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**Title:**

**Influence of heterogeneities of density on the hydromechanical behaviour of pellet-based bentonite materials in imbibition experiments**

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**Abstract:**

An experimental investigation on the hydromechanical behaviour of pellet-powder mixtures, candidate materials for sealing the galleries in radioactive waste disposal concepts, was described. Imbibition experiments were performed using a specially-designed column, with square cross-section and glass sides allowing pictures to be taken by a camera. Relative humidity and swelling pressure were measured at different elevations of the column. Three mixtures with an identical pellet volume fraction and various powder volume fractions were used to study the influence of heterogeneities of density on the hydromechanical behaviour of the mixture under repository conditions. Results demonstrated that hydration of the material was dominated by water vapour flux, with pellets and inter-pellet pores not at hydraulic equilibrium. In mixtures with low powder content, the evolution of swelling pressure was controlled by contact forces between pellets. The material progressively lost the initial granular structure upon hydration. Note that hydration was made by imbibition, instead of injecting liquid water at high pressure, to avoid unrealistic liquid water flow rates in the initial large inter-pellet voids. In addition, sensors four times larger than the pellets were used to measure a representative value of swelling pressure.

**Keywords:**

Bentonite, Pellet mixture, Imbibition experiment, Swelling pressure, Relative humidity, Water migration

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## 1. Introduction

Bentonite materials have been considered as sealing materials in radioactive waste disposal concepts of the French National Agency for Radioactive Waste Management (Andra) for their low permeability, good radionuclide retention capacity and ability to swell upon hydration (Andra, 2005). In the gallery, the bentonite swelling in the radial direction is prevented by the host rock and a swelling pressure develops. This latter contributes to the sealing of fractures in the excavation damaged zone. Experimental studies demonstrated that the permeability and swelling pressure of a bentonite material in fully-saturated state were related to its dry density (Karnland et al., 2006; Lloret and Villar, 2007; Wang et al., 2012; Kaufhold et al., 2015).

Owing to operational convenience, pellet-based bentonite materials have been regarded as candidate materials for the swelling clay plug and backfill (Imbert and Villar, 2006; Dixon et al., 2011; Bosgiraud and Foin, 2016; Sánchez et al., 2016; Darde et al., 2018; Bernachy-Barbe et al., 2020; Sellin et al., 2020). In these studies, the galleries were expected to be filled with pellet-based mixture. Screw auger can be used for this purpose as demonstrated by Andra in a full-scale experiment (Noiret et al., 2016). During the installation of the granular mixture in the gallery, heterogeneities of dry density can develop. Obtaining a homogeneous mixture was found difficult in a full-scale experiment (Noiret et al., 2016) as well as in the laboratory Molinero Guerra et al. (2017).

Following its installation, the material will be progressively hydrated by the pore water of the host rock. Upon hydration, experimental studies at the laboratory scale (Hoffmann et al., 2007; Darde et al., 2020a) and discrete element method (DEM) simulations (Darde et al., 2021, 2020b) suggested that the evolution of swelling pressure depended on the arrangement of the particles (pellets and powder grains). This initial granular structure became less obvious during hydration (van Geet et al., 2005; Hoffmann et al., 2007; Molinero Guerra et al., 2018a; Darde et al., 2020a; Villar et al., 2021).

In order to demonstrate the influence of the initial granular structure and progressive homogenisation of the mixture on the performance of the seals in the repository, the present work aims at investigating the hydromechanical behaviour of bentonite pellet mixtures in the course of hydration using imbibition columns. A total of three imbibition experiments were performed on three different pellet and powder mixtures, one of them is a candidate sealing material in the French concept for radioactive waste disposal (Noiret et al., 2016), using hydration columns.

