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1 **Macroscopic effects of nano and microscopic phenomena in clayey soils and**
2 **clay rocks.**

3

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13

14 **Abstract:** Based on long term investigations of the microstructure of various geomaterials as
15 different as sensitive low plastic clays from Eastern Canada, unsaturated compacted silt from
16 the Paris area, highly plastic compressible deep marine clay from the Gulf of Guinea,
17 compacted MX80 bentonite and the Callovo-Oxfordian claystone, considered as possible
18 barriers in deep geological radioactive waste disposal, two fundamentals and distinct nano
19 and micro mechanisms governing their macroscopic volume changes have been identified. In
20 low plastic structured clays and dry compacted soils, in which an aggregate microstructure has
21 been evidenced, the decrease in volume under mechanical compression result from the
22 collapse of inter-aggregates pores in an ordered manner, from the larger to the smaller, with
23 no effect on the intra-aggregate porosity. The soil skeleton can hence be modelled as an
24 elastic fragile porous matrix, affected by the ordered collapse of its pores. Things are different
25 in plastic soils, due to the significant reactivity of the montmorillonite minerals to changes in
26 water content that results in significant changes of the initial porous matrix. The ordered
27 adsorption of layers of water molecules in clay platelets with respect to the suction or stress
28 applied, evidenced long time ago in Soil science through X-ray diffraction techniques, appear
29 to be able to help better understanding various phenomena like the compression of plastic
30 compressible soils and the hydration of compacted bentonites. Interestingly, it also applies to
31 understand the effects of smectite minerals in the volume changes behaviour of the Callovo-
32 Oxfordian claystone. These two mechanisms at nano and microscopic scales hence provide a
33 better understanding of the macroscopic volume changes of a large variety of natural and
34 compacted clayey soils and rocks.

35 **Keywords:** clayey soils and rocks; microstructure; montmorillonite; pore size distribution;
36 nano scale; water adsorption.

37

38 Introduction

39 It is well admitted that microstructure effects have significant consequences on the
40 macroscopic mechanical response of clayey soils and rocks. This was first observed when
41 investigating the compression behaviour of sensitive clays of Eastern Canada, in which a
42 sudden collapse is observed once overpassing the yield stress (e.g. Locat and Lefebvre 1985).
43 This collapse was related to microstructure changes by Delage and Lefebvre (1984), who
44 showed that sensitive Champlain clays are characterised by an aggregate microstructure, and
45 that their compression is related to changes in pore size distribution. This concept, that will
46 be detailed later in this paper, has been extended to unsaturated dry compacted soils, but
47 could however not be extended to plastic soft compressible soils, because of the particular
48 role played by the montmorillonite minerals (also called smectite) responsible of the high
49 plasticity of these soils. In this case, pioneering data from X-ray diffraction techniques from
50 Soil Scientists on the hydration of smectites evidenced some nano-scale mechanisms
51 governing the clay-water interactions in hydrated smectites. These mechanisms also appeared
52 to be valid to better understand the swelling behaviour of plastic soils, in particular in the case
53 of compacted bentonite used as engineered barriers in radioactive waste disposal. More
54 recently, it has been demonstrated that they also govern the swelling behaviour of claystone
55 considered as possible host-rocks for radioactive waste disposal.

56 In this paper, the macroscopic hydro-mechanic volume changes of various clayey soils and
57 rocks are interpreted with respect to these mechanisms occurring at the nano and micro-
58 scales, for various saturated and unsaturated geomaterials of various plasticity indexes.

59 Pore collapse compression mechanisms in low plastic clay soils and dry 60 compacted soils

61 The microstructure of sensitive clays was characterised by using a Jeol Cryoscan scanning
62 electron microscope (SEM) that allowed to observe (Delage et al. 1982) the freeze fractured
63 plane of a sensitive clay from Saint-Guillaume, a moderately plastic clay (low and moderate
64 plastic soils are defined by $I_p < 50$ according to Casagrande's classification) from the Champlain
65 Sea, in the Province of Québec, with $80\% < 2 \mu\text{m}$, $I_p = 22$, clay minerals mainly containing illite.
66 The Cryoscan device allowed to minimise sample perturbations thanks to ultra-rapid freezing
67 followed by freeze-fracturing. In such conditions, crypto-crystalline ice is formed, with no
68 significant volume expansion (Gillott et al. 1973, Tovey and Wong 1973, Tessier and Berrier
69 1978, Delage and Pellerin 1984). Defining an observation plane by fracturing a frozen
70 specimen also benefits from the fact that fracture is governed by the breakage of the ice, that
71 acts as an impregnation resin keeping the microstructure intact during fracturing.

72 Observation of Figure 1 shows a typical aggregate microstructure, with inter-aggregates
73 bridges made up of clay platelets and large inter-aggregates pores. This aggregate
74 microstructure has also been observed in various other sensitive clays from the Champlain sea
75 by Delage (2010), who investigated microstructure by the combined use of mercury intrusion
76 porosimetry (MIP) and scanning electron microscopy. Figure 2 shows a SEM photo of Saint-
77 Marcel clay, and Figure 3 shows the pore size distribution (PSD) curves of intact, remoulded
78 and dried freeze-dried specimens of Saint Marcel clay. The Figure shows a well sorted curve
79 at intact state, with an average pore diameter of $0.18 \mu\text{m}$ and 80% of the intruded pore

80 volume in the range 0.1 – 0.3 μm . As shown in Delage (2010), this section of the PSD curve
81 corresponds to the inter-aggregates porosity, whereas the smaller pores below 0.1 μm
82 corresponds to the intra-aggregate porosity. The average entrance diameter of 0.18 μm is
83 smaller than the average pore diameters of about 0.5 – 1 μm observed in the photo of Figure
84 2 because of the constriction (or ink-bottle effect) resulting from the various clayey bridges
85 crossing the inter-aggregate distance (see Figure 1 and 2). In other words, the average
86 diameters detected by mercury during mercury intrusion in the inter-aggregates pores are
87 smaller than the real pore diameters. This is also due to the tortuosity effects in the inter-
88 aggregates pores.

89 Another important point observed on Eastern Canada sensitive clays is the conservation of
90 aggregates after remoulding. This was directly observed in SEM photos in Delage & Lefebvre
91 (1984) on a specimen manually remoulded in the lab by using a spoon, and can also be seen
92 on the PSD curve of a remoulded Saint-Marcel clay (Figure 3). The curve differs from that of
93 the intact soil in the range of inter-aggregates porosity, and is superimposed in the intra-
94 aggregate porosity range, for pores with radius smaller than 0.8 μm . The weak points of the
95 microstructure affected by remoulding are the clayey bridges joining the aggregates together,
96 that are destroyed by remoulding. The order of magnitude of the inter-platelets bonds within
97 the aggregates is obviously stronger, since they are not affected by remoulding, that keeps
98 their intra-aggregates pores intact. This observation that the elementary microstructure unit
99 involved (but not destroyed) by remoulding is the aggregate has also been made in Lapierre
100 et al. (1990) who adopted the same methodology on another eastern Canada sensitive clay
101 (Louiseville), and has recently been confirmed on a Swedish sensitive clay (Utby) by Birmpilis
102 et al. (2019), based on an investigation conducted by using Small Angle X-ray Scattering (SAXS)
103 and Digital Image correlation techniques (DIC).

104 Figure 3 also shows that the aggregates didn't resist to drying, that resulted in significant
105 shrinkage, as shown by the much smaller intruded volume and average pore diameter. Only
106 one well sorted pore population of very small radius (38 nm) is observed, showing that the
107 inter-aggregate pores have been completely erased.

108 Unsurprisingly, the lower strength level at the inter aggregates level also governs the
109 compression behaviour, as shown in Delage and Lefebvre (1984) on the Saint-Marcel
110 Champlain clay. Indeed, the change in PSD curve of specimens of Saint Marcel clay submitted
111 to various level of vertical stress in the oedometer (124, 421 and 1452 kPa, Figure 4) shows
112 that compression progressively affects, in an ordered fashion, smaller and smaller inter-
113 aggregates pores, starting from the largest ones (at around 1 μm entrance radius) at yield
114 stress (54 kPa). As an example, the PSD curve at 124 kPa clearly show that all pores with radius
115 larger than 0.4 μm have been collapsed, whereas those smaller than 0.3 μm are still intact.
116 Actually, the simultaneous presence of (larger) collapsed pores and (smaller) intact pores has
117 been also observed in SEM, as described in Delage and Lefebvre (1984). Note also that in all
118 cases, all PSD curves are similar in the intra-aggregate range, showing that the maximum
119 vertical stress of 1452 kPa is not high enough to compress the aggregates.

