Microstructure Observations in Compacted Clays Subjected to Thermal Loading

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Abstract:

The response of clays to thermal variation has been the subject of extensive studies, given the wide range of applications which involve subjecting soils to substantial temperature fluctuations. A number of hypotheses have been proposed to explain the volumetric changes induced in the clays as a result of temperature variations. Most associate the observed volumetric changes to re-orientation as well as changes in the clay microstructure, with no microstructural experimental evidences to date. The work presented in this note is a first attempt at studying the evolution of the internal structure of two types of saturated and normally consolidated clays, an Illite and a Kaolin, subjected to thermal loading. A series of thermal oedometer, mercury intrusion porosimetry (MIP) and tomography tests were conducted in order to induce, detect and quantify microstructural alterations within the clay as a consequence of temperature changes. Results of heating and cooling tests on Illite showed a thermal contraction which could be attributed to the deformation/collapse of macro-pores in its dual-porosity structure assemblage. The magnitude of

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the observed contraction varied with the level of pre-imposed effective vertical stresses. Higher effective vertical stresses resulted in larger shear stresses at the contacts of clay-assemblages, and thus in easier deformation of the macro-pores. The Kaolin samples which presented a unimodal pore size distribution, with a relatively small dominant pore size (0.25 μm), did not exhibit changes in the microstructure which could be captured by the MIP.

Keywords

Clays; deformation; structure of soils; temperature effects; microstructure

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1. Introduction

Clays are expected to exhibit volumetric thermal deformation of different magnitude depending on several factors such as the clay type, clay plasticity and the loading history. A normally consolidated clay will irreversibly contract as temperature increases, a highly over-consolidated clay will undergo elastic thermal expansion while a slightly overconsolidated clay will first dilate and then tend towards contraction at high temperature (Cekerevac and Laloui, 2004; Di Donna and Laloui, 2015). The driving mechanisms behind such thermal response were attributed to various physical phenomena occurring in the clay microstructure. For instance, Campanella and Mitchell (1968) reported that the main reason behind the thermal volume change is the expansion of the soil solids and pore water during heating, which can be conveniently used to explain the elastic expansion of over-consolidated clays. Campanella and Mitchell (1968) suggested an additional potential contributor which is the increase in the thermal energy due to heating causing a decrease in in-particle shearing strength followed by a particle re-arrangement and thus a sample contraction in case of normally consolidated clays. A different conjecture was proposed by Towhata et al. (1993), in which they suggested that the thickness of the diffuse double layer decreases with temperature, leading to closer contact between

particles and thus, a denser configuration. Conversely, Plum and Esrig (1969) hypothesized that the particles re-arrangement is initiated by the expansion of diffuse double layer with temperature. Another contributing mechanism, described by Paaswell (1967), is the decrease of pore water viscosity with increasing temperature leading to higher permeability of the clay and a faster sample consolidation which was also noticed by Delage et al. (2000) and Towhata et al. (1993). Thus, the physico-chemical interaction between particles controls the clay thermal response which depends also on the plasticity index and the clay percentage (Skempton, 1953; Laloui et al., 2014). However, experimental confirmations of the microstructure alterations upon thermal loadings are not yet available. This note presents a first attempt to detect such alterations by comparing the microstructure of heated and un-heated compacted clays using mercury intrusion porosimetry (MIP) and X-ray tomography techniques. These techniques offer qualitative and quantitative insights into the pore size distribution of clay samples, which can help explain the thermal contractive behaviour of normally consolidated compacted clays.

