Modelling of oedometric compression and swelling behaviour of the Callovo-Oxfordien claystone

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Abstract. Callovo-Oxfordien (COx) claystone, as a potential host rock for nuclear waste disposal, is investigated for its oedometric compression and swelling behaviour in this paper. Based on results of high pressure oedometric compression experiments, a model is proposed to explain the swelling behaviour of COx claystone under different stress. A method using the variation of porosity is also proposed to determine the damage of rock quantitatively. Mercury Intrusion Porosimetry (MIP) and Scanning Electron Microscopy (SEM) were applied in order to get a clear map of the swelling process in micro scale.

1. Introduction

Annul rising of high activity radioactive waste leads to the need to find the proper sites with a stable rock structure where used nuclear fuel could be stored safely without any significant leak in the long term. Callovo-Oxfordien (COx) claystone of the Meuse\Haute-Marne region, is supposed as a potential host rock due to its low hydraulic conductivity, compressibility, diffusion coefficient and high sorption capacity for radionuclide. The depth of COx claystone is between 420m and 550m, and the underground laboratory of ANDRA (the French Agency for Radioactive Waste Management) is located at the depth of 445m and 490m.

Requested by ANDRA, a series of experiments have been done in order to investigate the physical, hydraulic, and thermal behaviours of COx claystone. In this article, oedometric compression and swelling behaviour of the COx claystone is investigated and a new model is proposed based on the experimental results of Mohajerani (2011) and Hamza (2013). This model tries to bridge the variation of microstructure and macro behaviour, aiding by Mercury Intrusion Porosimetry (MIP) and Scanning Electron Microscopy (SEM).

2. Materials and methods

The tested COx claystones originate from the cores in the boreholes EST28518 and EST28522 at depths of 490 m and 481 m, respectively. The characteristics of tested samples are listed in Table 1.

| Samples       | Water content (%) | Degree of saturation (%) | Porosity (%) | Suction(MPa) | Sample height(mm) |
|---------------|-------------------|--------------------------|--------------|--------------|-------------------|
| EST28518 n°1  | 6.46              | 77                       | 17.4         | 34           | 13.02             |
| EST28518 n°2  | 7.1               | 68                       | 22           | 34           | 13.55             |
| EST28522 n°1  | 6.1               | 56                       | 22           | 17           | 10                |
| EST28522 n°2  | 6.1               | 56                       | 22           | 32           | 10                |
The mineralogical composition of COx claystone is determined by X-ray diffraction method. Clay minerals, quartz and calcite constitute nearly 95% of the rock, with their relative proportion varying with depth. For EST28518, clay minerals represent 45% of the sample with nearly 60% interstratified illite-smectite, 30% illite and a little percent kaolinite. Clay minerals, especially smectite, expanding considerably due to the lamellar structure, is the key for the swelling behaviour of claystone. 28% calcite and 23% quartz are also contained in the claystone, while feldspars, pyrite and iron oxides form the rest.

The no-wetting property of mercury make it possible to find out the distribution of the connected porosity with Mercury Intrusion Porosimetry technique. The pore size could be determined by mercury pressure with Jurin’s law as following:

\[ p = \frac{2\sigma \cos \theta}{r} \]  \hspace{1cm} (1)

Where \( \sigma \) is the surface tension for mercury, equalling to 486.5 mN/m at 20°C and \( \theta \) is the contact angle of mercury environ 140°, \( r \) is the pore throat radius.

The measured range of pressure is from 0.001 to 400 MPa, corresponding to the pore diameter from 6 nm to 360 μm. The total porosimetry is measured by hydrostatic weighing, basing on Archimedes' principle. The total volume of sample is determined by the immersed water and the volume of grain is obtained by the ratio of dry mass and grain density. With these two values obtained, it is easy to obtain the total porosimetry.

WP4 dewpoint potentiometer, produced by Decagon Devices, is used to measure suction, by determining the relative humidity of the air above a sample in a closed chamber. Its range is from 0 to 300 MPa with a margin of 0.1 MPa.

Rock samples desaturate during the collecting and preserving process, thus, resaturation of rock should be done by infiltrating water under pressure into porous discs before the tests. The resaturation leads to swelling, so a load should be applied in order to remain the volume of sample. As presented in Mohajerani (2011), a series of doubled loads is applied to avoid vertical displacement reaching 2 μm. According to Mohajerani (2011) and Hamza (2013), swelling dominates the deformation until 3.5 MPa pressure is applied.

3. Experimental program

Oedometric experiments were carried out with a high pressure oedometer developed at Navier/CERMES (Marcial et al. 2000) and the results are presented in Mohajerani et al. (2011). For sample EST28522 n°2, it was firstly loaded to 28 MPa; then unloaded to 0.4MPa; after the unloading, reloaded to 113MPa; finally, unloaded to zero (Seen as Figure.1). For sample EST28518 n°1 and EST28518 n°2, they were loaded and then unloaded to zero, with a maximal stress 56 MPa and 113 MPa, respectively.