Several imbibition experiments performed on pellet materials have been described in the literature (Imbert and Villar, 2006; Molinero Guerra et al., 2018a, 2018b; Bernachy-Barbe et al., 2020). The originality of the tests described in the present work stems from the following points: *(i)* columns with a square cross-section and glass sides were especially designed for the study, allowing a direct observation of the soil texture changes during hydration; *(ii)* hydration was performed through imbibition of water instead of injection of water, to avoid significant water flow rates in the inter-pellet space, reproducing repository conditions more realistically and avoiding an early loss of the granular structure (actually, in the case of Andra's concept, water flows from the host rock, claystone with very low permeability, with negligible pressure during the re-saturation phase); *(iii)* swelling pressure was measured using sensors of diameter four times as large as the pellet diameter, providing rather an average value than local ones as with pellet-sized sensors (Darde et al., 2021). This issue actually is a critical point, since the duration of hydration tests performed on bentonite materials is significant, and the opportunity to perform several tests is thus very limited, so high variability of the measurement can affect the interpretation of results.

## 2. Materials and Method

### 2.1. Pellets and powder

The materials in this study were provided by Laviosa MPC, France. Pellets were made of MX80 sodium bentonite. Its mineral composition is given in Table 1. Its cation exchange capacity (CEC), specific surface area ( $S_{SA}$ ), and amount of  $Na^+$  and  $Ca^{2+}$  cations in interlayer space are presented in Table 2. Pellets had a subspherical shape, with a cylinder-shaped part and two spherical caps at the top and

50 bottom (Figure 1). The pellet's properties are presented in Table 3; additional data can be found in  
 51 Molinero Guerra et al. (2017) and Darde et al. (2018).

52

53 Table 1. Mineral composition of the MX80 bentonite from X-ray Diffractometer. Data from Laviosa MPC, cited by  
 54 Molinero-Guerra et al. (2017).

Mineral	Mass proportion
Pyrite	< 1%
Montmorillonite	80%
Albite	2%
Anorthite	8%
Quartz	4%
Muscovite	4%

55

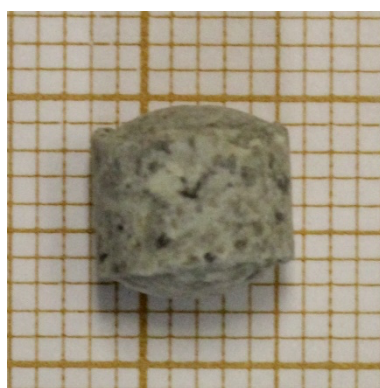
56

Table 2. Properties of the MX80 bentonite. Data from Saiyouri et al. (2004)

CEC	82.3	meq/100 g
S <sub>SA</sub>	522	m <sup>2</sup> /g
Na <sup>+</sup>	80	meq/100 g
Ca <sup>2+</sup>	5	meq/100 g

57

58



59

60

Figure 1. Picture of a pellet. The size of each square on the picture is 1 mm × 1 mm.

61

62

Table 3. Properties of pellet.

Pellet properties	Value
<i>Geometrical properties</i>	
Diameter <sup>a</sup>	7 mm
Height of the cylinder part <sup>a</sup>	5 mm

Total height <sup>a</sup>	7 mm
Curvature radius of the spherical caps <sup>a</sup>	6.5 mm

*Physical properties*

Density of solid particles <sup>b</sup>	2.77 Mg/m <sup>3</sup>
Initial dry density <sup>a</sup>	1.91 Mg/m <sup>3</sup>
Initial void ratio <sup>a</sup>	0.45
Initial suction <sup>a</sup>	89 MPa
Initial water content <sup>a</sup>	12%

a: Darde et al. (2018); b: Saba et al. (2014)

63

64 The powder was obtained by crushing pellets (“Concassé d’expangel SP32” following the  
65 nomination of the producer, Laviosa MPC). The initial suction and water content of the powder were  
66 180 MPa and 3.5%, respectively. The average diameter of the powder grains was 0.65 mm (Molinero  
67 Guerra et al., 2017) which was ~10 times smaller than the pellets.

68 **2.2. Pellets-powder mixtures**

69 Three mixtures of pellets and powder were used in the imbibition experiments. The reference mixture  
70 was a pellet-powder mixture in proportion 70/30, respectively, in dry mass, a candidate sealing material  
71 in the French concept of radioactive waste disposal. The dry density of the mixture,  $\rho_d$ , was 1.50 Mg/m<sup>3</sup>.  
72 The pellet initial volume fraction (volume of pellets / total volume),  $\phi_{p0}$ , was 0.553. The powder initial  
73 volume fraction (volume of powder grains / total volume),  $\phi_{g0}$ , was 0.236. During the installation of the  
74 mixture in the galleries, powder might trickle through the inter-pellet space, causing local  
75 heterogeneities. To investigate the influence of heterogeneities of density resulting from powder  
76 trickling on the mixture behaviour, two other mixtures were studied. These mixtures had a value of  $\phi_{p0}$   
77 identical to that of the 70/30 reference mixture, but different values of  $\phi_{g0}$ . These mixtures are referred  
78 to as “70/15” and “70/0” to indicate the identical pellet volume fraction and reduced powder volume  
79 fraction. The properties of the mixtures are summarised in Table 4.