2. Testing Programme:

Two similar compacted clay samples for each clay type were tested in parallel as a part of the experimental campaign. The samples were first consolidated in an oedometer cell up to the desired vertical stress (60 kPa or 500 kPa). Figure 1 shows the compression curves of both clay samples indicating that the pre-consolidation pressure is approximately 15 kPa for the Illite and less than 10 kPa for the Kaolin, which confirms that all samples were normally consolidated under the desired vertical stresses. Subsequently, one of them was subjected to a thermal cycle whereas the second was considered as a control sample that was loaded only mechanically. Once the oedometer tests were finalized, the samples were unloaded and prepared for the MIP and tomography analyses. These analyses can provide valuable insight into the pore distribution of each sample, covering a pore diameter range from 0.004 μm to 500 μm. As such,

a comparison between the structures of the heated and unheated samples is possible. Although the unloading phase is inevitable and it induces a slight rebound, its effect can be considered negligible on the pore distribution (Cetin, 2004). Moreover, since both the control and the heated samples were unloaded, the comparison is all the more valid.

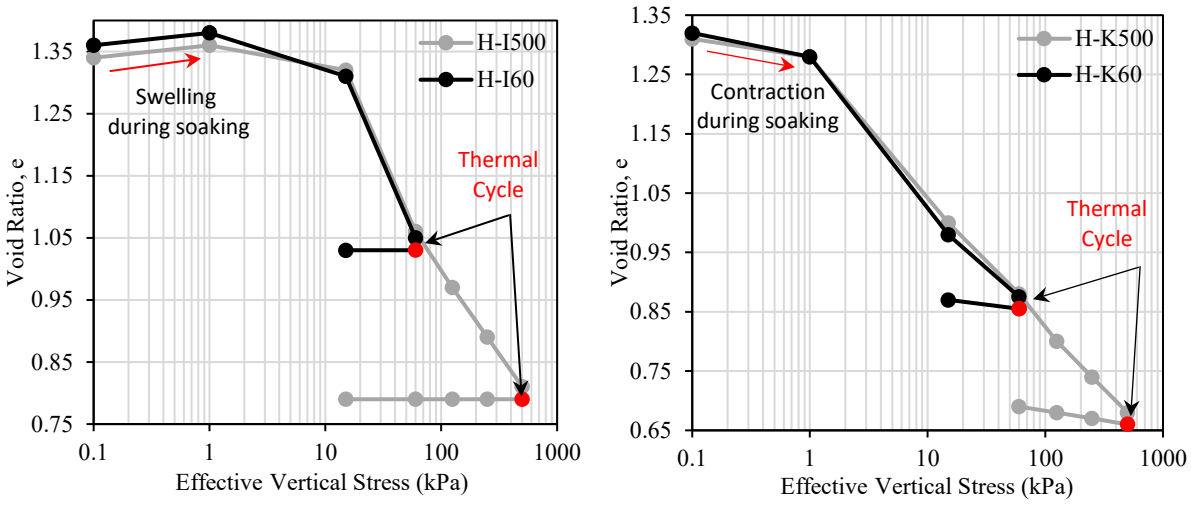


Figure 1. Compression Curves of the heated (a) Illite and (b) Kaolin samples

3. Material and Methods

3.1 Tested Soils

The clays used in this experimental study were commercially available: Illite from Arginotec-GI and Kaolin from PROMAFOR. The mineralogical contents of the Illite sample were 77% illite, 10% kaolinite and 12% calcite and those of the Kaolin sample were 67% kaolinite, 2% illite and 31% quartz. According to the Unified Soil Classification Scheme (USCS), the Illite and Kaolin soils used can be classified as CH and CL, respectively. Table 1 presents a summary of all the relevant soils properties.

Table 1. Soils Properties and Classifications

Soil	G_s	Fraction by weight (%)			USCS	Atterberg Limits (%)		
		Sand	Silt	Clay		LL	PL	PI
Illite	2.650	10.6	17	72.5	CH	56	32	24
Kaolin	2.625	0	66	44	CL	45	24	21

