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Experimental investigation of the free swelling of crushed argillite

L. Wang*, M. Bornert+, S. Chancole* and E. Héripré*

Callovo–Oxfordian argillite is studied in France as a possible host rock for repository of high-level radioactive wastes. A new experimental procedure is proposed to quantify the free swelling of its main constitutive clay phase, which is a key parameter to model the hydromechanical behaviour of this rock. The material was pulverised finely enough to remove any local stress that may prevent the free swelling induced by a change in water content. Powders were observed using an environmental scanning electron microscope and images recorded at various degrees of relative humidity were analysed by digital image correlation techniques to measure swelling strains. The investigation showed that the free swelling is moderate in comparison to other expansive clays (e.g. bentonite MX80). The magnitude of deformation becomes significant at high relative humidity. This swelling is not reversible and residual contraction is found after a wetting/drying cycle. These results confirm and provide a possible interpretation of some macroscopic phenomena.

KEYWORDS: clays; deformation; fabric/structure of soils; microscopy; particle-scale behaviour; radioactive waste disposal

NOTATION

a	mechanical interaction coefficient
E_1	minor principal strain
E_2	major principal strain
E_{xx}	x-component of strain tensor
f_p	volume fraction of inclusion
ε^F	free swelling of clay minerals
ε	total deformation of argillites under wetting/drying

INTRODUCTION

In the context of high-level radioactive waste disposal, the hydromechanical behaviour of argillite has been intensively investigated in recent decades in view of the safety assessment of long-term repositories. When the relative humidity (RH) varies, argillite undergoes a volume change. This variation of volume essentially consists of two terms – free swelling/shrinking of clay minerals and mechanical deformation induced by the incompatibility of the first term at local scale. Indeed, during wetting, the water-sensitive constitutive clay minerals in argillite swell because of various mechanisms such as crystalline or double-layer swelling (Laird, 2006), but this physico-chemical swelling cannot operate freely because of the heterogeneous microstructure of the rock. Firstly, carbonate and quartz grains do not deform and inhibit the swelling of the surrounding clay minerals and, secondly, various types of clay minerals with contrasting swelling properties and various local orientations swell heterogeneously at local scale. Incompatible free swelling leads to an internal stress field (even if macroscopic stress is null), which generates mechanical deformation as well as possible damage phenomena. The free swelling capacity of the constitutive clay minerals is a key parameter not only for the analysis of

damage but also for the micromechanical modelling of the total deformation of such rocks under wetting or drying conditions.

Quantitative experimental data on free swelling at local scale are still missing because most of the existing investigations have been performed at macro-scale at which free swelling is always coupled with mechanical deformation. A new methodology to quantify free swelling at micrometric scale is proposed here. The argillite was first pulverised finely enough to remove the local stress. The free swelling of the obtained powder particles, essentially made of individual clay aggregates, was then investigated by a combination of environmental scanning electron microscopy (ESEM) and digital image correlation (DIC) techniques.

MATERIALS AND EXPERIMENTAL METHOD

Callovo–Oxfordian (COx) argillite, taken from the ANDRA underground research laboratory at Bure in France, was studied. It is composed of 40–45% (by weight) clay minerals (predominantly inter-stratified illite/smectite), 30% carbonate and 25–30% quartz and feldspar. Samples were crushed using a grinder for 24 h in standard laboratory conditions, without controlling RH. The powder was sieved using a screen with an opening of 100 μm .