120 Indeed, this clear segregation in size of collapsed pores based on the vertical stress applied
121 allowed to consider, in these sensitive clays, that the compression curve of intact samples
122 could also be considered as a pore size distribution curve, as demonstrated by the good
123 correspondence between the slope of the PSD curve (C_p) and that of the compression curve

124 (compression coefficient C_c) observed on 6 different Champlain clays by Delage & Le Bihan
125 (1986), Delage 2010 and 2014. In other words, the MIP investigation showed that a given
126 compression stress increment will collapse a given population of pores defined by their size,
127 starting from the larger ones (lower stress) and progressively collapsing smaller and smaller
128 ones (at higher and higher stress). In each case, the decrease in volume provides the pore
129 volume corresponding to the collapsed pores, that have their size related to the stress
130 increment applied. The two parameters necessary to define a pore size distribution curve are
131 hence met: the stress increment, related to the pore size, and the resulting decrease in
132 volume, providing the corresponding porous volume (see Delage 2010 for more detail). As a
133 consequence, Delage et al. (2010, 2014) observed that a more compressible soil (high C_c) had
134 a better sorted PSD (high C_p).

135 Following the methodology of Delage and Lefebvre (1984) based on the combined use of SEM
136 and MIP, similar observations have been made (Delage et al. 1996) on a low plasticity Jossigny
137 silt compacted on the dry side ($w = 14.9\%$, $\rho_d = 1.63 \text{ Mg/m}^3$, $S_r = 59\%$) of the Proctor optimum
138 ($w_{\text{Opt}} = 18.3\%$, $\rho_{d-\text{Opt}} = 1.68 \text{ Mg/m}^3$, $S_{r-\text{Opt}} = 77\%$), observations that have been confirmed by
139 others afterwards (e.g. Romero et al. 1999, Lloret et al. 2003). Figure 5a clearly shows that the
140 microstructure on the dry side is also composed of aggregates, with a $110 \mu\text{m}$ diameter
141 aggregate composed of silt particles of $20 - 30 \mu\text{m}$ average diameter. A $28 \mu\text{m}$ inter-aggregate
142 pores can also be observed below the aggregate, together with other ones surrounding the
143 aggregate. Unsurprisingly, this work confirmed previous observations from Ahmed et al.
144 (1974) about the bimodal nature of the PSD curve on the dry side of the Proctor optimum
145 (Figure 6), that defines both the inter-aggregate porosity (average pore entrance radius 2.7
146 μm , 52% of the intruded porosity) and the intra-aggregate porosity (average pore entrance
147 radius $0.28 \mu\text{m}$, 48% of the intruded porosity). Again, due to ink-bottle effects, the average
148 entrance pore diameter of the inter-aggregate porosity detected by MIP is significantly smaller
149 than the inter-aggregate pores observed in SEM.

150 The change in pore size porosity during compression has been derived (Delage and Graham
151 1995, Delage 2009) from earlier data of Sridharan et al. (1971) who run MIP tests in samples
152 compacted at various densities at same water content of 21% . The various PSD curves of
153 Sridharan et al. (1971) are plotted together in Figure 7. Based on the hypothesis that, due to
154 both capillarity and adsorption, water is contained in smaller pores, one can consider that the
155 pores corresponding to intruded porosity below the $w = 21\%$ line (with entrance diameter
156 smaller than $1 \mu\text{m}$) are saturated. The changes in PSD curves with decreased porosity show
157 that compression mainly concerns the larger inter-aggregate larger pores, located above the
158 21% line. Based on the zone in which PSD curves are superimposed, a limit value between the
159 intra-aggregate porosity (the common section of the curves) and the inter-aggregate one (the
160 section that changes with decrease in porosity), can be estimated at around $0.5 \mu\text{m}$, which
161 corresponds to a suction around 600 kPa (using an air-solid interfacial tension value $\sigma_s = 72.75$
162 $\times 10^{-3} \text{ N/m}$ and, for the contact angle, $\cos \theta = 1$). This would mean that, for suctions smaller
163 than 600 kPa , some inter-aggregate menisci could exist, while having most of the inter-
164 aggregate porosity filled by air. For suction larger than 600 kPa , menisci develop within the
165 aggregate, and the inter-aggregate bonding results from adsorption effects through
166 connecting contact governed by hydrated clay particles. The void ratio of 0.59 corresponds to
167 a stage close to saturation, that is reached at $e = 0.56$.

168 A macroscopic consequence of this feature can be derived from monitoring the changes in
169 suction at constant water content with density observed in various compacted soils, and
170 reported in Figure 8 (Li et al. 1995, see also Gens et al. 1995) from data on a compacted loess
171 ($I_p = 10$). One can observe that constant suction extrapolated curves are close to vertical on
172 the dry side, showing that compression at constant water content occurs at (almost) constant
173 suction, with $s \approx 90$ kPa at $w = 16.5\%$, $s \approx 80$ kPa at $w = 18\%$, $s \approx 70$ kPa at $w = 19\%$. This trend
174 changes when approaching to the Proctor optimum water content ($w = 20.5\%$), where the
175 constancy in suction only holds until dry a density value of $\gamma_d = 15.75$ kN/m³. At larger dry
176 densities, iso-suction curves become curved and progressively adopt, at largest values,
177 inclinations close to that of the saturation curve. The constancy of suction at constant water
178 content on the dry side is explained by the fact that, as observed in Figure 7, compression
179 occurs by compression of the aggregate porosity full of air and mutual rearrangement of the
180 aggregates. Suction, that is governed by capillary and physico-chemical clay water interaction
181 within the aggregates, is not affected by compression, because the aggregates themselves are
182 not compressed in the range of stress corresponding to this range of density and water
183 content. The conservation of aggregates already observed in saturated sensitive clays is hence
184 confirmed in unsaturated compacted soils, as also confirmed by others (e.g. Lloret et al. 2003).

185 The data of Figure 8 show that this is no longer valid on the wet side of the optimum (with
186 $w = 21.5\% > w_{opt} = 18.3\%$, $\rho_d = 1.63$ Mg/m³, $S_r = 83.5\%$), an area where the aggregate structure
187 is no longer observed. Indeed, as observed in Figure 5b, the microstructure is characterised
188 by a clay matrix. In other words, the clay fraction of the Jossigny silt ($34\% < 2$ μ m) is fully
189 hydrated, and the clay particles are no longer stuck along the faces of the silt grains as in the
190 case of the dry compacted specimen. Once hydrated, their volume is much larger, and they
191 are able to act as a clay matrix in which silt grains are embedded. As a consequence, as
192 observed in Figure 6, the PSD curve is no longer bi-modal, as in the case of aggregate
193 microstructures (see Delage et al. 1996 for more details). Although it is not clearly observable
194 in the SEM photo, this area of high degree of saturation ($S_r > 85\%$) should correspond to a
195 zone with individual air bubbles not connected together. In this area, compression affects the
196 clay matrix that governs the soil suction, which results in a decrease in suction with
197 compression.

198 The general conclusion drawn from compression data in sensitive clays and dry compacted
199 soils is that both soils are characterised by a rigid porous skeleton made up of aggregates
200 bonded together with fragile bonds, and that compression occurs by the ordered and
201 progressive collapse of inter-aggregate pores, with no effect on the intra-aggregate porosity.
202 Of course, for natural saturated clays characterised by a rigid porous skeleton, this observation
203 is only valid on intact specimens, given that the condition of rigid skeleton would not be valid
204 for remoulded clays, due to the breakage of the rigid natural inter-aggregate bonds. The
205 compression of an elastic fragile porous matrix containing pores of various diameters
206 progressively collapsed according to their size has been modelled by means of the boundary
207 elements method in Cerrolaza and Delage (1997), who observed a satisfactory shape of the
208 compression curve.

209

210

211 **The compression of a highly plastic marine soil**

212 The previous mechanism can only work when the soil skeleton behaves as a rigid porous
213 matrix, like in sensitive clays, that are known to have a Yield Stress Ratio (YSR) larger than 1
214 thanks to some bonding agents (Locat et al. 1984) that ensure their stability at intact state and
215 are destroyed by remoulding. This is also true for dry compacted soils thanks to the inter-
216 aggregate bonding at low water contents, that results from both capillary actions and physico-
217 chemical effects. This mechanism cannot however be extended to plastic compressible clays,
218 as seen in Figure 9 that presents the PSD curves obtained at different levels of compression of
219 a highly plastic clay ($w_p = 52$, $w_L = 139$ and $I_p = 87$) from the Gulf of Guinea (De Gennaro et al.
220 2005, Le 2008). The Figure does not present any superimposition of the PSD curves at various
221 vertical loads, and one cannot conclude that the same ordered pore collapse mechanism
222 occurs here as well. Note however that the SEM photo presented in Figure 10 shows a
223 microstructure not so different from that of the Saint-Guillaume clay (Figure 1). The Gulf of
224 Guinea clay is characterised by large pores (diameter often larger than $1 \mu\text{m}$) with clayey
225 bridges linking together aggregated clay particles, with an overall microstructure somewhat
226 similar to a honeycomb microstructure. The PSD curves present different shapes, with
227 superimposition only observed on small pores (entrance pore radius smaller than $0.06 \mu\text{m}$).
228 Indeed, most of the pores are affected by loading under 50 kPa, showing that almost all
229 microstructure levels are affected by compression. Note that some micro-organisms have
230 been observed in this clay, as also done by Tanaka and Locat (1999) in some plastic Japanese
231 marine clays. Tanaka and Locat however concluded that the effect of microorganisms was not
232 significant at low stress, due to the mechanical resistance of the shells, compared to that of
233 the surrounding assembly of clay particles. This is thought to be valid in our case, at least when
234 applying the 50 kPa load, during which a significant shift of the PSD curve was already
235 observed. It is true that these effects would deserve a specific investigation at larger stress.