3.2 Sample preparation

A specific sample preparation procedure was devised to ensure reproducible samples of approximately the same initial void ratios of 1.4 and 1.25 for Illite and Kaolin samples, respectively. First, water was gently added to the clay powder with a spray gun along with continuous mixing with a spatula to reach a water content of 35% for the Illite and 28% for the Kaolin. The mix was then left for three days to enable full moisture homogenization. Afterwards, a specific clay mass was compacted statically, dry of optimum, until reaching a target sample height at a constant piston velocity of 0.5 mm/min to prevent the generation of excess pore water pressure knowing that the optimum water contents were 39% and 33% for the Illite and Kaolin, respectively. After compaction, the samples were carefully taken out, trimmed and fitted inside the ring of 60 mm diameter and 15 mm height for the oedometer tests. At the end of the oedometer tests, the samples were trimmed into small specimens of 5 mm x 5 mm x 10 mm for MIP and tomography tests. Since the MIP test requires a fully dried specimen, the freeze-drying method was adopted to prepare the samples, owing that it is the best technique to prevent any structural alteration during dehydration and any capillary effect (Delage and Lefebvre, 1984). Consequently, the trimmed pieces were immediately submerged in liquid nitrogen bath at –196°C for about half an hour to rapidly freeze the pore fluid. The frozen samples were then placed in a freeze-drier for 24 hours, which dried them by sublimation at a temperature of –60°C and a vacuum of 0.03 mBar.

3.3 Testing Procedure:

Following static compaction, the clay samples were fitted in an invar ring and placed inside the oedometer cell. Distilled water was then added to ensure sample saturation. The consolidation process began by adding calibrated dead weights every 24 hours, according to the following loading steps: 1, 15, 60, 125, 250, 500 kPa. After loading the samples to the desired vertical stress for 24h, the control sample was left under this stress and creep displacements were recorded (Standard oedometer test). In parallel, the second sample was subjected to one thermal cycle (Thermal oedometer test) by circulating water inside the copper tubes placed around the oedometer ring under a controlled temperature and flow using a Huber thermostat. Starting with a base temperature of 20ºC, the maximum temperature reached inside the cell was 70°C. This high temperature was held constant for 4 days, at the end of which the sample was cooled back to the ambient temperature (20°C). The heating and cooling rates were 10°C per 3 hours guaranteeing the dissipation of the induced excess pore pressures (Cekerevac and Laloui, 2004; Di Donna and Laloui, 2015). During testing, the temperature inside the cell was monitored using type-K thermocouples with an accuracy of 0.1°C and the sample settlements were measured using an LVDT (operating scale OS ±2.5 mm, sensitivity 0.099 mm/mV, accuracy ±0.21% OS). Prior to any test, the setup was carefully calibrated under the same temperature changes. More details about the thermal oedometer setup can be found in Di Donna and Laloui (2015).

Subsequently, the samples were unloaded and prepared for the MIP and tomography tests. The MIP tests were performed on the freeze-dried samples using the Thermo Electron Pascal 140/440 equipment. The mercury intrusion pressure, P, was used to calculate the pore diameter, D, based on Equation 1.

$$D = \frac{-4\gamma cos\theta}{P} \quad (1)$$

Where γ is the surface tension of the mercury (0.485 N/m at room temperature) and θ is the contact angle which is assumed to be 140° for clays (Diamond, 1970). The pore-size density (PSD) function for each sample was generated using Equation 2, where *e* is the cumulative intrusion void ratio defined as the intruded pore volume of mercury normalized by the solid volume.

$$PSD = \frac{-de}{d(\log D)} \quad (2)$$

The tomography analyses were performed using the 3D X-Ray tomography "UltraTom" from RX-SOLUTIONS. The samples were scanned with X-rays having a source voltage of 50 kV and a current of 150 µA. Around 1450 slice images were acquired with a resolution of 1450 x 1500 pixels. The X-Act CT software was then used to reconstruct the 3D sample with a voxel size of 2.1725 μm^3. The image processing, 3D visualization and the volume analysis were then carried out using the software AVIZO 9.1. The CT images were first filtered for noise reduction. A 2.127 x 2.25 x 1.14 mm sub-volume was then extracted from the center of the reconstituted sample to eliminate the border effects and any disturbed areas during sample preparation and handling. Thresholding was applied to the gray scale images to segment the pore volume. It is worth mentioning that the same gray scale intensity was used for both samples in order to get comparable results.

Table 2 shows all the tests detailing the different conditions. The heated samples are designated by H, the unheated by U for both the Illite (I) and Kaolin (K).