Wetting/drying tests on the obtained grains were performed in an ESEM. The RH of the sample surroundings was prescribed by combined control of the sample temperature by a Peltier module and vapour pressure in the ESEM chamber. Images were recorded and analysed by DIC for local and average strain evaluation. DIC consists of matching homologue domains in two images, from which displacement and strain (by derivation of the former) fields can be subsequently determined. Wang (2012) gives more details on strain measurements using the combination of ESEM and DIC techniques. The in-house software CMV was used for two-dimensional (2D) DIC analysis (Bornert *et al.*, 2010). The in-plane components of local and average deformation tensors were calculated over some regions of interest (ROIs) on the selected grains, according to the procedures described by Allais *et al.* (1994). These

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tensors can also be decomposed into rotational and distortion parts, whereby the two principal Green-Lagrange strains (E_1 , E_2 with $E_2 > E_1$) are obtained. The ESEM images are essentially 2D projections of the grain surface, so that neither out-of-plane components of the displacement field nor displacement inside the grains are quantified here. Hence, the actual volume change cannot be quantified exactly. However, the so-called iso-strain ($E_1 + E_2$) is used as a qualitative indicator of volumetric deformation. It is emphasised that the selected ROIs are located on top of the aggregates so that artefacts in strain measurements due to possible global rotations of the grains are limited, and that only grains that do not undergo such rotations, as indicated by the evolution of their contrast in the periphery of the grains, are considered in the analysis.

The general experimental procedure was as follows. The powder of crushed argillite was put directly on the Peltier module and underwent a multi-step swelling/drying cycle in the ESEM chamber. Micro-scale observations and DIC analysis were performed on dozens of clay aggregates. Only isolated ones (as shown in Fig. 1) were chosen to avoid confinement effects and to ensure free swelling. Between each wetting/drying step, the RH was varied up to the new prescribed value at a moderate rate (1–7% RH/min) to prevent the violent break-up of samples, which would be induced by too strong local gradients of water content in the grains during the transient stage. This also avoided condensation of water vapour on the Peltier module. The RH was then maintained constant, waiting for hydric equilibrium of the aggregates. ESEM images were recorded every 2 min for strain measurements. Once the average deformation increment between two successively recorded images became extremely small ($<10^{-4}$), the aggregates were considered to have attained their steady states and the subsequent wetting/drying step was triggered (Fig. 2). This procedure ensures that the strain heterogeneities described hereafter were induced by microstructure only, not by fluctuations of moisture.

RESULTS AND DISCUSSION

Monotonic swelling tests were performed on samples 1 to 6 with RH ranging from about 25 to 80%, while sample 7 underwent a wetting/drying cycle. In crushed argillite, the mesoscopic confinement due to non-swelling inclusions is eliminated. It should, however, be noted that the individual

grains or ‘aggregates’, with typical size ranging from 20 to 50 μm , are indeed assemblies of clay particles with various orientations and inter-particle pores, so that the swelling field still exhibits fluctuations at the scale of a few micrometres, as shown in Fig. 1. The average strains over the considered ROIs (typically 10–20 μm in size) measured here as a function of RH should therefore be considered as the free swelling strain at the scale of clay aggregates, not of individual clay particles. An enlarged view of sample 7 is shown in Fig. 3. In general, the ESEM image seems smoother after wetting. This is associated with the absorption of water on the particles’ surfaces. Moreover, some menisci, associated with the capillary effect, are observed in some inter-particle pore spaces.

Figure 4(a) shows a plot of steady iso-strain as a function of RH during the wetting process for samples 1 to 6. Broadly speaking, the deformation due to wetting is several percent at the highest increment in RH but strong heterogeneity was observed: a maximum of 3.5% was recorded for sample 6, while the minimum was 0.8% for sample 5. The observed heterogeneity can be explained by the anisotropy of local swelling induced by preferential orientation of clay particles on the aggregates. Assuming an axial-symmetric behaviour with two of the three eigenstrains equal to a minimum swelling E_{\min} and the third equal to the maximum E_{\max} , measurements of 2D iso-strains would then range from $2E_{\min}$ to $E_{\min} + E_{\max}$, depending on the orientation of the observed aggregates. In addition, the non-uniformity of measured strains may be related to the different types of clay minerals owning variable swelling capacities; for example, smectite can swell greatly whereas illite and kaolinite hardly deform during wetting processes. The measured results suggest that $E_{\min} = \sim 0.4\%$ and $E_{\max} = \sim 3.1\%$ (i.e. three-dimensional local volume change would be about 4%). These values are far less than the swelling magnitude of $\sim 15\%$ recorded by Montes *et al.* (2003) for bentonite MX80 aggregates subjected to a similar RH increase.