![Figure 1 Oedometric compression curve of EST28522 n°2 (left), EST28518 n°1 and EST28518 n°2 (right)](image-url)
Before and after the experiment, MIP tests were made to investigate the evolution of porosity of samples, and microstructure of sample was also observed under SEM. For both of them, before measurement and observation, samples need to be frozen to -210 °C with nitrogen so as to avoid the derangement of microstructure during the preparation.

4. Results of tests
The pore size distribution of intact sample, EST28518 n°1 and EST28518 n°2 is presented in Fig. 2. According to this figure, the peak of pore size for these three is around 20nm, and the sample that suffered higher stress has a lower peak and a wider dispersal. Meanwhile, the samples after oedometric compression test have another peak environ 30 μm. As there is nearly no pore larger than 1 μm for intact sample, we can say that the pore larger than 1 μm propagate during the cycle of loading-unloading.

![Figure 2 Pore size distribution of intact sample, EST28518 n°1 and EST28518 n°2](image)

Although the pore distribution is different, cumulative porosity curve (Fig.3) shows that the total volume of pores with the diameter smaller than 1 μm doesn’t change much. Precisely, the porosity under 1 μm for intact sample is 12.75%, while those of EST28518 n°1 and EST28518 n°2 are respectively 12.70% and 12.19%. For intact sample, pores larger than 1 μm are negligible, constituting only 0.35% volume of the rock. Due to the appearance of cracks, the volume of pores larger than 1 μm of sample EST28518 n°1 and EST28518 n°2 increase to 3.55% and 5.96%. And this augmentation is same as the augment of total porosity measured by hydrostatic weighing method. Therefore, although knowing no detail about the pore size distribution under 3 nm, we can suppose these pores do not vary with the applying of loads.

According to the results of test EST28522 n°2, EST28518 n°1 and EST28518 n°2, the loading curve is nearly linear, which shows that 113 MPa is beyond the elastic limit of COx claystone, whereas the unloading curve is non-linear. It is remarkable that the sample swells significantly at each unloading.
step and finally its volume comes back to or even bigger than the initial state. Therefore, a simple conclusion could be drawn that swelling behavior of COx claystone doesn’t play an important role in strain variation during the loading phase, while it dominates strain variation during the unloading phase.

As seen in Figure 4, although the loading-unloading procedure and maximal loading are different, these samples exhibit a similar behavior during the loading and unloading phases.

![Figure 4 ε/ε<sub>max</sub> - σ/σ<sub>max</sub> curves](image)

**Figure 4 ε/ε<sub>max</sub> - σ/σ<sub>max</sub> curves**

![Swelling of samples under axial stress](image)

**Figure 5 Swelling of samples under axial stress**

Before investigating the swelling behavior, elastic deformation and real swelling should be distinguished. Supposing that swelling at high pressure is negligible, elastic unloading moduli E could be calculated using the first loading step. Here the value is 4.5 GPa and 7.8 GPa for EST28518 n°1 and EST28518 n°2, respectively. Another hypothesis that elastic unloading moduli is constant during the whole unloading process should be added. A semi-log linear relationship between axial stress and swelling is presented as Fig.5. As seen in the figure, COx claystone swells much more at a lower stress and the one suffered higher maximal stress exhibits more significant swelling trend.

5. Modelling

5.1 Determination of damage variable D
Obviously swelling behaviour has a certain relationship with the damage of samples, which is presented by a damage variable D. Defined as the ratio between the surface intersections of the micro-cracks and overall section surface (Kachanov, 1958; Lemaitre, 1971), D is commonly used in the continuous damage mechanics to describe the degradation of the material during the loading phase. Even with this definition, it is difficult to obtain the exact value of D due to the lack of quantitative information about the micro-cracks.

Under limited stress, the rock matrix doesn’t break and its deformation is negligible compared with that of pores. So the porosity variation offers us a clue to the evolution of micro-cracks and to estimate the value of D. If we suppose that micro-crack doesn’t propagate during the unloading phase, thus, D at unloading process is the same value as that at the final of test. Limited by the measure range of MIP, the distribution of pore size < 3nm is unknown. However, because the difference between the total porosity measured by Hydrostatic Weighing and porosity measured by MIP is very close in the intact and damaged situation, it is reasonable to presume that the pores under 3nm don’t change during the whole loading-unloading process, which means that those pores make no influence on the variation of D.

