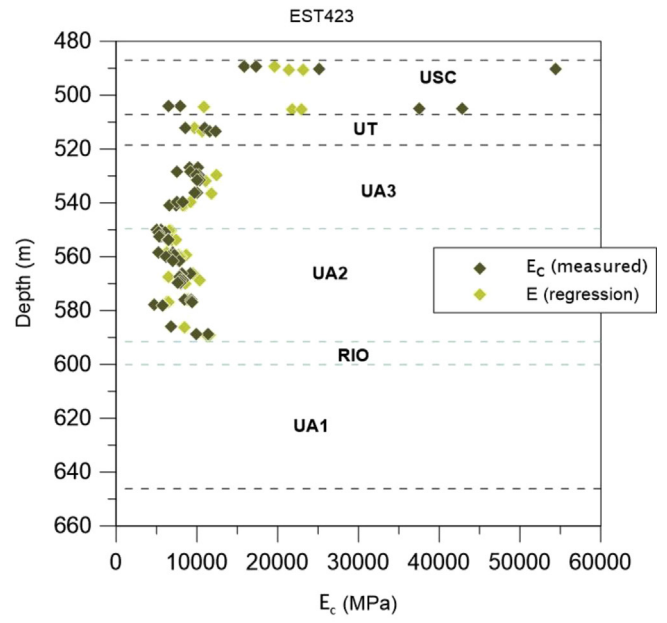


Fig. 7. Change in degree of saturation with suction of Teguline clays during drying and wetting (Zeng et al., 2016).



(a)

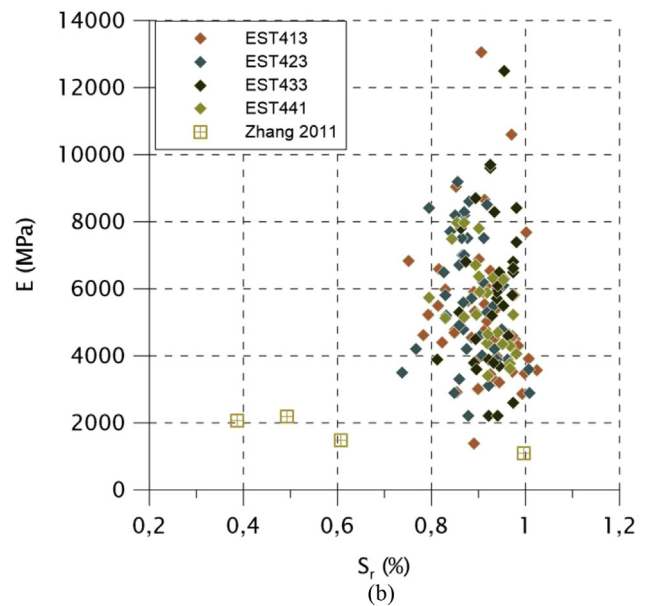


Fig. 8. Change in Young's modulus as a function of mineralogy in borehole EST423 (a) and the degree of saturation (b) of samples taken from deep boreholes.

modulus values are very low with a clear decrease as a result of the total sample resaturation. This effect is also observed by Wang (2013) who, using image correlation, showed that the artificial changes in samples saturation cause microcracks to appear in the material along with the changes in the material mechanical characteristics.

To illustrate this, percolation tests with reconstituted site water were carried out in a Peek (PolyEtherEtherKetone) triaxial cell using X-ray tomography. The applied confining pressure was 12 MPa. Images were obtained periodically: before confinement, after confinement, during the water injection phase, during the unloading stages and then at the end of the test (see Fig. 9). Continuous measurements of water flows were used to assess the

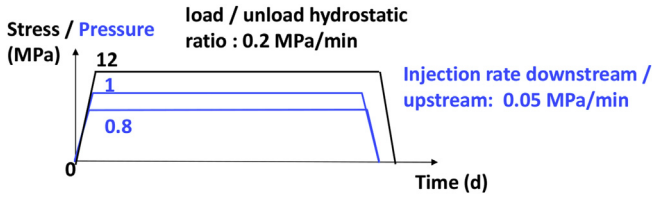


Fig. 9. Schematic of hydraulic load tests under tomography.

permeability of the material during the test. Fig. 10 shows the frontal and cross-sections of the sample at a resolution of 24 μm per pixel. Before confinement, the sample appears perfectly undisturbed. The second tomography was carried out after 28 d at the end of hydraulic loading; again, no cracks were found. The injected water pressure is indicated in Fig. 9. Two unloading stages were then applied from 12 MPa to 8 MPa and then from 8 MPa to 4 MPa before the total discharge. The images of the internal sections show the appearance of a crack at 8 MPa stage. The unloading to 4 MPa and then total unloading amplify the phenomenon: several new cracks appear and the opening of these increases. The three-dimensional (3D) representations (see Fig. 11) before and after the test allow clearly visualising the size of the crack compared to the sample. It was observed at the end of the tests that the sample was extremely degraded. The inverse analysis performed from the permeability measurement yields a deformation modulus of about 500 kPa instead of 5000 MPa that was conventionally measured on undisturbed sample. This study confirms that percolating water through claystones is not neutral for the sample even when confinement is applied. On the other hand, the water permeability value measured during this test is within the range of values obtained for the undisturbed COx claystones, i.e. around 10^{-20} m^2 .