80 The material is characterised by three levels of porosity. The following terms are used in a broad  
81 sense in the following: “micropores” refers to intra-aggregate pores in the pellets or powder grains;  
82 “macropores” refers to inter-aggregate pores in the pellets or powder grains; “inter-pellet pores” refers  
83 to the large pores between pellets and powder grains.

84

Table 4. Properties of the three pellet-powder mixtures.

Mixture	70/30	70/15	70/0
$\rho_d$ : Mg/m <sup>3</sup>	1.50	1.275	1.05
$\phi_{p0}$ : dimensionless	0.553	0.553	0.553
$\phi_{g0}$ : dimensionless	0.236	0.118	0

85

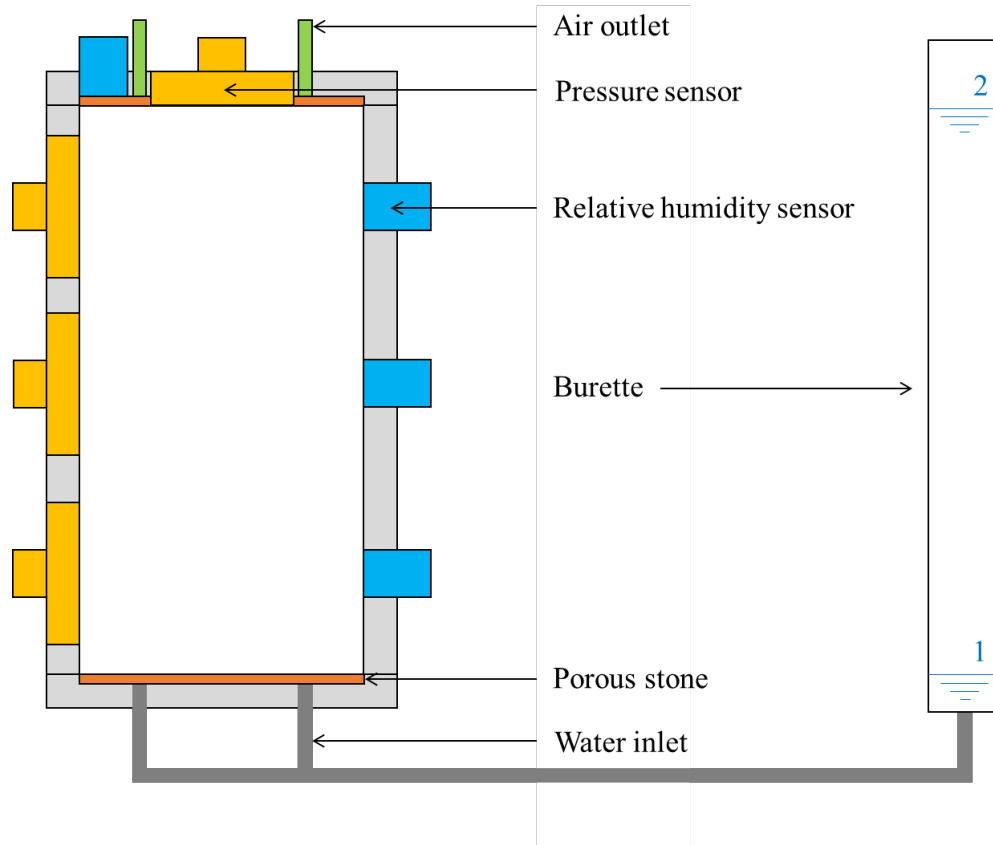
86 **2.3. Imbibition column**

87 The imbibition column was a square cuboid, two faces of which were made of glass. Its inner height  
88 was 120 mm (~17.1 times the initial pellet diameter) and its inner width was 60 mm (~8.6 times the  
89 initial pellet diameter). The glass faces were made of tempered glass of thickness 9 mm. Their  
90 deformation during the test was expected to be negligible such that the imbibition column remained at  
91 constant volume.

