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Stress release and suction generation in the Callovo-Oxfordian claystone

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Abstract. Stress release effects in a specimen of claystone extracted at great depth (465 m) in the layer of the Callovo-Oxfordian claystone in Eastern France is examined by mimicking the stress change during extraction in the oedometer. To do so, a specimen first submitted to hydration with swelling impeded is afterwards unloaded at constant water content. Comparable mercury intrusion pore size distribution curves are obtained before and after stress release, showing little change in the configuration of inter-platelets pores within the clay matrix of the claystone. The swelling observed at constant water content is then related, to a water transfer from inter-platelets pores to intra-platelets pores, resulting in crystalline swelling within the smectite fraction of the inter-layer illite-smectite minerals.

1 Introduction

The Callovo-Oxfordian (COx) claystone is considered as a possible host rock for deep geological disposal of radioactive waste in France. Many investigations on its hydromechanical behaviour are carried in the Underground Research Laboratory (URL) managed by Andra (the French Agency for radioactive waste disposal) close to the village of Bure (Eastern France). The URL has been excavated at a depth of 490 m in a 150 m thick layer COx claystone.

Prior to be tested in surface laboratory, claystone specimens have to be cored and extracted from the claystone mass. It is important to figure out how far the specimens tested in the laboratory are representative of the in-situ intact state, prior to extraction. The effect of stress release is quite significant at depths as high as 490 m. Given that COx claystones specimens provided to surface laboratory are generally unsaturated, particular attention has been paid to ensure fully saturation under stress conditions close to in-situ and full drainage conditions during compression tests. In low permeability rocks (the saturated permeability is in the order of 10^{-13} m/s in the COx claystone), saturation can be quite long and pore pressure homogeneity during testing can be problematic. In this regards, specific devices with reduced length of drainage have been developed (Monfared et al., 1, Hu et al., 2).

Desaturation of specimens of the COx claystone also occur in-situ in the gallery walls due to ventilation. This will not be considered here and only the effects of stress release will be considered. It is well known that saturated clay specimens carefully extracted at standard geotechnical depths (various tens of meters) exhibit suction (Skempton and Sowa, 3, Bishop et al., 4, Doran et al., 5, Delage et al., 6). In the case of the "perfect sampling" of an isotropic elastic sample and provided the sample remain fully saturated once extracted, this suction is supposed to be equal to the mean effective stress the specimen was submitted to prior to extraction (4). A necessary condition for the sample to remain saturated is that its air entry value should not exceed the mean effective in-situ stress. In the case of the Boom clay, suction effects have been investigated by (6) who found that specimens extracted at a depth of 225 m in the URL of Mol (Belgium) exhibited a suction value between 2 and 3 MPa, close to the mean in-situ effective stress (2.12 MPa). This work also confirmed that swelling clays to be tested in the triaxial apparatus should not be put in contact in water without previously being submitted to a mean effective stress as close as possible to that existing in-situ to avoid swelling and damage that would degrade the mechanical response and reduce the volumetric yield stress.

Compared to clays, claystones have been consolidated and strengthened during very long diagenesis periods during much longer geologic history, resulting in significantly lower porosity, strong diagenesis bonds and higher mechanical resistance. Following the previous investigation in the Boom clay, suction effects were investigated in the Callovo-Oxfordian (COx) claystone by testing specimens extracted at a depth of 479 m in the Bure URL in Eastern France. Investigation of the effects of desaturation in claystones (Pham et al. ,7, Jougnot et al., 8, and Wan et al., 9) is of some interest due to the consequences of the ventilation of the galleries along the gallery walls.

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In this paper, one examines what happens during stress release at constant water content on a specimen of COx claystone. To do so, a COx specimen was submitted to hydration at constant volume in the oedometer and stress release was imposed at constant water content. The change in microstructure were investigated by using mercury intrusion porosimetry.

2 Experimental methods

Tests were carried out in a room at controlled temperature and relative humidity (20°C and 80% respectively). Following Mohajerani et al. (10), the saturation of the sample was conducted on a high stress double lever arm oedometer (Marcial et al., 11) by carefully limiting volume changes during water infiltration. To do so, the vertical displacement gauge was manually followed and the specimen was progressively loaded to compensate any swelling with a limit fixed at 2 µm.