At this stage, it should be mentioned that the accuracy of the measurements is sufficient for these results to be meaningful. Indeed, the various sources of error on strain measurements on COx argillite based on the combination of ESEM and DIC were thoroughly investigated by Wang (2012) and the accuracy was found to be of the order of 10^{-4} for overall strain measurements and 10^{-3} for local strains. In the present case, strains were averaged over

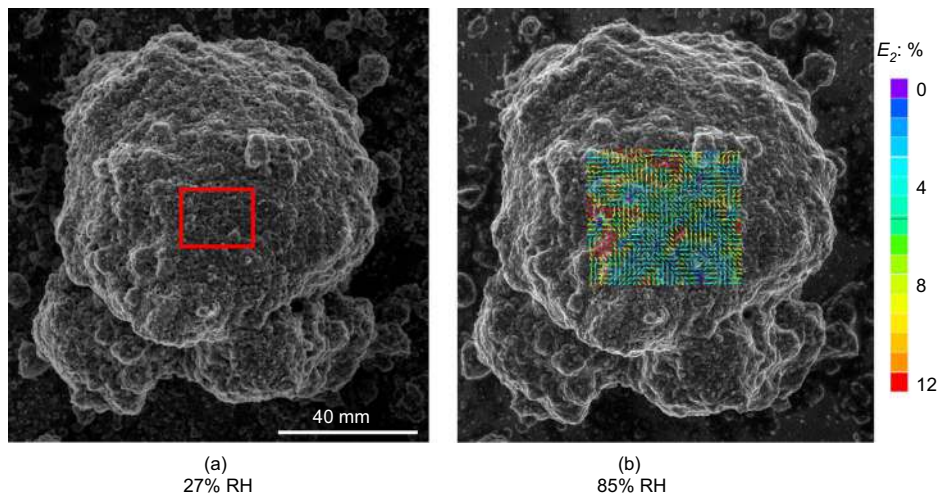


Fig. 1. ESEM image of sample 7 in reference configuration at 27% RH (a) and its swelling strain field at 85% RH (b). The domain outlined in red in (a) is enlarged in Fig. 3

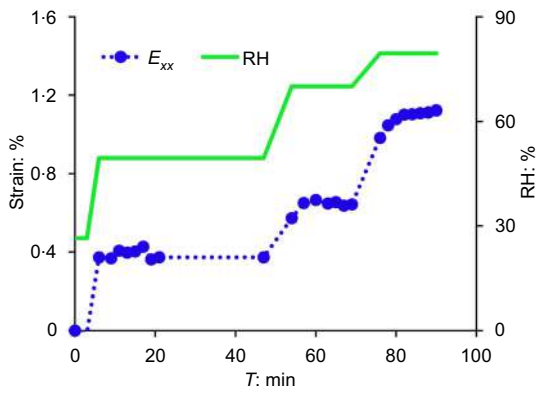


Fig. 2. Wetting history and deformation evolution of sample 6

intermediate areas and the accuracy should be somewhere in between these two values, which is sufficient for the rather large deformation levels (of the order of 10^{-2}) considered in this work. It should be noted, however, that in the present study the evolution of the local image contrast used by DIC for pattern matching is somewhat modified by the absorption of water, as commented on earlier, so that the subpixel accuracy of local displacement measurements is degraded with respect to the above mentioned studies. But there is still sufficient contrast provided by the local topography of the samples at a scale slightly larger than the menisci and still smaller than the DIC windows, so that DIC is still accurate. Even with the pessimistic assumption that the displacement accuracy would only be of the order of a couple of pixels, and when applying the general relations described by Allais *et al.* (1994), the accuracy of the strain averaged over the ROIs considered in this study is better than 10^{-3} .