The following expression is defined to quantify the damage of rock:

$$D = 1 - \frac{\Phi_{\text{intact}}}{\Phi_{\text{damaged}}}$$

Where $\Phi_{\text{intact}}$ is the porosity of intact sample and $\Phi_{\text{damaged}}$ is the porosity of sample after loading-unloading process. Both of them are the values measured by MIP. The values of porosity and D in different samples are listed in the Tab. 2.

| Sample       | Porosity | Mercury Intrusion Porosimetry | Hydrostatic Weighing | D   |
|--------------|----------|-------------------------------|----------------------|-----|
| Intact       | 13.1     | 17.4                          |                      | 0   |
| EST28518 n°1 | 16.2     | 20.4                          |                      | 0.191|
| EST28518 n°2 | 18.1     | 22.2                          |                      | 0.273|

According to the definition of D, (1-D) represents the effective resisting area in unit section, effective stress is expressed as following:

$$\sigma = \sigma/(1-D)$$

Where $\sigma$ is axial stress during oedometric test, $\sigma_{\text{eff}}$ is effective stress.

Therefore, the relationship between elastic unloading moduli and effective elastic unloading moduli is obtained:

$$E = E_{\text{eff}}(1-D)$$

Where $E_{\text{eff}}$ is effective elastic unloading moduli.

### 5.2 Normalized swelling and normalized stress

As mentioned above, swelling varies inversely with the axial stress, and is more significant after suffering a higher stress. Naturally the following equation is developed:

$$\frac{\lambda}{\sigma} \sigma_{\text{max}} d\sigma = E_{\text{eff}} d\varepsilon$$

Where $\lambda$ is a parameter which presents the swelling ability of rock, $\varepsilon$ is swelling, and $\sigma_{\text{max}}$ is the maximal stress in its stress history.

Integrating both sides of Equation 5, we can find the relationship between normalized swelling and normalized stress.

$$\frac{2.303 \lambda \sigma_{\text{max}}}{E_{\text{eff}}} \log \left( \frac{\sigma}{\sigma_{\text{max}}} \right) = \frac{\varepsilon}{D}$$

Plotting the normalized swelling and normalized stress, we could obviously observe that the points are located in the same line with a slope $C_s = -0.062$ (Seen in Fig.6).
According to Eq. 6,

\[ \frac{2.303\lambda \sigma_{\text{max}}}{E_{\text{eff}}} = C_s = -0.062 \]  

(7)

\[ E_{\text{eff}} = 37.1\lambda \sigma_{\text{max}} \]  

(8)

It is logical that \( E_{\text{eff}} \) varies with the applied maximal stress, and it corresponds with our hypothesis that elastic unloading moduli is constant during the whole unloading process. Combining Eq. 7 and Eq. 8, for EST28518 n°1 and EST28518 n°2, we can obtain the same value of \( \lambda \), equal to 2.68.

For EST28522 n°2, the porosity after the loading-unloading test is unknown. Supposing that D of EST28522 n°2 after 113MPa loading is the same value as that of EST28518 n°2, we could get normalized swelling and normalized stress and also find that these points are located at the same line as EST28518.

6. Conclusion
In this paper, two parts of works about some characteristics of hydro-mechanic behaviour COx Claystone are presented. In the experimental part, suction controlled tests and MIP test were conducted on specimens of the COx claystone to investigate the water retention properties and the influence of
suction on the pore size distribution. In the theoretical part, the coupling between swelling and damage of COx claystone under stress was analyzed and a model was proposed based on the analysis of the evolution of microstructure during the loading-unloading cycle.

The results of suction controlled tests show a good correspondence and demonstrate a good repeatability of the method. The water retention curve corresponds well with the results of Wan et al. (2013). Along the drying path at higher suction, a linear relationship is observed between the degree of saturation and the water content. COx claystone exhibits a significantly different behaviour above and below a crucial point with suction of 11 MPa and water content of 7%.

The results of MIP tests exhibit the influence of suction on the variation of pore size distribution. The transition from the initial state to 150 MPa is reflected by shifting towards the left indicating a decrease in pore size of the clay matrix. The part of the macro-porosity is unaffected by swelling (no cracks created), swelling occurs at the micro-structural level (change within the clay matrix).

As shown in this research, COx claystone exhibits significant water sensitivity due to the expansive minerals. The results of oedometric compression tests demonstrate that the swelling capacity of COx claystone increases with the maximal supported stress. With the study of MIP results, a simple sketch about the evolution of microstructure could be drawn. The porosity decreases linearly with the compression illustrating the ordered collapse of pores, starting from the largest and then the smaller pores. A damage variable D is introduced and a new approach based on the variation of porosity to define D is proposed to quantitatively describe the extent of rock damage. With the damage variable D and maximal supported stress, the model between swelling strain and stress was developed. The result of the model corresponds well with the results of oedometric compression tests upon loading.

Due to the limited period of this work, tests haven’t all been completed up to now. After submitting this report, tests will continue to be carried out. The study about swelling behavior of other samples of COx claystone, even other types of claystone will be carried out in the future to check the validity of the proposed model.

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