236 The main difference compared to Eastern Canada sensitive clays is that the clay fraction of the
237 Guinea clay is characterised by a significant content of smectite (probably contained in illite-
238 smectite species, common in clays of detritic origin occurring in complex natural media,
239 Thomas et al. 2005), that explains the high plasticity of the clay. It is hence necessary to
240 consider how smectite minerals behave under compression, based on literature data
241 observed on montmorillonite suspensions. The area of the Gulf of Guinea is a place where the
242 sedimentation of clay particles provided by the river Niger still occurs in a saline environment
243 (around 35g/l of salt, mainly NaCl), in quiet conditions, in such a way that the comparison with
244 artificial clay suspensions is reasonable. Tessier et al. (1991) performed an investigation of
245 the change in microstructure of saturated clay suspensions submitted to increased suction by
246 means of the combined use of SEM, transmission electron microscopy (TEM) and Small Angle
247 X-ray Scattering (SAXS). As observed by Marcial et al. (2002) on bentonite suspensions, the
248 effect of increased suction in saturated conditions, in terms of global volumetric changes, is
249 reasonably comparable to that of oedometer compression loading, showing that the
250 contribution of the deviatoric part of the stress state in the oedometer has no significant effect
251 in terms of global macroscopic volumetric response.

252 The SEM photo of a Na^{++} smectite suspension submitted to increase suction (3.2, 100 kPa and
253 1 MPa) presented in Figure 11 shows an initial honeycomb microstructure ($w = 369\%$ under
254 3.2 kPa) characterised by large pores (up to $2 \mu\text{m}$) separated by walls made up of the stacking
255 of montmorillonite layers. The TEM photo of Figure 12 shows in more details the structure of

256 the wall, made up here of 30 – 50 elementary layers in face to face arrangements. The SEM
257 photos clearly show how the pores are compressed when increasing suction (the changes in
258 height of the photos corresponds to the change in height due to compression), with elongated
259 pore shapes observed under 100 kPa. Better understanding of the change in microstructure
260 are provided by SAXS measurements, who are able to give the changes in thickness of the
261 walls as well as the inter-layer distances. Actually, it is known for long time (e.g. Mooney et al.
262 1952, Norris 1954, Méring & Glaeser 1954 and, more recently Berend et al. 1995 and Ferrage
263 et al. 2005) that the inter-layer distance between montmorillonite layers within stacks
264 depends on the number of layers of water molecules adsorbed along the montmorillonite
265 minerals, which in turn depends of the suction applied. Typically, to give orders of magnitudes,
266 various hundreds of MPa correspond to one adsorbed layer (1W hydration) with an inter-basal
267 spacing of 12.6 Å, various tens of MPa correspond to two layers (2W hydration) with an inter-
268 basal spacing of 15.6 Å whereas three layers (3W hydration) with an inter-basal spacing of
269 18.6 Å appear for suctions smaller than 7 – 9 MPa. Tessier (1991) investigated in more details
270 the change in the wall configuration of a Ca⁺⁺ montmorillonite and provided the changes in
271 wall thickness and number of layers of the wall with respect to suction. He showed that, at
272 lower suctions (3.3 and 100 kPa), the walls were made up of 55 layers with an inter-basal
273 spacing of 18.6 Å (3W hydration), whereas this number increased up to 225 with the same 3W
274 hydration under 1000 kPa. At higher suction, the inter-basal spacing reduced to 15.6 Å (2W
275 hydration) whereas the number of layers of the walls increased to 400. There is unfortunately
276 no such data for Na⁺⁺ montmorillonite, but SAXS data indicate that the compression
277 mechanisms simultaneously involve the decrease in the size of pores, as observed in SEM, and
278 a change in the configuration of the wall, with a significant enlargement of the number of
279 layers with stress. Although the link with the changes in PSD curves is not obvious, one can
280 suspect that the changes in microstructure that govern the shape of the various PSD curves
281 observed in Figure 9 are affected by this kind of phenomenon with a thickening of the walls
282 of the honeycomb microstructure and, in the range of stress considered (up to 1 MPa), a 3W
283 hydration with three layers of water adsorbed along the montmorillonite layers within the
284 walls. Note also that the water molecules located in the large pores close to the surface of the
285 walls may also be affected by Diffuse Double Layer effects resulting from the electrical deficit
286 of the montmorillonite layer along the wall. The main difference of this compression
287 mechanism involving smectite minerals, compared to the previous case corresponding to a
288 porous fragile matrix typical of low plastic soils, is linked to the reactivity of smectite minerals
289 with water, that changes the nature of the clay matrix under compression, with the combined
290 effect of decrease in pore size and modifications in the wall structure due to both change in
291 the nW hydration within the wall, and in number of layer that constitute the walls.

292 **The swelling of heavily compacted bentonites**

293 Various Engineered Barriers Systems (EBS) are foreseen as barrier and plug for galleries in
294 geological radioactive waste disposal. EBS can be used as compacted bricks, pellets or granular
295 bentonite and placed around the waste canisters to provide a supplementary isolation layer
296 with respect to that provided by the host rock. EBS are made up of relatively pure
297 montmorillonites, like those described in Table 1 (FoCa7 from France, Kunigel from Japan,
298 MX80 from Wyoming, USA and GMZ from China), considered as possible components of EBS
299 in various countries. As seen in the Table, bentonites are characterised by significantly high
300 liquid limit (up to 520 for MX80), specific surface (up to 700 m²/g for MX80) and cation

301 exchange capacity (up to 73.2 mEq/100g for Kunigel). The old findings on bentonite hydration
302 mentioned above appeared also valid for compacted bentonites, as shown in Figure 13, from
303 a SAXS investigation of hydration conducted by Saiyouri et al. (2004). The suction limits
304 corresponding to changes in nW hydration are clearly defined with the transition 1W – 2W
305 occurring at 50 MPa and that between 2W and 3W at 7 MPa. It is important to also note the
306 decrease in number of layers per platelet that starts at 350 above 50 MPa, decreases to 150
307 at the 2W – 3W transition at 7 MPa prior to reduce at 10 layers below 3 MPa. Note also that
308 the constitution of a 4th layer occurs at low suction below 100 kPa. Interestingly, these
309 hydration data follow the same trend as what was observed by Tessier (1991) on
310 montmorillonite suspensions along a drying path, with 3W hydration under 1 MPa and 2W
311 hydration under 10 MPa, coupled by an enlargement in thickness of the wall from 55 layers at
312 100 kPa to 225 at 1 MPa and 400 layers at 10 MPa. Saiyouri et al. (2004) observed similar
313 trends on other compacted bentonites than MX80. Based on this, one can then consider that
314 the microstructure of the compacted Kunigel bentonite hydrated a low suction with free
315 swelling, as observed in Figure 14 (Cui et al. 2002), has walls made up of around 10 layers with
316 4W hydration and an inter-basal thickness of 21.6 Å, resulting in an estimated thickness of
317 around 21 nm. These walls delimit pores of various micrometres diameters, comparable with
318 those observed in the montmorillonite suspension of Figure 11.

319 The nW hydration mechanism also helps better understanding the changes in PSD curve of
320 MX80 compacted samples at same density but different water content, presented in Figure
321 15. Both samples have a void ratio e close to 1, but the wetter sample at a water content $w =$
322 28.5% has a suction of 2 MPa, whereas that at $w = 12.5\%$ has a larger suction of 30 MPa. Both
323 curves exhibit a bi-modal PSD allowing to distinguish the (comparable) inter-aggregate
324 porosity, with entrance pore radius close to 2 μm , from the intra-aggregate porosity where
325 more difference is observed, with an entrance radius of 19 nm for the drier sample at $s = 30$
326 MPa, compared to 10 nm for the wetter sample at $s = 2$ MPa. Another significance difference
327 is observed in the infra-porosity, defined in Delage et al. (2006) as corresponding to the pores
328 with entrance radius smaller than 3.5 nm, too small to be intruded by mercury in MIP. The
329 infra-porosity of the wetter sample (with $e_{\text{infra}} = 0.156$) is smaller than that of the drier sample
330 ($e_{\text{infra}} = 0.363$).