Table 2. Experimental Program

Test Number	Designation	Soil Type	Test Type	Temperature	Desired Stress	Characterization Test
1	U - I60	Illite	Standard	20°C	60 kPa	MIP – XRAY
2	H - I60	Illite	Thermal	70°C	60 kPa	MIP - XRAY
3	U - I500	Illite	Standard	20°C	500 kPa	MIP
4	H -I500	Illite	Thermal	70°C	500 kPa	MIP
5	U - K60	Kaolin	Standard	20°C	60 kPa	MIP
6	H - K60	Kaolin	Thermal	70°C	60 kPa	MIP
7	U - K500	Kaolin	Standard	20°C	500 kPa	-
8	H - K500	Kaolin	Thermal	70°C	500 kPa	-

4. Experimental Results

4.1 Microstructure of Compacted Clays

The difference between the microstructure of the Illite and Kaolin samples is highlighted by the MIP results in Figure 2. The pore-size density of the Illite shows a bi-modal distribution with dominant pore diameters of 0.015 μm and 9 μm, which indicates that the compacted Illite samples present a dual porosity, e.g. macro- and micro-porosities as described by Collins and McGown (1974). The transition between these porosities can be set at 0.2 μm where the pore size density drops to a minimum value of 0.06. Accordingly, the pore sizes lower than 0.2 μm can be defined as the intra-assemblage pores formed between elementary particles (within clay aggregates). Those greater than 0.2 μm can be considered as the inter-assemblage pores formed between clay aggregates. On the other hand, the Kaolin shows a unimodal distribution with one dominant pore diameter of 0.25 μm, which corresponds to the intra-assemblage pores. This unimodal distribution was probably a result of soaking the sample for saturation which

caused the collapse of Kaolin aggregates initially observed when water was added to the oedometer box under a vertical stress of 1 kPa (Koliji et al., 2010). Once submerged in water, the Kaolin sample contracted as shown in Figure 1(b) where the void ratio decreased at 1 kPa, which points to the high sensibility of Kaolin aggregates to water, leading to the destruction of macropores. In contrast, the Illite samples expanded slightly during the same phase (Figure 1 (a)).

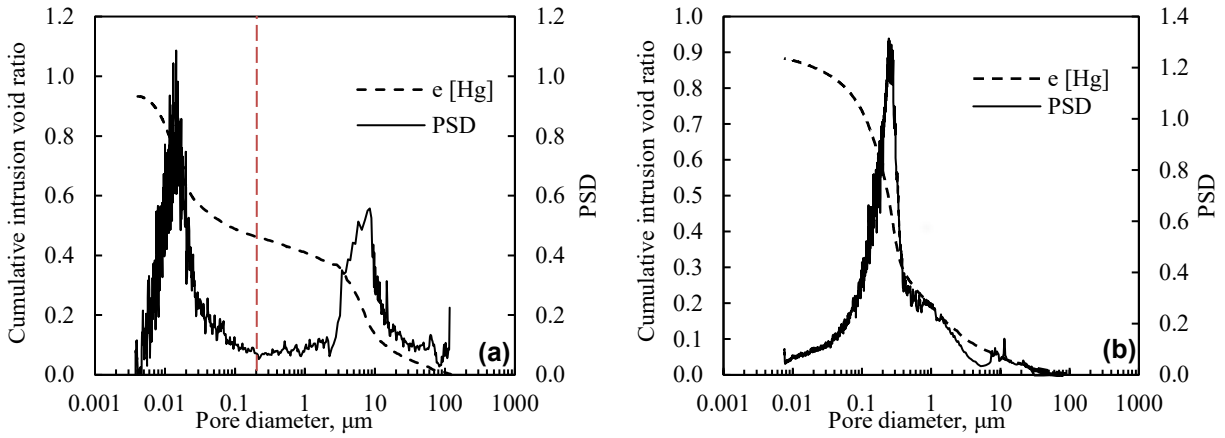
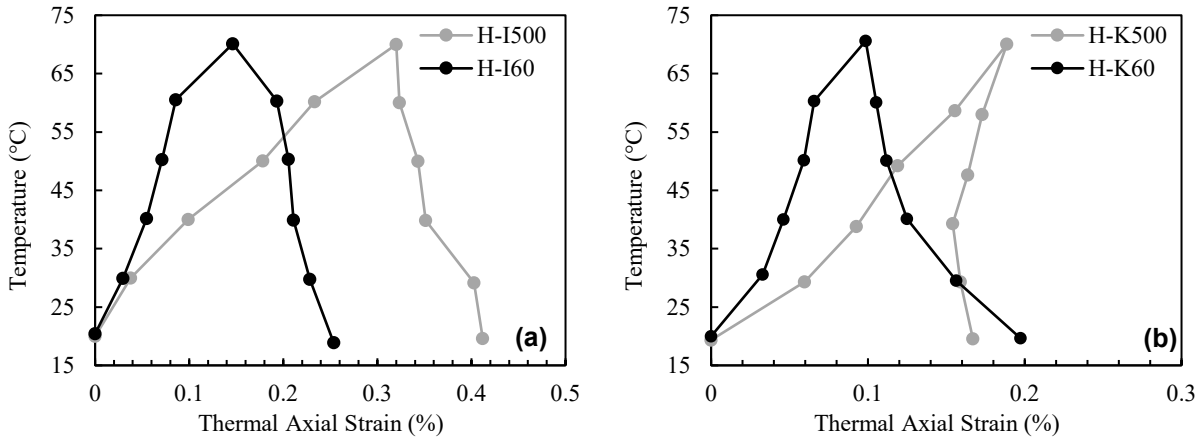