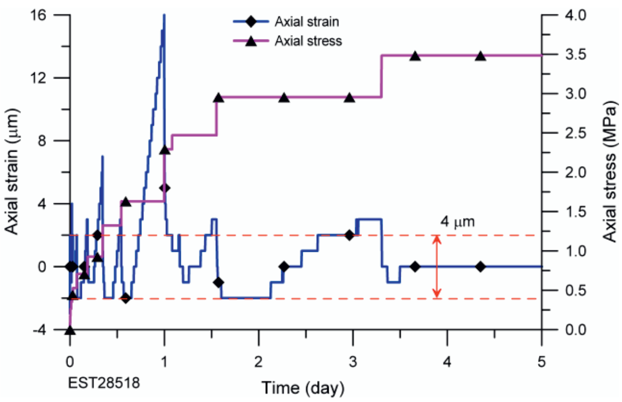

Figure 1. Resaturation with limited swelling

The detail of the procedure is illustrated in **Figure 1** that shows that the maximum stress applied to neutralize the swelling was equal to 3.48 MPa for a sample with an initial degree of saturation (S_r) of 77% and a suction of 34 MPa (WP4 measurement). The relatively high displacement of 16 □m observed at the end of first day is due to imperfect control during night time. To investigate suction effects on such a specimen resaturated with limited volume change, the specimen was afterwards quickly unloaded at constant water content. To do so, the porous stones were first desaturated by air flushing.

Pore-size distribution measurements (Mercury Porosimeter micromeritics-AutoPore IV 9500**)** were conducted on freeze-dried specimens to investigate microstructure changes. Specimens were previously frozen at - 210°C by using slush nitrogen (obtained by previously putting liquid nitrogen under vacuum, Delage et al. 2006, 12). The range of pores investigated was between 363.6 µm (minimum pressure of 3.4 kPa) to 5.5 nm (maximum pressure of 227.5 MPa).

3 The Callovo-Oxfordian claystone

The COx claystone is a sedimentary rock deposited 155 millions years ago between the Callovian and the Oxfordian ages in the western area of the Parisian sedimentary basin on top of a layer of Dogger limestone. The claystone was afterwards covered by an Oxfordian limestone layer. It has been since that time submitted to various tectonic effects including some horizontal stresses resulting from the Alpine orogenese. The COx claystone is composed of a clay matrix containing some detritic grains of quartz and calcite. At the depth of the Bure URL (490 m), the average mineralogical composition of COx claystone is as follows (Gaucher et al., 13): 45-50% clay fraction (mainly interstratified minerals of illite–smectites with a fraction between 50 and 70% of smectites, 20% carbonate, 22% quartz and 9% other minerals (feldspars, pyrite, dolomite and siderite). The total porosity varies in the COx layer between 14% in carbonated levels and 19.5% in the more argillaceous levels (Yven et al., 14).

The stress state measured in the 490 m deep Bure URL is as follows (Wileveau et al., 15):

- σ_v = 12.7 MPa
- σ_h = 12.4 MPa
- σ_H = 12.7 – 14.8 MPa
- u = 4.9 MPa

The vertical stress σ_v is close to the minor horizontal stress □h and smaller than the major horizontal stress □H. For this reason and some others (Mohajerani et al., 10), usual concepts of preconsolidation valid in clays do not apply to the COx claystone. Based in the above values, the equivalent effective Terzaghi mean stress:

$p'=1/3\ (\sigma'_v + 2\ \sigma'_h)$

is between 7.7 and 8.4 MPa (with $\sigma'_i = \sigma_i - u$).

Table 1 provides a comparison between the geotechnical properties of the Boom clay (at a depth of 225 m) and that of the COx clay (at a depth of 479 m with maximal clay fraction of 48-50%, Gaucher et al., 13).