Let us now compare the current measurements to earlier ones performed on the same rock but at more macroscopic scales. To do so, it should be recalled that the total deformation of argillite under wetting actually involves different complex phenomena. At the inclusion–matrix composite scale, total swelling is the sum of the free swelling of the clay matrix and the mechanical deformation induced by the heterogeneity of the material (in particular, the interaction between the swelling matrix and the non-swelling

inclusions). These effects can be captured, at least qualitatively, by micromechanical models. Assuming, for simplicity, that quartz and calcite inclusions are much stiffer than the clay matrix, describing the latter as an elastically isotropic phase with a Poisson ratio of 0.2 and adopting the Hashin and Shtrikman lower bound for isotropic two-phase media as an appropriate scale transition model, standard micromechanics lead to the following relation between the local volumetric free swelling strain in clay aggregates ε^F and the macroscopic volumetric free swelling of argillite ε

$$\varepsilon = \frac{1-f_p}{(1+f_p)} \varepsilon^F \quad (1)$$

where f_p is the volume fraction of non-swelling inclusions. For a volume fraction $f_p = 40\%$, such a simple estimate would lead to $\varepsilon \approx 1.7\%$ for a 30 to 80% RH change. Such a simple model does not account for the complexity of the real microstructure. To improve it, numerical simulations making use of realistic phase distributions would be required. Preliminary 2D finite-element simulations based on real microstructures observed by ESEM (see Wang (2012) for details) allowed estimation of the ratio of overall free strain to free clay swelling of 0.45–0.70 for f_p varying from 40 to 20%. Such relations allow us to qualitatively compare the present local measurements to overall swelling measurements of argillite for similar RH variations. For instance, Pham *et al.* (2007) recorded a volume strain of 1.7% for a swelling from 33 to 76% RH, while Yang *et al.* (2012) found it to be 0.5% for a 36 to 79% RH increase. Orders of magnitude are very similar, with limited discrepancies probably related to the variability of microstructure and over-simplifications in the above preliminary models. These results emphasise that the free swelling capacity of clay minerals in COx argillite is rather limited, which is consistent with the abundance in these rocks of inter-stratified illite/smectite, in which illite hardly swells.

The evolution of swelling as a function of RH is fairly linear at low RH: this is the case until 80% RH for samples 1–5 and holds true at least up to 70% RH for samples 6 and 7. For higher RH (80% for sample 6 and 85% for sample 7), the deformation increases significantly. This type of non-linearity has also been found for the swelling curve of

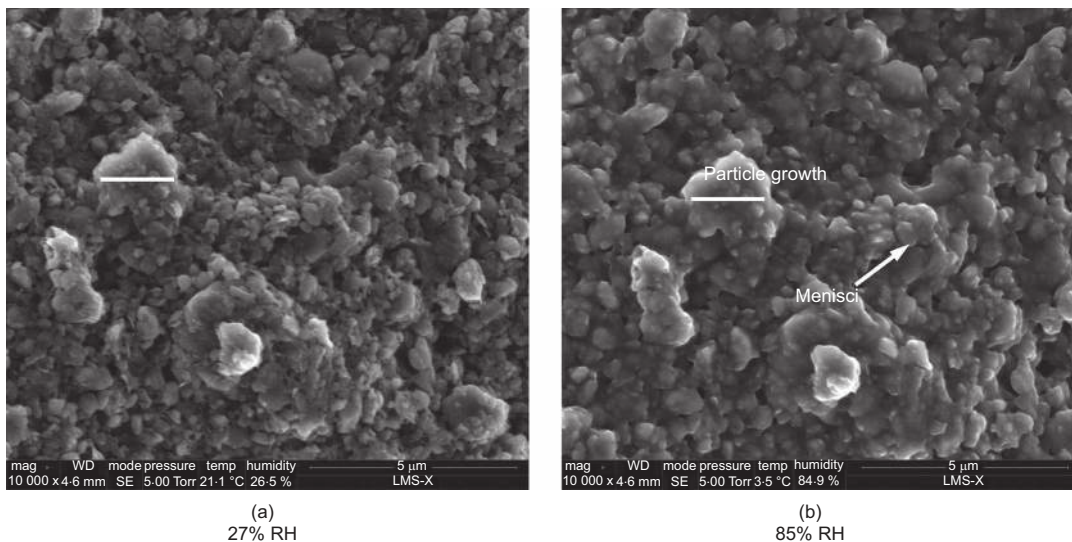


Fig. 3. Hydration of clay aggregate observed by ESEM (sample 7)