Figure 2. Pore-size density function of (a) Illite and (b) Kaolin samples at 60 kPa

4.2 Thermal Volumetric Response

The results of the thermal oedometer tests are presented in Figure 3 in terms of thermal axial strain, where the positive sign of strains refers to a downward displacement (contraction) and also in Figure 1 in terms of void ratio. All samples exhibited a contraction during heating and cooling except for the Kaolin loaded to 500 kPa (H - K500) which showed a slight expansion when cooled down to the ambient temperature. The irreversible deformations reached 0.26 % and 0.42 % for the Illite samples at 60 kPa and 500 kPa, respectively, compared to 0.2% and 0.17% for the Kaolin samples. Such difference can be attributed to the respective microstructures of each sample, more specifically the different pore sizes that can respond

158 distinctively to heating and cooling. This difference can also be a reason for the resultant stress
159 state dependency where plastic deformations are higher for the Illite sample loaded to 500 kPa
160 than that loaded to 60 kPa. This observation is in apparent contradiction with other studies,
161 which reported a stress level independency (Sultan et al., 2002; Abuel-Naga et al., 2007).
162 However, the mineralogy and composition of clays may also have a significant impact on their
163 thermal response. Having the higher plasticity index, Illite is expected to have higher potential
164 for thermal volume change, as demonstrated by Sultan et al. (2002).



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166 **Figure 3. Thermal axial strain at vertical effective stresses of 60 kPa and 500 kPa for: (a)**
167 **Illite and (b) Kaolin**

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169 ***4.3 Microstructure Evolution***

170 A better interpretation of the clay thermal response is achievable by comparing the MIP results
171 between the heated and unheated samples as shown in Figure 4. Thermal loadings clearly
172 affected the macropores of the Illite samples by either reducing their density or their size. A
173 small reduction in the macropore density is noticed when comparing the pore-size density of U-
174 I60 and H-I60 without affecting the range of dominant pores, which is reflected by a slight
175 decrease of the cumulative void ratio from 0.935 to 0.923 and the downward shift of the void

ratio distribution. At 500 kPa, the macropores shifted to the left after thermal loadings conserving approximately the same density and the dominant macropore became at around 1 μm. The same shift is seen in the cumulative intrusion void ratio plots where the pores having a size higher than 10 μm constitute a void ratio of 0.2 in the case of the unheated sample, which decreased to only 0.03 in the case of heated sample. Such prominent macropore deformation is revealed by the higher thermal contraction observed during the oedometer test for H-I500. Conversely, the micropores of all samples seem to remain intact implying that the intra-elemental distances do not vary with temperature variations.