Table 1. Geotechnical characteristics of the Boom clay and COx claystone

	Boom clay	Callovo-Oxfordian claystone
Geology	Oligocene (Rupelian)	Jurassic (Callovo-Oxfordian)
Age	30 millions years	160 millions years
Depth	225 m	479 m
Water content *w* (%)	20-30	< 6.5
Porosity *n* (%)	35-40	14-19
Clay fraction (%)	40-70	48-50
Young's modulus (GPa)	0.2-0.4	3.6-8.5
Initial specimen suction	2.8 MPa[1]	34 MPa[2]

[1] *Measured by the filter paper method*
[2] *Measured by the Decagon dew-point tensiometer*

The differences in water content (w), Young's modulus and unconfined shear strength illustrate the significant difference between a stiff clay (Boom clay) and a claystone (COx).

Further understanding about the microstructure, mineralogy and porosity of the COx claystone was gained by Yven et al. (14) by using different methods (including scanning electron microscope, autoradiography, mercury intrusion porosimetry, oil, helium and nitrogen adsorption), resulting in the conceptual model presented in Figure 4. The model shows how individual calcite or quartz grains are contained into a clay matrix with interconnected porosity.

Figure 2 presents a typical pore size distribution curve of the COx claystone obtained by mercury intrusion porosimetry (MIP) on a freeze-dried specimen, showing a well defined single pore population (with an average diameter of 32 nm) that is typical of the clay matrix.

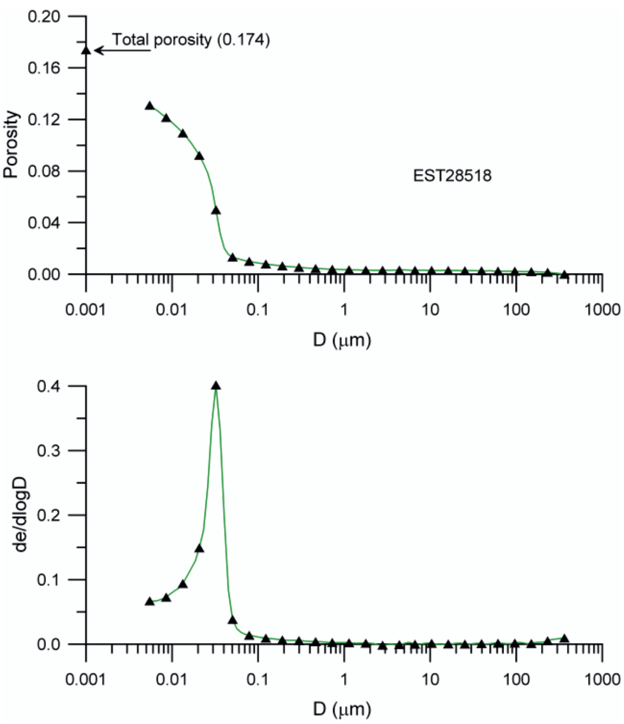

Figure 2. Pore size distribution curve initial state

The intruded porosity represents 75% of the total porosity (n = 17.4% and also represented in the graph). This data is in good agreement with that of Yven et al., (14) who estimated the porosity of small dimension (3 - 10) to 33.5 % of the total porosity in accordance with the 34% of porosity smaller than 10 nm found here.

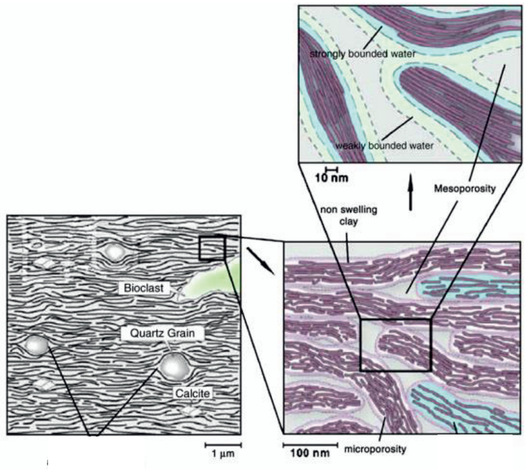

Figure 3. Microstructure model (after Yven et al., 14)

The remaining porosity is located within the clay matrix in smaller pores that cannot be investigated by MIP. This is a well known feature in clay materials containing smectites, due to very thin intra-platelets (or intra-crystalline) porosity (see Delage et al., 12 in compacted smectites).