331 The hydration mechanisms described in Figure 13 indicates that, under 2 MPa, the wetter
332 sample is in a 3W hydration state with inter-basal spacing of 18.3 Å and thin platelets made
333 up of around 10 layers, resulting in an estimated thickness of 18 nm. Based on the brick model,
334 in which clay platelets within the aggregate are compared to a brick assembly, the brick
335 thickness can be estimated as the average entrance diameter detected in MIP (see Yven et al.
336 2007 and Menaceur et al. 2016). This provides an estimated thickness of 20 nm (for an
337 entrance radius of 10 nm, see Figure 15), in good agreement with the 18 nm estimation
338 derived from the hydration mechanism. At 30 MPa, the drier sample is in a 2W hydration state
339 with 15.3 Å inter-basal spacing, and much thicker particles of around 250 layers, resulting in
340 an average thickness of 38 nm. With an entrance pore radius of 19 nm (Figure 15), the
341 thickness estimated from the brick model and MIP is 38 nm, quite in good agreement with the
342 estimation from the hydration mechanism.

343 The difference in infra-porosity can also be commented. The amount of inter-layer water
344 molecules adsorbed along the montmorillonite layers is higher in the wetter sample, with 4W
345 hydration, compared to that in the drier sample at 2W hydration. Assuming that the

346 montmorillonite surface along which water molecules are adsorbed in both cases is the same
347 because they have the same density, one can conclude that there are twice more water
348 molecules adsorbed in the wetter case, a proportion which corresponds well with the ratio of
349 infra-porosity (0.156 in the drier case and 0.363 in the wetter case). All these observations
350 show the validity of the conclusions drawn from SAXS investigations on the hydration
351 mechanisms in compacted bentonite, and how MIP data can be further interpreted to get a
352 more complete understanding of the effects of changes in water content on the
353 microstructure of compacted bentonites.

354 **Volume changes in swelling claystones**

355 Particular attention has been paid in the last decades on the hydro-mechanical behaviour of
356 claystones considered as possible host-rocks for deep geological waste disposal in Europe, i.e.
357 the Callovo-Oxfordian claystone in France and the Opalinus Clay in Switzerland. A detailed
358 investigation on the volume changes in free conditions of the Callovo-Oxfordian (COx)
359 claystone under controlled suction changes (Menaceur et al. 2016) demonstrated that the
360 hydration mechanisms commented above also play a role within the montmorillonite minerals
361 present in illite-smectite mixed layer clay minerals of the COx clay matrix (see also Delage et
362 al. 2014). This matrix (with more than 45% < 2 μm) can be observed in the SEM photo of a
363 freeze-fractured plane of a COx sample hydrated at zero suction, presented in

364 Figure 16 (Menaceur 2014). The clay matrix is clearly apparent with a well-marked bedding
365 plane close to horizontal, and some footprints of detritic minerals (25% calcite grains, 20%
366 quartz grains, see Gaucher et al 2004) that have been extracted from the fracture plan during
367 freeze-fracturing (after ultra-quick freezing of small sticks of claystone in slush nitrogen at –
368 210°C, see Delage et al. 2006). Pyrite crystals are also observed. The photo also clearly shows
369 that (free) swelling under zero suction (around 6%) resulted in the development of large sub-
370 horizontal cracks following the bedding plane, with 4 μm width and more than 10 μm length.
371 Note that those cracks, also detectable by MIP, are typical of fully hydrated specimens and
372 have not been detected in specimens extracted and kept at initial water content (Menaceur
373 et al. 2016), showing that they are not due to stress release. Actually, as shown by Bornert et
374 al. (2010) based on Digital Image Correlation on Environmental SEM images, swelling is also
375 related, besides those mesoscopic cracks, to the development of smaller cracks and to some
376 bulk deformation within the clay matrix.

377 The PSD curves of three freeze-dried COx specimens at suctions of 34 MPa (initial state, as
378 received in the lab), 150 MPa, 331 MPa together with that of a dry specimen (48h in an oven
379 at 105°C) are presented in Figure 17, together with the corresponding water retention curve,
380 represented in terms of volume changes (measured by hydraulic weighing on waxed
381 specimens) with respect to suction changes (Wan et al. 2013, Menaceur et al. 2016). One
382 observes that there is a decrease in the average entrance pore diameter from 32 nm at 34
383 MPa suction down to 28 nm for both specimens under 150 and 331 MPa suctions. The
384 diameter then decreases to 21 nm in the dry state, in accordance with the data of Boulin et
385 al. (2008). Considering that, based on the brick model and a simplified model of the
386 microstructure of the COx claystone (see Yven et al. 2007 and Menaceur et al. 2016), the
387 average entrance pore diameter provides a reasonable order of magnitude of the average
388 platelet thickness, one can deduce from the dry state (with 0.96 Å inter-basal spacing) that the

389 average number of layers per platelet is equal to $21/0.96 = 22$. By considering a smectite
390 proportion of 50 – 70% (Yven et al. 2007) in the mixed-layered illite-smectite platelets and
391 considering 1W hydration (12.6 Å inter-basal spacing) at 150 and 330 MPa, one estimate an
392 average platelet thickness of 24.3 – 25.6 nm, in reasonable agreement with the average
393 entrance pore radius of 28 nm detected in MIP. Similarly, 2W hydration (15.6 Å inter-basal
394 spacing) for specimens at 34 and 9 MPa provides an average thickness of 27.6 – 30.2 nm,
395 comparable to the 32 nm average entrance diameter detected by MIP. This good
396 correspondence is illustrated in Figure 18, that shows the measured and estimated platelet
397 thicknesses along the drying path. An interesting similarity is observed with the data
398 presented in Figure 13, showing that the same hydration mechanisms occurs along the faces
399 of the smectite layers contained in the COx clay matrix. This feature is further confirmed by
400 considering the wetting path between 34 and 9 MPa, along which no significant volume
401 change is observed in the water retention curve, in spite of a significant decrease of 25 MPa
402 in suction. It is shown in Menaceur et al. (2016) that both PSD curves at 34 and 9 MPa are
403 similar, with comparable entrance pore diameter (as also observed in Figure 18), indicating
404 that the COx claystone is probably in a 2W hydration state at natural state.

405 MIP investigation also provided better understanding of the significant swelling (around 6%)
406 occurring below 9 MPa (up to full saturation), a suction value below with 3W hydration starts,
407 followed by 4W hydration at zero suction. The micro-cracks observed in Figure 16 at full
408 saturation have also been detected by MIP by Menaceur et al. (2016), with some detected
409 pores at 12 and around 0.1 µm.

410 Although they are quite different in nature, the same mechanisms govern the hydration of
411 smectite suspensions, compacted bentonite and a swelling claystone like the COx claystone.
412 In the latter case, this shows that the level of energy governing crystalline swelling in
413 claystones is strong enough to break the long term diagenetic bonds that have developed
414 during the long geological history of this claystone (158 millions years old). Swelling occurs
415 both at the crystalline nanoscale level, with the ordered adsorption of layers of water
416 molecules along the smectites layers within the platelets, and larger micrometre scale, by the
417 development of micro-cracks that are responsible of most of the swelling at low suction (see
418 Wan et al. 2013 and Menaceur et al. 2016). Note also that this significant reactivity of the
419 smectite phase in swelling claystone is responsible of the remarkable self-sealing properties
420 observed in both the Callovo-Oxfordian claystone and the Opalinus clay, in which open cracks
421 are progressively filled by hydrated smectites that finally bring back the permeability of the
422 cracked claystone to that of the intact rock (e.g. Davy et al. 2007, Menaceur et al. 2015).

423 **Concluding remarks**

424 The nano and micro-scale mechanisms described in this work to interpret the macroscopic
425 volume changes of various geomaterials result from long-term investigations carried out on
426 various clayey soils and rocks specimens as different as low and high plasticity compressible
427 soils, smectite suspensions, compacted bentonites and swelling claystones. Two main
428 mechanisms have been identified. In low plastic saturated and unsaturated materials, like
429 sensitive clays or dry compacted silts, an aggregate microstructure was evidenced, and it was
430 shown that compression is due to the progressive and ordered collapse of the inter aggregates
431 pores, from the larger to the smaller. The soil skeleton hence reacts as an elastic fragile porous

432 matrix, in which a given level of stress collapses a pore population whose diameter can be
433 related to the stress applied. Also, it was shown that aggregates are able to resist to the action
434 of stress currently applied in common geotechnical practice, and up to 1.5 MPa in the case of
435 sensitive clays from Eastern Canada. Another consequence of the aggregate stability in dry
436 compacted soils is that compression at constant water content does not result in any change
437 in suction.

438 The mechanisms of smectite hydration and compression that have been evidenced by Soil
439 scientists in smectite suspensions based on X-ray diffraction techniques appeared to be
440 relevant to better understand the compression of a highly plastic clay from the Gulf of Guinea,
441 that is governed by quite different mechanisms than the above-mentioned ones. Indeed, the
442 high reactivity of smectite minerals result in more complex mechanisms upon compression,
443 with no clear collapse of any pore population, but a decrease in size of the pores coupled to
444 both a reorganization affecting the thickness of the walls and the nW hydration of the platelets
445 they are made of. Interestingly, nW hydration steps also govern the hydration of compacted
446 bentonite, and help better understanding the effects of changes in water content on their
447 microstructure. They are also valid when describing the volume changes of swelling claystones
448 due to crystalline swelling, and their high-energy level appears able to break, upon hydration,
449 the diagenetic long term bonds that are at the origin of the high cohesion that they have
450 gained and that progressively transformed the clay sediment in a clay rock.