The same deformation of macropores can be observed in the X-Ray tomography results (Figure 5) that covered the pore range undetected by the MIP for U-I60 and H-I60 (i.e. more than 100 μm). Referring to the 3D view of the pore space, the macropores in the heated sample are smaller and more dispersed compared to those of the un-heated one. This is also clear in the transversal section where the maximum detected pore size reached 500 μm in the unheated sample compared to 170 μm in the heated sample. On the other hand, the superposed pore-size densities of the heated and unheated Kaolin samples (Figure 6) indicate that the temperature did not modify the internal structure. The limited thermal strains detected during the oedometer tests can be one reason of such observation, in addition to the small pore sizes of the Kaolin samples, which are less likely to collapse with temperature variations. However, it could be argued that the macropores of Kaolin, undetected by MIP, have been deformed inducing minor thermal contraction.

As a conclusion, macropores would gradually deform upon temperature variations leading eventually to a change in the pore-size density and thus a permanent thermal contraction without affecting the micropores in case of compacted clays similar to the ones used in this study. Such contraction is highly related to the clay type, the stress state and the initial pore size density. The larger the sample pores, the longer the bridges between the particle assemblage

and thus the more prone they become to collapse upon thermal and mechanical loading. In addition, increasing the vertical stress would increase the shear stresses between the particle assemblages facilitating the deformation of macropores once the structure is subjected to additional thermal loading. These observations indicate that the diffuse double layer thickness would not change with the tested levels of temperature variations, as reported by Mitchell and Soga (2005), since only the macropores will be affected. These macropores are large enough in a way that they could not be altered by the double layer (Delage et al., 1983) and thus their effect would be minimal.