This section shows that, apart from the difficulty of resaturating claystone samples due to their low permeability, resaturation can have a negative effect on the HM properties of the materials. Methods used for resaturation can never restore the ‘natural’

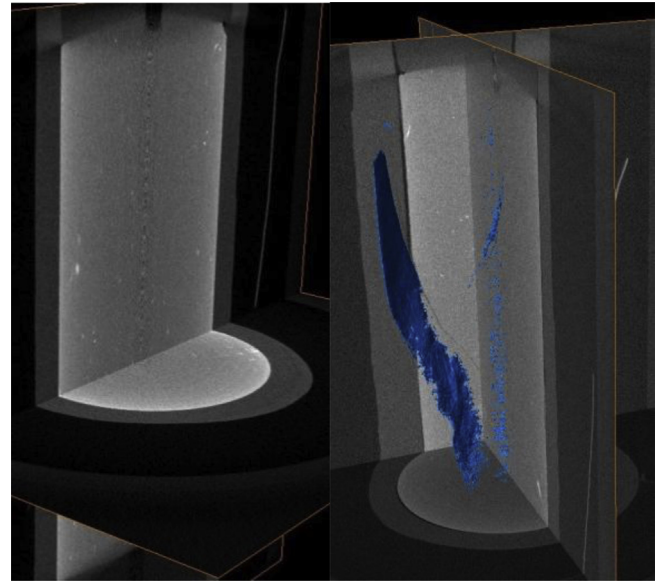


Fig. 11. 3D representation of the sample before and after testing; the blue volume represents the main fracture.

condition of the samples. The choice has therefore been made for the mechanical measurements, to use only properly preserved samples whose water saturation remains high enough, i.e. above 90%. This requires a highly controlled sampling process and conservation chain in order to preserve the material structure.

6. Conclusions

COx claystones show a natural variability due to their mineralogical compositions. This variability must be taken into account in the analysis of HM characterisation tests. However, it is important to distinguish this variability to that related to the sample

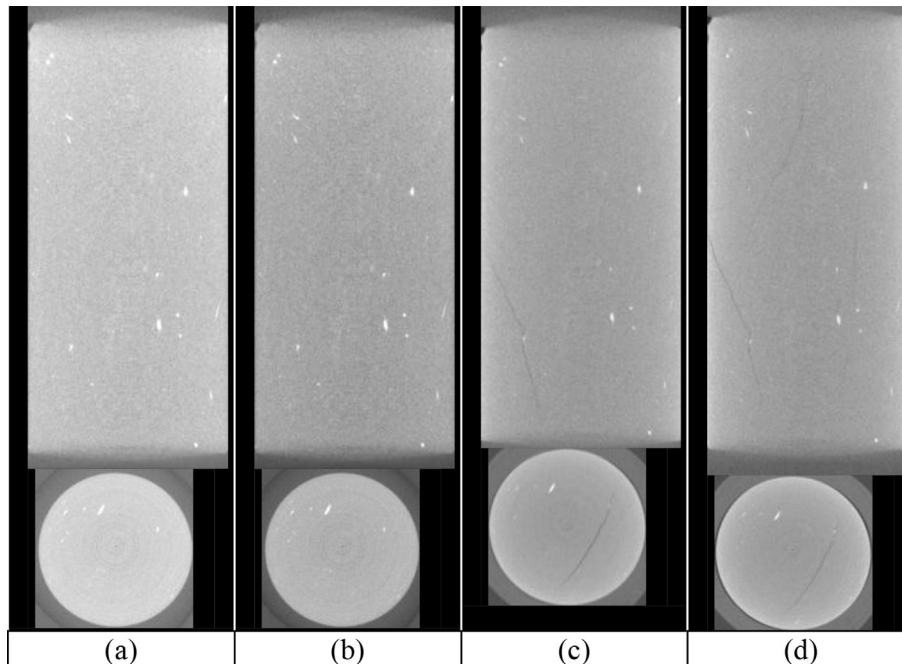


Fig. 10. Cross-sections along the XZ and XY axes, resolution 24 μm per pixel (a) before hydraulic loading, (b) after hydraulic loading, (c) during mechanical unloading at 4 MPa, and (d) after total unloading.

condition. Andra has optimised and improved the sampling method from borehole extraction to testing in order to deliver the most representative samples for characterising the properties and response of the natural clay rock.

The samples preparation for laboratory tests may be accompanied by damage, which is unfortunately difficult to be assessed. The initial deconfinement, the conservation of the material, the desiccation, and the production of the samples from the T1 cells are all 'traumatic' elements, which can induce microcracking. In order to better control all these effects and to be able to understand them, it is important to have the sampling history from the core extraction to the realisation of the laboratory tests. An improved sampling procedure (with extraction and conditioning) as well as testing protocols is proposed to ensure the high quality of the samples.

The time from core extraction to confinement in cells has to be shortened within an hour to avoid desaturation and relaxation of stresses. Preserving desiccation by adding a new protective layer (self-adhesive aluminium foil) between the core and the latex sock has also contributed to improving the seal function. The production of the total set of samples should be implemented as soon as the T1 cell is opened. The degrees of water saturation in the samples are to be kept above 90%. The variation of mechanical characteristics with the degree of saturation of highly saturated samples is lower compared to that associated with the variability of the mineralogical composition. By means of sophisticated imaging methods, it shows that the artificial resaturation of the samples leads to alteration of the microstructure and degradation of the strength. For an optimised characterisation, this study shows that HM tests must be carried out on the least desaturated samples. The development or improvement of sampling methods is still on-going in the Andra's research programme, in particular, to fix a better method of resaturation which does not damage the sample.

Conflicts of interest

The authors wish to confirm that there are no known conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome.

Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.jrmge.2018.02.004>.

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