451 Among other things, the investigations described in this paper didn't address the effects of
452 pore water salinity on the macroscopic behaviour of intact clayey soils and rocks. Such effects,
453 particularly significant in the smectite fraction of plastic clayey materials, can be described by
454 using the Diffuse Double Layer theory, that provides some modelling of the effects of both the
455 valence and the concentration of dissolved ions. This is however mainly valid in clay
456 suspensions, and it seems that accounting for such effects in structured clays is still a pending
457 problem, that obviously needs further investigation.

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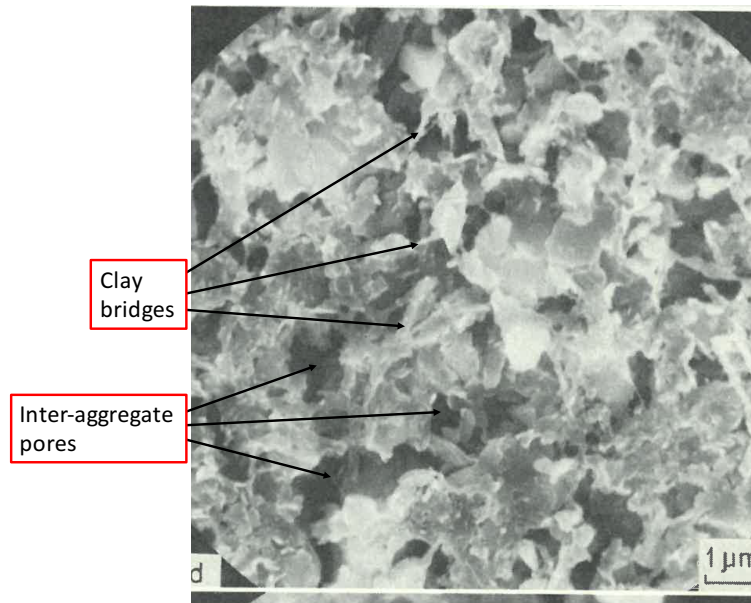
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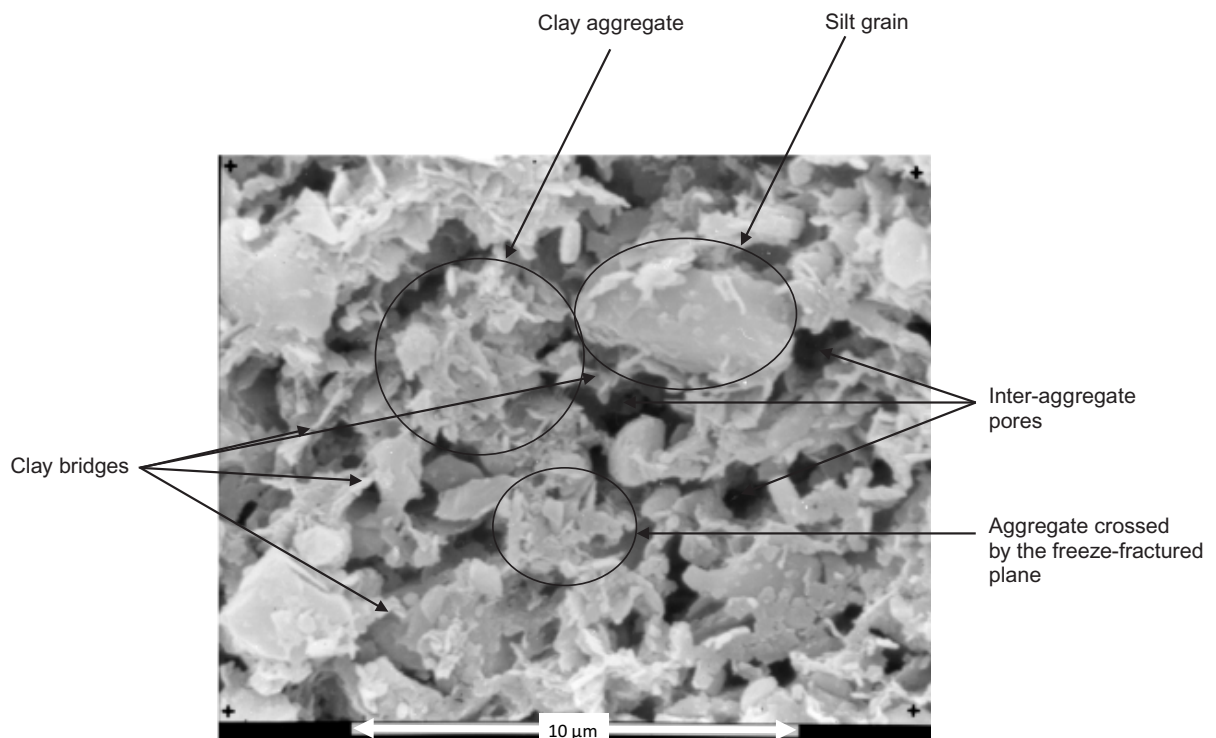
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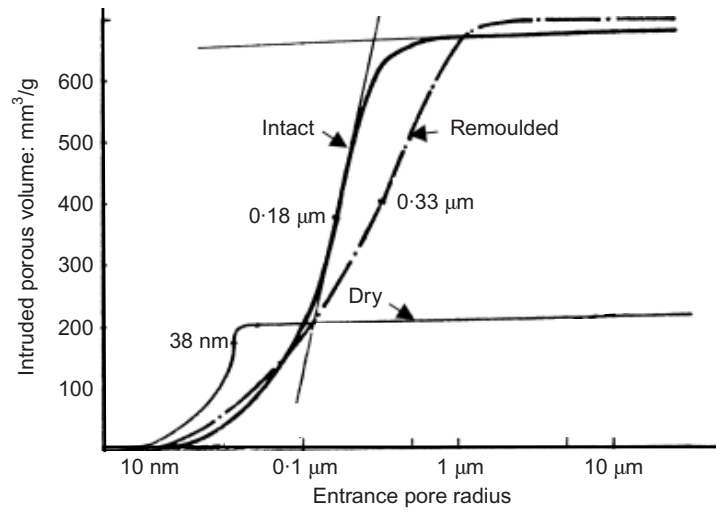
587 *Figure 1. Scanning electron microscope photo of the Saint-Guillaume Eastern Canada sensitive clay.*



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589 Figure 2. Scanning electron microscope photo of the Saint-Marcel Eastern Canada sensitive clay (Delage 2010)

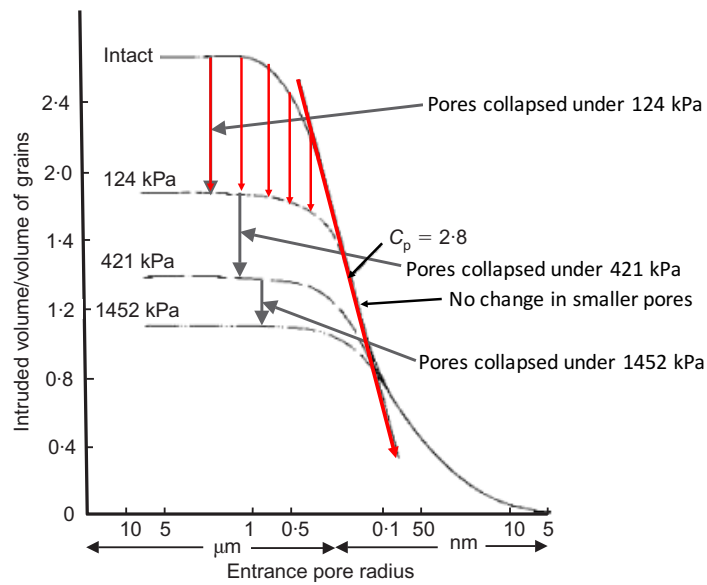
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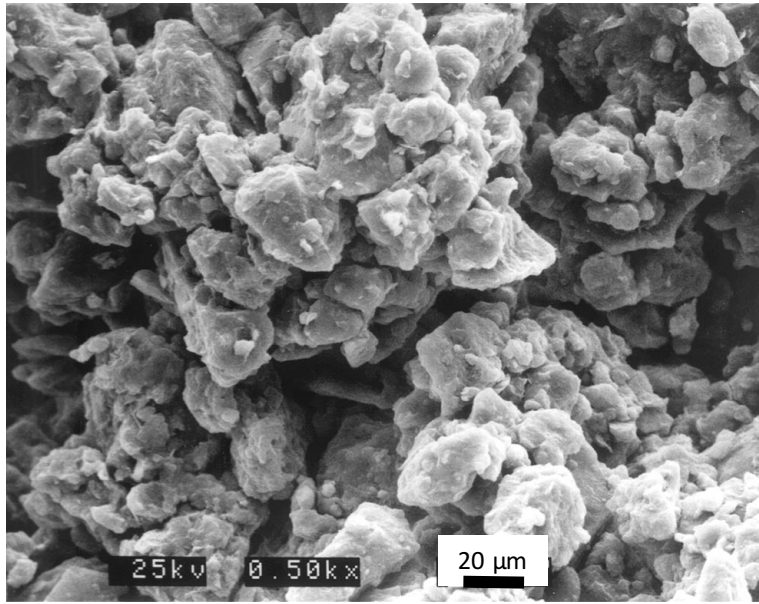
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593 Figure 3. Pore size distribution curves of the Saint Marcel clay, intact, remoulded and dry states (Delage 2010).