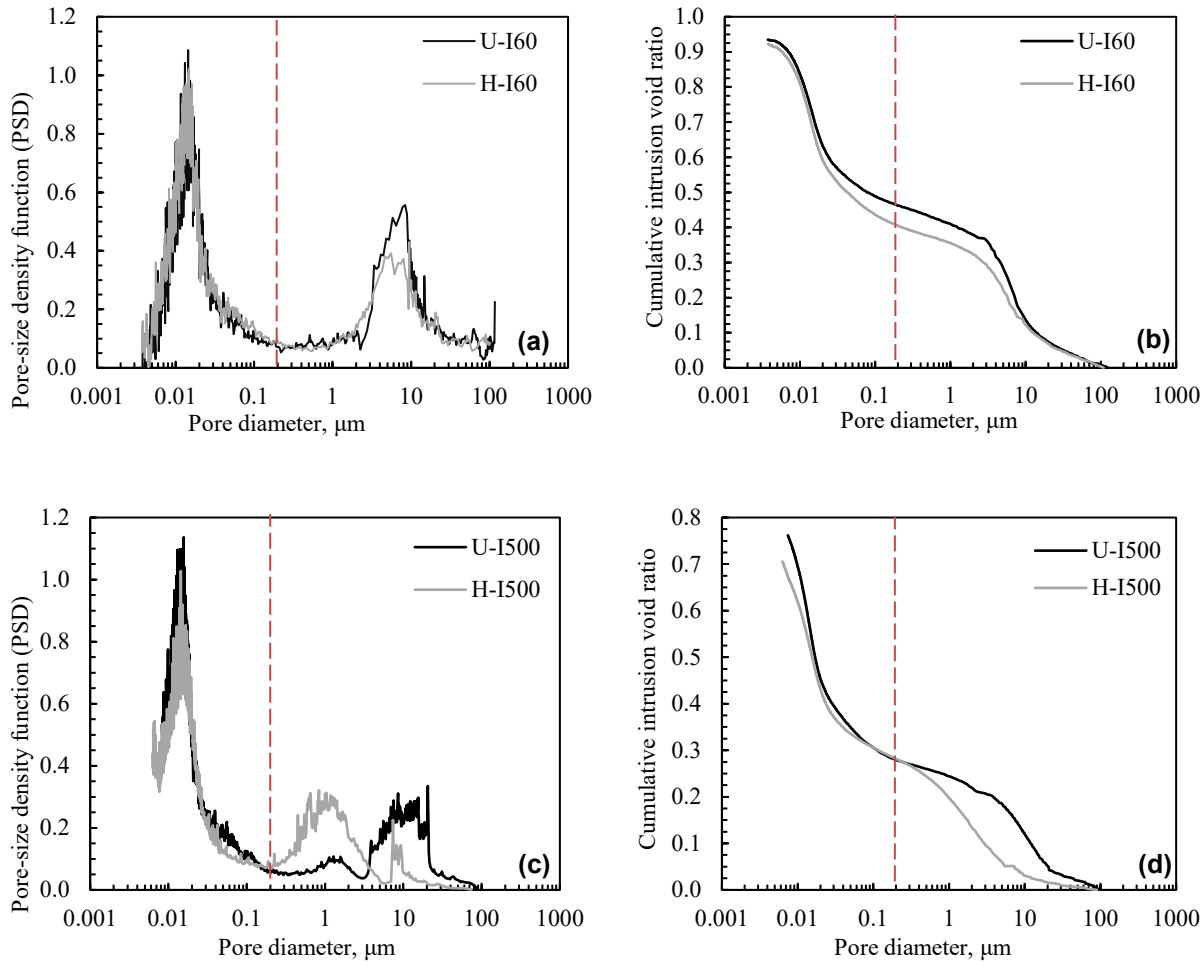
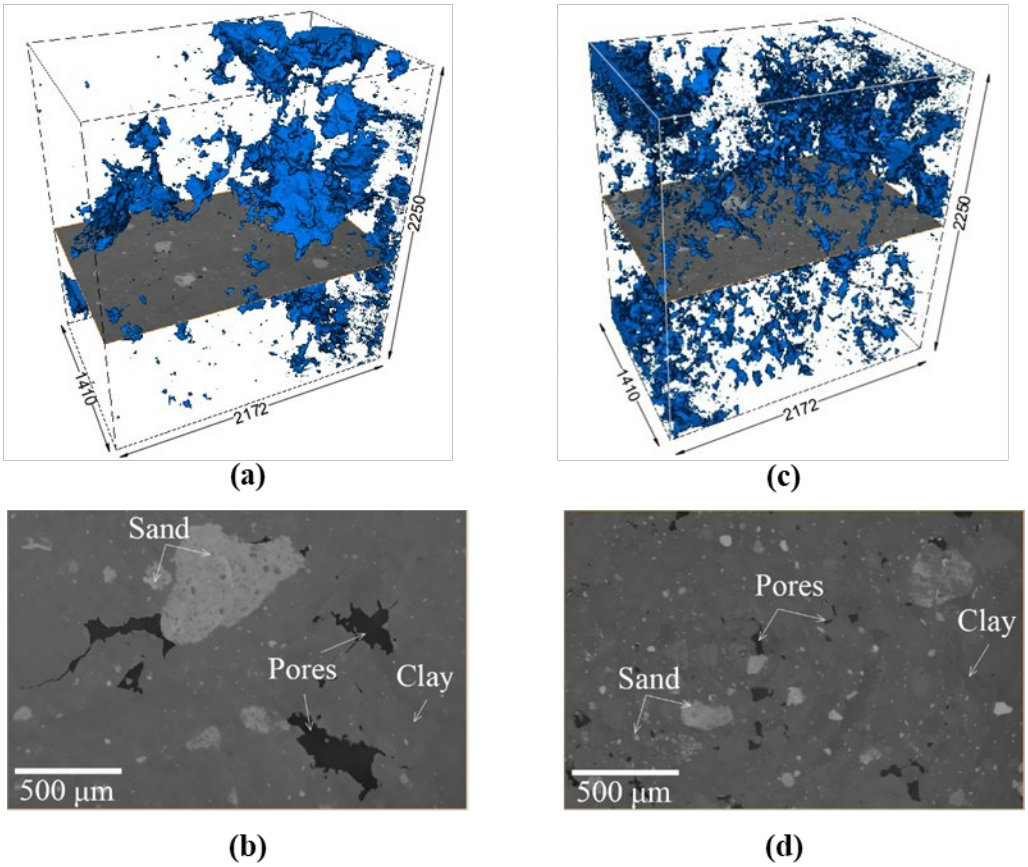