92 Four pressure sensors (BER-A-5MP15S from Kyowa, rated capacity of 5 MPa, and accuracy - related  
93 to nonlinearity and hysteresis - of 4% of rated output) were used. Three were located on one side at  
94 different elevations (20 mm; 60 mm; 100 mm) to measure lateral pressure, one was located at the top  
95 face of the column to measure axial pressure. The diameter of all the pressure sensors was 30 mm. The  
96 ratio of sensor diameter to pellet diameter was thus ~4.3. The expected coefficient of variation (standard

102 deviation / mean value) of the measured pressure for this ratio is  $\sim 0.1-0.2$  (Darde et al., 2021). Four  
 103 relative humidity sensors (HIH-4000 Series from Honeywell) were used. Three were located at the same  
 104 heights as pressure sensors, but on the opposite side, one was located on the top face next to the axial  
 105 pressure sensor. The accuracy of the relative humidity sensor was  $\pm 3.5\%$  of the measured value,  
 106 corresponding to less than 5 MPa of suction for 100% RH at 20°C. Room temperature was controlled  
 107 and kept constant ( $20^\circ\text{C} \pm 1^\circ\text{C}$ ).

108 Porous stones were placed at the top and bottom of the column. A camera was used to take pictures  
 109 at different moments of all three samples. A sketch of the column is presented in Figure 2.



105  
 106 Figure 2. Sketch of the imbibition column, with the water levels associated to each phase of the experiment.