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595 Figure 4. Pore size distribution of the Saint Marcel clay, intact and submitted to oedometer compression under
596 124, 421 and 1452 kPa (Delage and Lefebvre 1984)

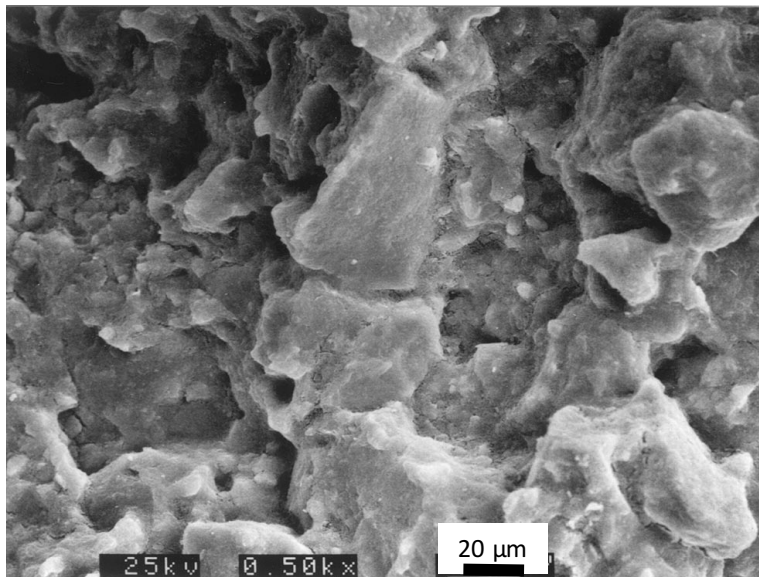


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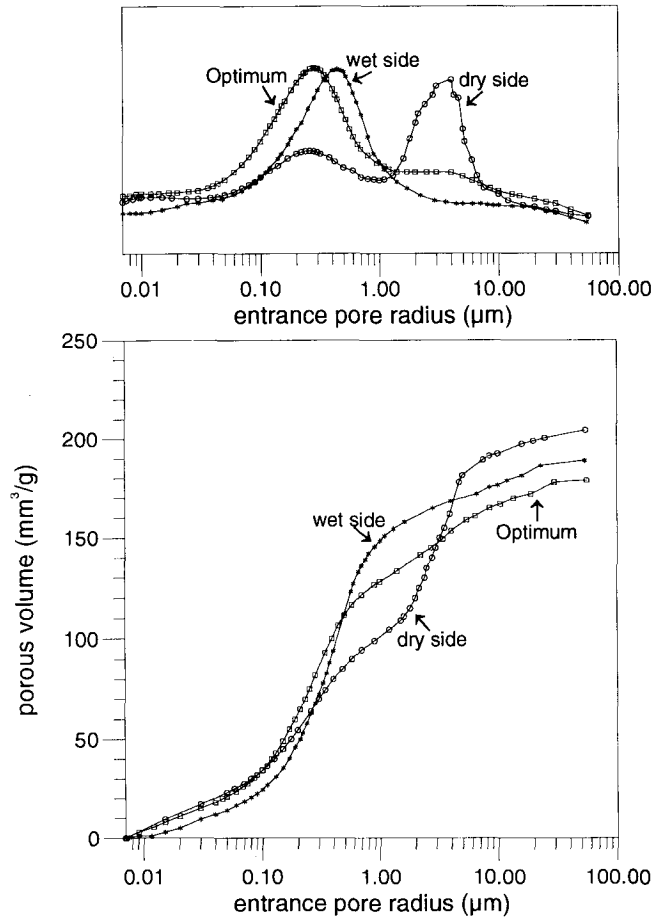
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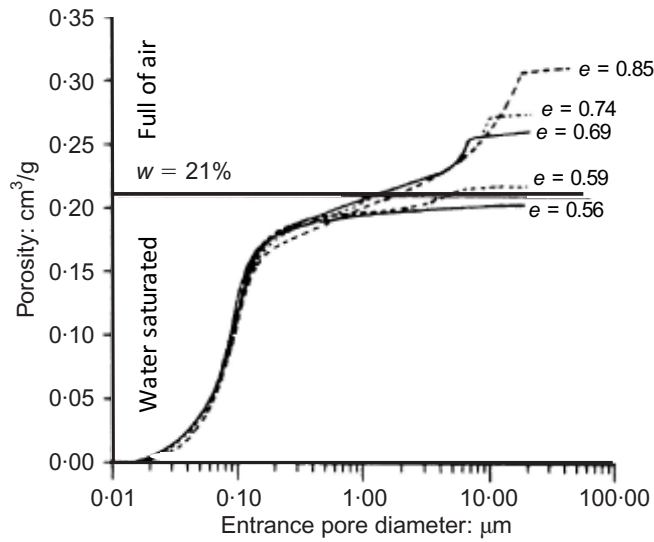
602 *Figure 5. SEM observation of compacted Jossigny silt: a) dry side of optimum; b) wet side of optimum.*

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605 *Figure 6. PSD curves of compacted Jossigny silt (Delage et al.1996).*



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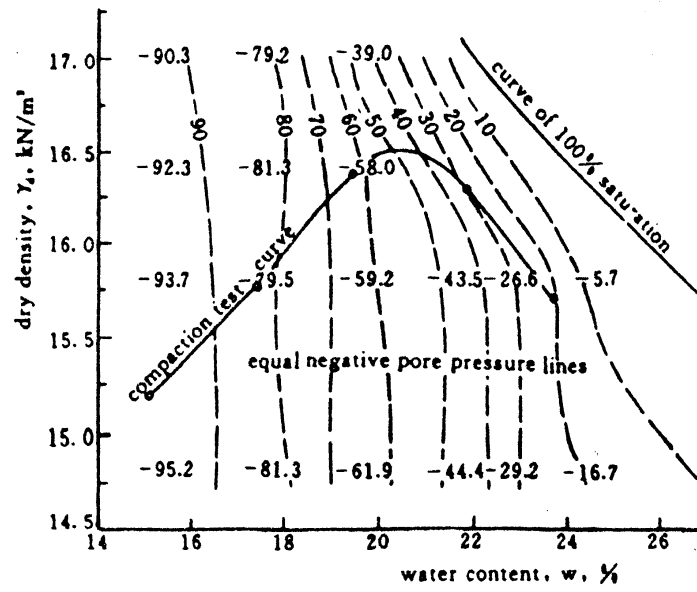
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608 *Figure 7. PSD curves of a compacted soils at various densities and constant water content of 21%.*