Figure 4. Comparison of MIP results between the heated and unheated Illite samples at: (a,b) 60 kPa and (c,d) 500 kPa



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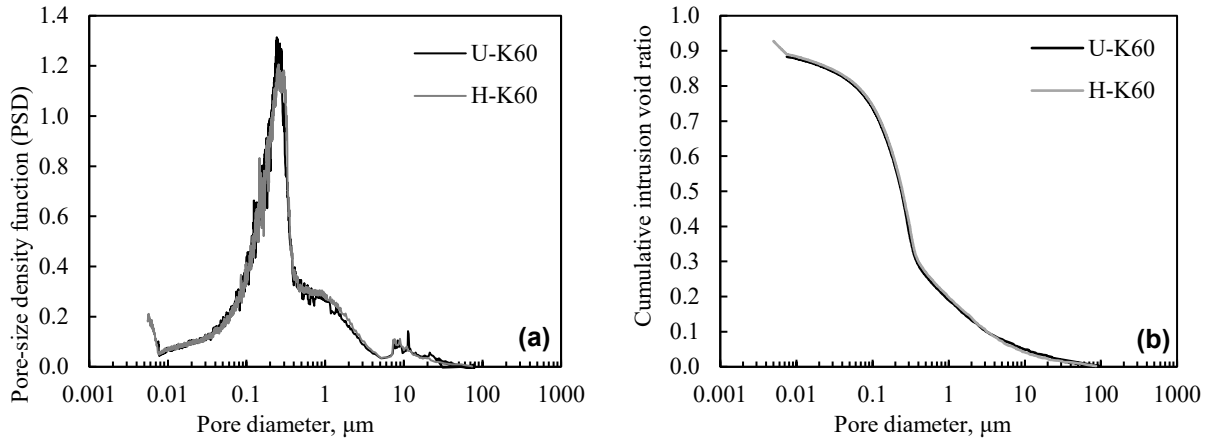
214 **Figure 5. Reconstructed pore volume and a detailed transversal section of: (a,b) un-**

215 **heated and (c,d) heated Illite samples**



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217 **Figure 6. Mercury intrusion porosimetry results for the Kaolin samples at 60 kPa**

5. Conclusions

In this study, the microstructural response of compacted Illite and Kaolin samples subjected to mechanical and thermal loads was quantitatively and qualitatively investigated using MIP and tomography tests. After thermal oedometer tests, the clay samples permanently contracted showing a dependency on the pore size density, the stress state and the clay composition. As revealed by MIP and tomography tests, the Illite, having a bimodal distribution, was more susceptible to contraction due to the collapse of macropores upon temperature variations. Higher vertical effective stresses induced significant deformation of macropores and thus a more pronounced plastic thermal contraction of the Illite. However, the micropores of the Illite and those of the Kaolin having a unimodal distribution remained intact after thermal loadings, indicating that modifications of the diffuse double layer are not expected to be a main reason behind the observed thermal volumetric deformations.

6. CRediT author statement

Roba Houhou: Conceptualization, Methodology, Investigation, Original draft preparation.

Melis Sutman: Supervision , Methodology, Reviewing and Editing, Resources

Salah Sadek: Writing- Reviewing and Editing, Funding Acquisition

Lyesse Laloui: Funding Acquisition, Problem statement, Scientific approach definition, Methodology Analysis and evaluation, Project administration

7. Data Availability

The data will be available upon request.

8. Acknowledgements

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