The interpretation of the single pore population observed in Figure 2 can be made by assimilating the clay matrix to an assembly of bricks made up of platelets, as can be seen in Figure 3. The mean pore radius hence provides an estimate of the average platelets thickness. With an inter-basal spacing of 9.6 Å (0.96 nm) typical of dry smectites and illites and an average platelet thickness of 20 nm corresponding to the average entrance diameter determined from the PSD curve of the dried specimen, an average number of 21 layers by platelets can be roughly estimated.

4 Results

The stress-strain path corresponding to the instantaneous constant water unloading phase from the 3.48 MPa vertical stress is represented in Figure 4.

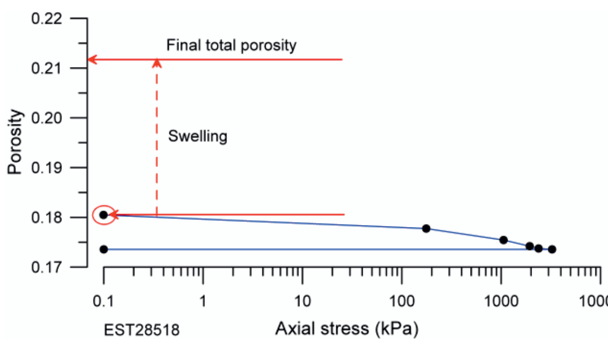

Figure 4. Unloading phase at constant water content

Unloading was performed stepwise at intermediate stresses of 0.8 MPa (in the semi-log graph, complete unloading is arbitrarily represented at a stress of 0.1 kPa). The unloading process lasted 5 minutes and exhibited an increase in porosity from 17.4 to 18% calculated from the swelling observed in Figure 4. The sample was afterwards extracted from the oedometer cell allowing for (slight) radial and axial expansion as shown in Table 2.

Table 2. Characteristics of the tested specimen

	Before test	After test
Height *h* (mm)	8.66	9.19
Diameter *d* (mm)	37.92	38.06
Mass *m* (g)	23.75	24.4
Water content *w* (%)	6.3	8.4
Void ratio *e*	0.21	0.27
Porosity *n*	17.4	21.2
Degree of saturation S_r	77	81
Suction *s* (MPa)	34	21

Careful calliper measurements carried out 15 minutes after unloading sample provided a porosity value of 20.9 %. Obviously, significant axial swelling occurred at constant water content once the sample was extracted from the ring. Note that hydrostatic weighing provided a porosity value of 21.2 % quite close to that obtained by calliper measurements, also represented in Figure 4. The specimen was afterwards isolated from evaporation by wrapping in a plastic film and rapidly frozen to keep constant water content.

One can observe that, in spite of having a constant water content (w = 8.4%), the swelling observed (final porosity n = 21.2% compared to 17.4%) resulted in a simultaneous desaturation of the specimen with a final value of the degree of saturation S_r = 81% corresponding to a 21 MPa suction (WP4 measurement). This means that the air-entry value of the sample has been exceeded. Indeed, this value, estimated between 7 and 9 MPa according to gas penetration tests (De La Vaissière and Talandier, 16) is significantly smaller than the final value of 21 MPa.

The pore size distribution (PSD) curve of the swollen sample is presented in Figure 5. Also reported in the graph is the total porosity (21.2%).