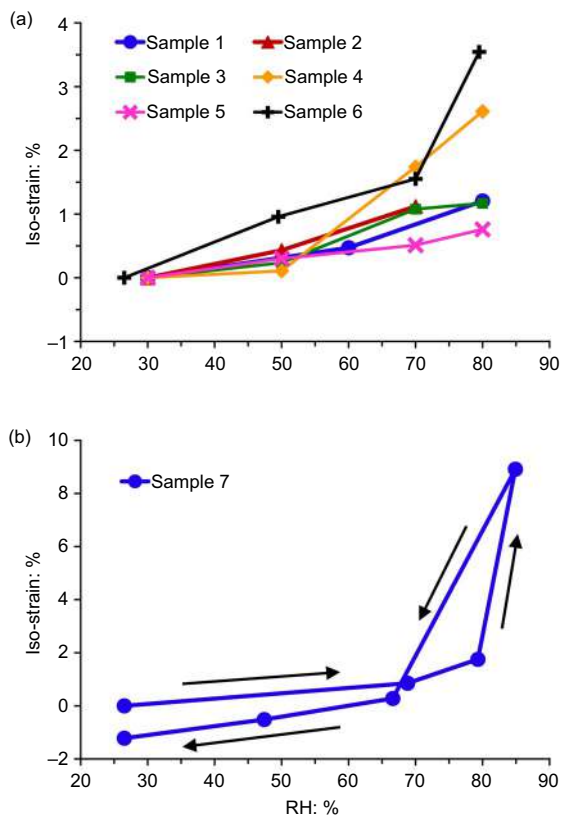


Fig. 4. Steady-state iso-strains measured during monotonic swelling (a) and swelling/drying cycle (b)

argillite. While such behaviour is often interpreted as being due to the micro-cracking induced by wetting, recent experimental investigations (Wang, 2012) have demonstrated that, besides this first mechanism, it might be also linked to the non-linear swelling of the clay matrix itself. The present study confirms this conclusion, both in terms of the existence of this non-linearity and the threshold at which it occurs (RH of about 80%).

Finally, it is also interesting to note the irreversible contraction observed after the wetting/drying cycle (about 1.5% iso-strain; see Fig. 4(b)). This phenomenon has also been found in micro-scale experimental investigations on argillite (Wang, 2012), but is not evident at macro-scale. Its interpretation is so far unclear and additional investigations are thus required.

CONCLUSION

The physico-chemical swelling of clay minerals combined with the deformation due to the local stress induced by its incompatibility contributes to the total deformation of argillite under wetting. Hence, its free swelling capacity is a key parameter for the analysis of damage phenomena and the prediction of total deformation of such rocks under wetting/drying. However, this parameter is hard to determine since it is often coupled with mechanical deformation.

In this work, argillite was crushed and an ESEM investigation was performed on small clay aggregates (dozens of micrometers in size). In this way, the mechanical interaction can be removed and the measurement of free swelling is possible. For COx argillite, the free swelling capacity of clay aggregates is relatively moderate compared with that of other expansive clays (e.g. bentonite MX80). The magnitude determined from experimental results confirms micromechanical predictions derived from the total swelling of argillite measured at macro-scale. Furthermore, the non-linearity of the free swelling curve was verified at local scale. Together with microcracking induced by wetting, this explains the non-linearity of the swelling behaviour observed at larger scale. Finally, some residual contraction was observed in clay aggregates subjected to a wetting/drying cycle.

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