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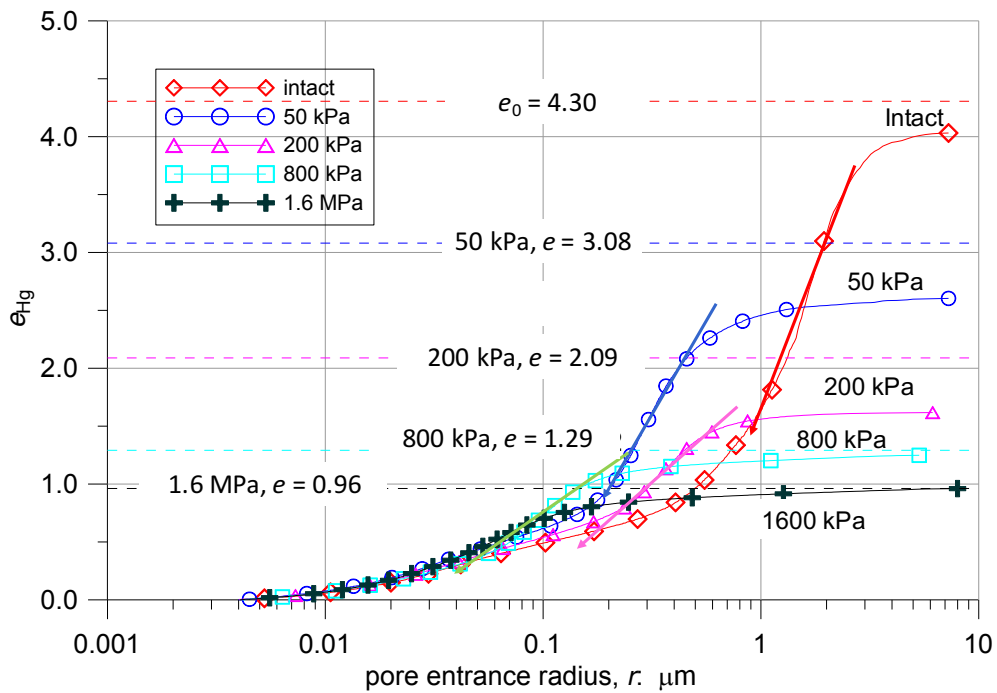
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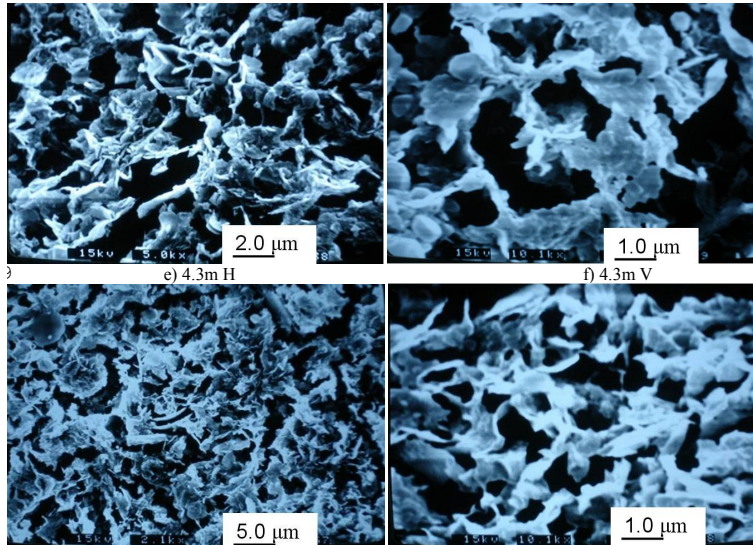
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613 *Figure 8. Changes in suction with density, at constant water content (Li et al. 1995)*



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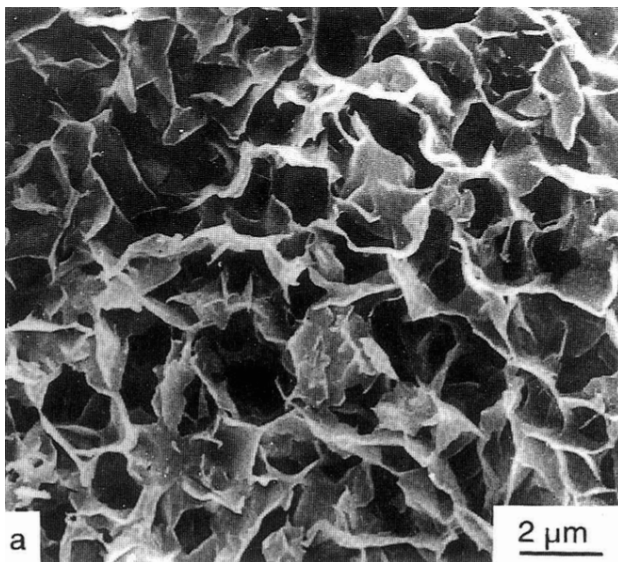
615 *Figure 9. Changes in pore size distribution curve of a clay from Gulf of Guinea compressed at various vertical*
616 *stresses in the oedometer (Le et al. 2008).*



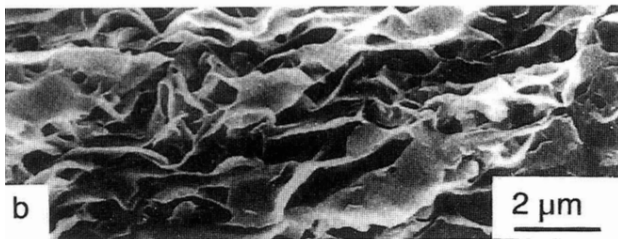
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618 *Figure 10. SEM photo of a highly plastic clay ($I_p = 100$) from the Gulf of Guinea (Le 2008)*

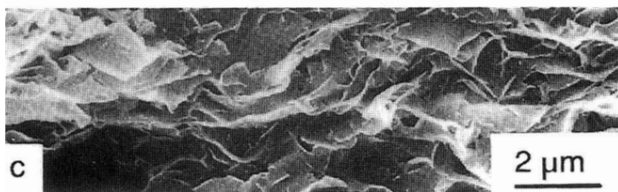
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$w = 369\%$
Suction 3.2 kPa



$w = 114\%$
Suction 100 kPa



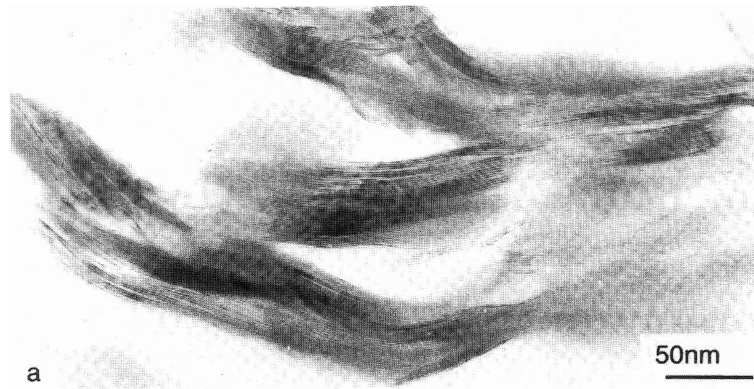
$w = 82\%$
Suction 1 MPa

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621 *Figure 11. SEM photos of Na⁺⁺ montmorillonite suspensions submitted to various suctions (w = 369% under a*
 622 *suction s = 3.2 kPa, w = 114% under s = 100 kPa and w = 82% under s = 1 MPa) (Tessier 1991).*

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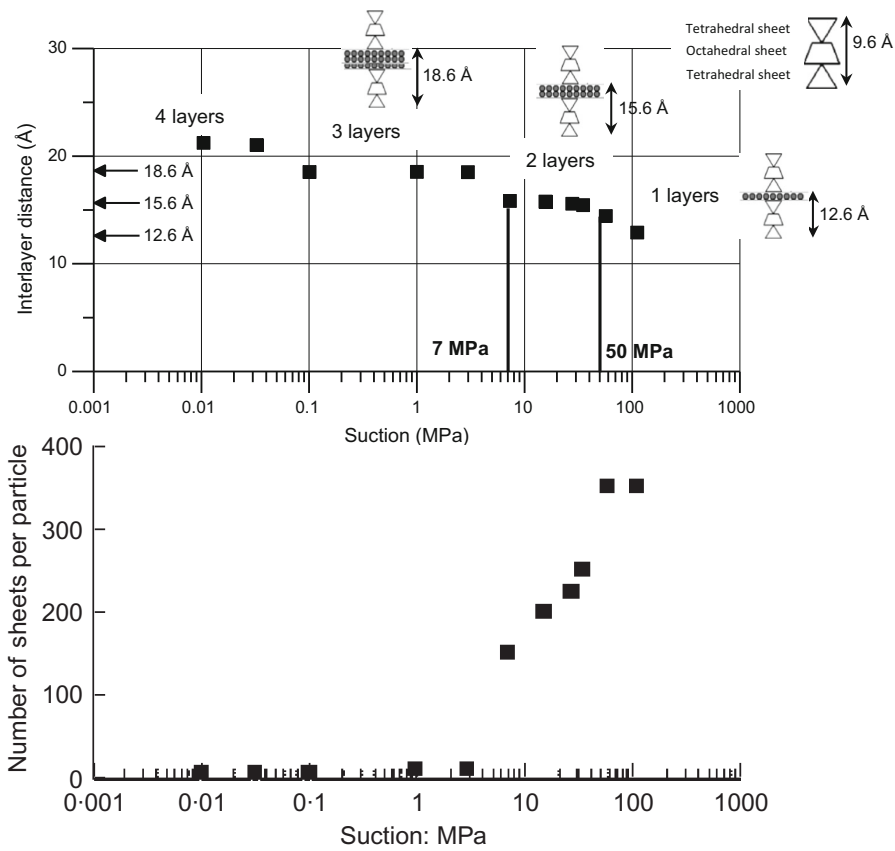


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626 *Figure 12. TEM photo of the wall of montmorillonite suspension (Tessier 1991).*