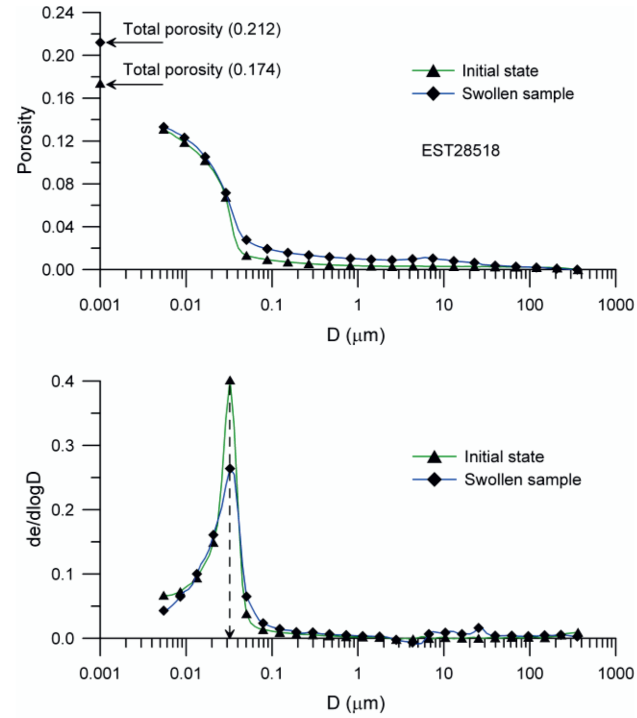

Figure 5. PSD curve of the swollen specimen

Surprisingly, the shape of the PSD of the swollen unsaturated specimen at constant water content is quite comparable to the initial one presented in Figure 2. It exhibits a single pore population with a mean radius of 32 nm, comparable to that of the intact sample (although slightly less well classified as indicated by the density function curve). No large pore population representing possible cracks due to hydraulic damage and swelling (Yang et al., 17, Wan et al., 9) is observed. Also, visual examination of the sample didn't evidence any visible cracks with thickness larger than the maximum diameter identified by mercury intrusion (363.6 µm).

5 Discussion

The global specimen expansion at constant water content is then only possible within the intra-platelet infra-porosity (too small to be intruded by mercury) with a transfer of water from the larger inter-platelets pores (average 32 nm) to the smaller intra-platelet pores that results in crystalline swelling. Here, swelling occurs within the inter-stratified illite smectite clay platelets.

As any clay material, the pore water of the COx clay includes both capillary and hydration water (e.g. Revil and Lu, 18). By only considering capillary effects, the suction s exerted when desaturating 32 nm diameter pores is given by the Young-Laplace equation:

$$s = \frac{2\sigma_{w/a}\cos\theta}{r} \tag{1}$$

in which $\sigma_{w/a}$ is the water/air interfacial tension equal to 72.8×10^{-3} N/m and θ the solid-liquid contact angle with $\cos\theta = 1$ for water. A pressure value of 9.1 MPa is obtained, slightly higher than the air-entry value of the

COx claystone, confirming the predominance of capillary effects in the clay water interaction in these inter-platelets pores schematically represented in Figure 3.

Comparing this value with the final suction of 21 MPa finally measured in the sample indicates that inter-platelets pores have been desaturated. The high suction value is hence mainly due to strong clay-water interaction occurring inside the platelets, confirming the intra-crystalline nature of the swelling.

The question about the rate of water transmission between the large inter-platelets pores and the thin intra-platelets ones arises given the very low permeability and slow water transfer suspected. This internal water transfer, comparable in nature to long term internal transfers observed in compacted bentonite at constant water content (Delage et al., 12) has been completed in around 30 minutes after unloading. Actually, the slowing effects due to the low permeability (around 10^{-20} m^2 in the COx claystone) are compensated by the small distance that water molecules have to travel from the inter-platelets pore to the intra-platelets ones.

6 Conclusion

Suction effects were investigated in a specimen of the Callovo-Oxfordian claystone extracted at great depth (479 m) in the underground research laboratory of ANDRA in Bure (France). The specimen was first saturated with limited volume change by progressively increasing vertical stress up to 3.48 MPa. The specimen was afterwards quickly unloaded at constant water content, exhibiting significant swelling and desaturation, 30 minutes after extraction from the oedometer ring with a suction significantly larger than the air entry value. The pore size distribution of the swollen sample was comparable to that of the initial one with no appearance of large pores, indicating that swelling was due to a water transfer within the clay matrix from inter-platelets pores to intra-platelet pores, in a pore range that is too small to be detected by mercury intrusion. This phenomenon may occur during gallery excavation in areas of the excavation damaged zone submitted to instantaneous stress release, as a first stage of desaturation, prior to the subsequent evaporation due to the ventilation of the galleries.

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