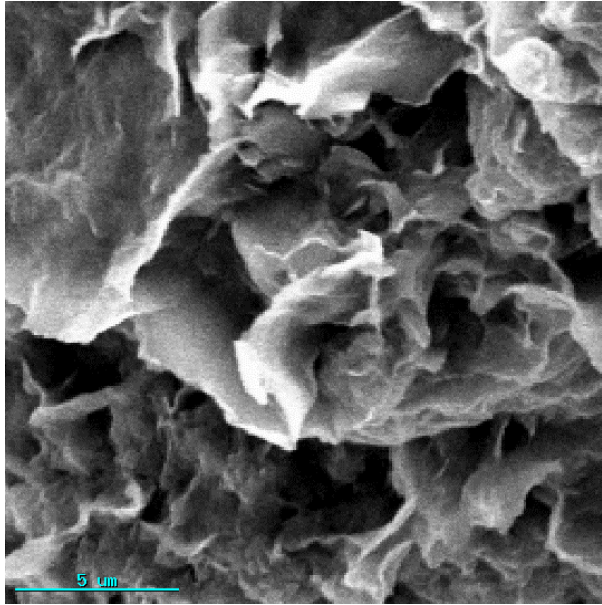
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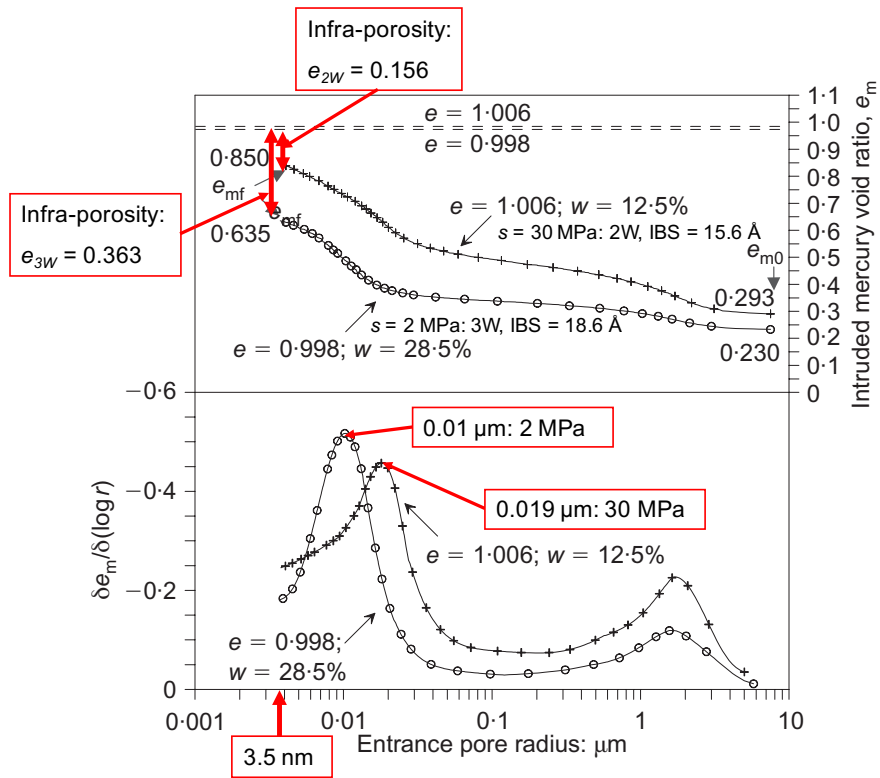
630 *Figure 13. Hydration mechanisms in a compacted MX80 bentonite (after Saiyouri et al. 2004).*



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632 *Figure 14. Fully hydrated and swollen Kunigel compacted bentonite (Cui et al. 2002).*

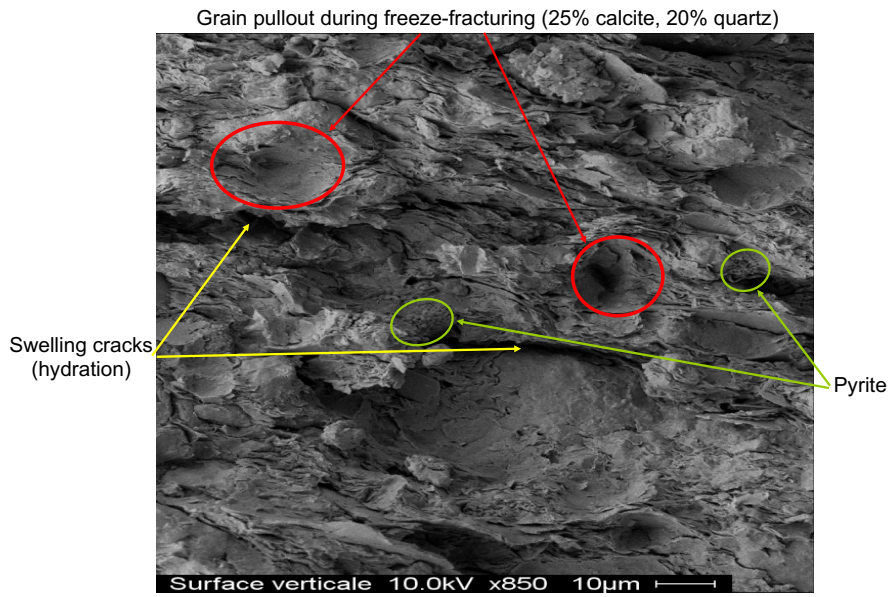
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635 *Figure 15. PSD curves of MX80 compacted samples at same density and different water contents (After Delage*
 636 *et al. 2006).*

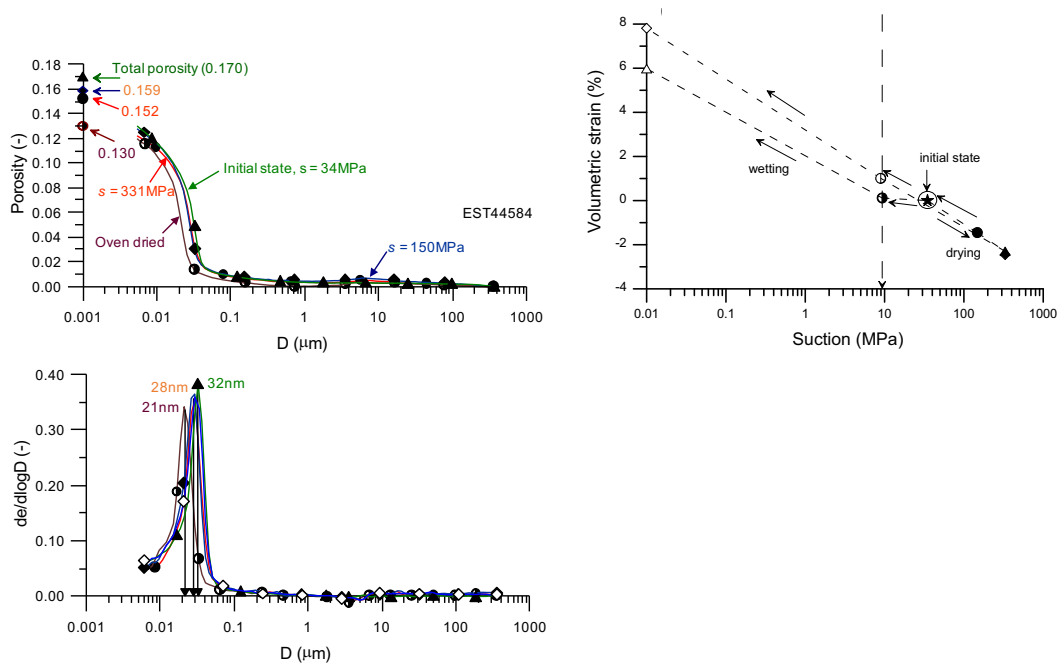
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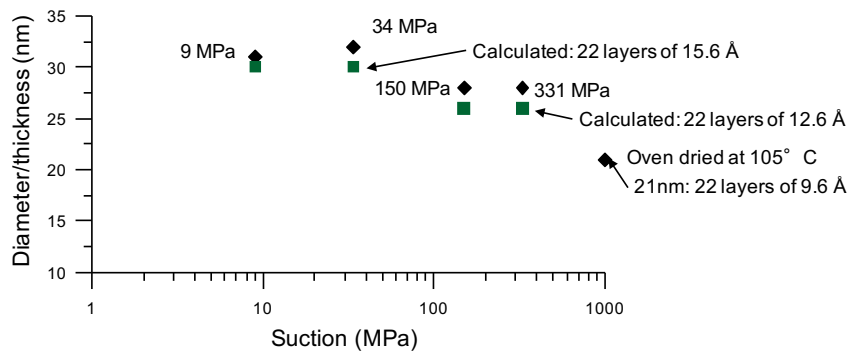
640 *Figure 16. SEM observation of a freeze-fractured plane of the Callovo-Oxfordian claystone (Menaceur 2014).*



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642 *Figure 17. PSD curves of COx claystones specimen from initial state along a drying path (suctions of 34, 150, 330*
 643 *MPa and dry state), after Menaceur et al. (2016).*

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645

646 *Figure 18. Measured and calculated platelet thicknesses along the drying path (suctions of 34, 150, 330 MPa*
 647 *and dry state), after Menaceur et al. (2016).*

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