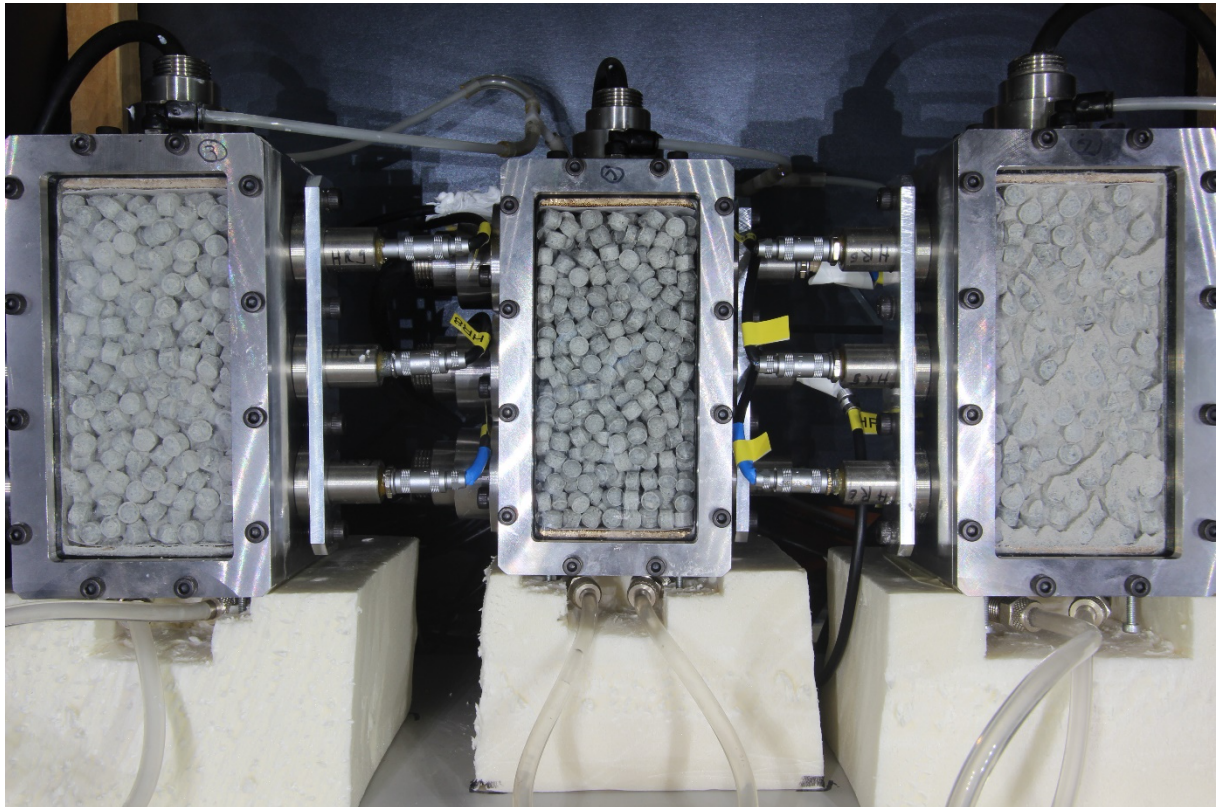
## 107 2.4. Sample preparation

108 During the preparation of the sample, the column was rotated so that a side glass face became the  
 109 bottom face. The mixtures were then prepared by pouring one layer of pellets, then adding powder in  
 110 the inter-pellet voids, in a quantity corresponding to the target proportion. This step was repeated to fill  
 111 the column with the mixture. This procedure was chosen to obtain an initial state as ideally mixed as  
 112 possible (Molinero-Guerra et al., 2017). Filter papers were used to prevent the occlusion of the porous  
 113 stones by the powder grains.

114 Once the column was filled with the mixture, the second glass face was then installed to close the  
 115 column. During this step, no pressure was applied on the mixture.

116 The column was finally carefully rotated back to limit powder trickling in the inter-pellet voids.  
 117 Three experiments are reported in the present work. The three columns were placed side by side to allow  
 118 pictures of all cells to be taken simultaneously by the camera (Figure 3).





119

120 Figure 3. Pellet-powder mixtures in the column following the preparation of the samples. From left to right: 70/15 mixture,  
 121 70/0 mixture, 70/30 mixture.

122 **2.5. Imbibition process**

123 The mixture hydration was performed by imbibition of synthetic water from the bottom of the cell.  
 124 The synthetic water was a solution with the same chemical composition as the pore water of the Callovo-  
 125 Oxfordian claystone from the underground research laboratory of Andra in Bure, France. The receipt of  
 126 the water used in experiments is given in Table 5.

127

128

Table 5. Receipt of the water used in imbibition experiments.

Salt	Concentration
NaCl	1.95 g/l
NaHCO <sub>3</sub>	0.130 g/l
KCl	0.035 g/l
CaSO <sub>4</sub> , 2H <sub>2</sub> O	0.630 g/l
CaCl <sub>2</sub> , 2H <sub>2</sub> O	0.080 g/l
MgSO <sub>4</sub> , 7H <sub>2</sub> O	1.020 g/l
Na <sub>2</sub> SO <sub>4</sub>	0.700 g/l

129

130 In the French concept of radioactive waste disposal, the host rock is a claystone, the permeability of  
 131 which is expected to be below  $10^{-17}$  m<sup>2</sup> (de la Vaissière et al., 2015). Under repository conditions,

132 bentonite-based sealing materials are hydrated by the pore water of the host rock. Considering the low  
133 permeability of the host rock, hydration is expected to be a slow process.

134 In this study, imbibition was performed to reproduce the hydration process under repository  
135 conditions more realistically: because of the presence of large inter-pellet voids, even a small hydraulic  
136 gradient would induce significant water flow rate (Hoffmann et al., 2007) and early loss of the granular  
137 structure as a consequence (because of pellet swelling). The volume of incorporated water was measured  
138 using a burette.

139 Imbibition was started when the sensors measured a constant value of relative humidity during  
140 several days after the sample preparation, indicating that pellets and powder were at hydraulic  
141 equilibrium at the beginning of imbibition. For the first five days of the test, the water level in the burette  
142 was maintained at the same level as the bottom porous disk (water level “1” in Figure 2). This allowed  
143 the pellets and powder grains in contact with the bottom porous stone to form a thin layer without large  
144 inter-pellet voids. Afterwards, the water level was set to the height of the cell (water level “2” in Figure  
145 2). The water head, 120 mm ( $\approx 1.2$  kPa), was small so that the thin homogeneous layer obtained during  
146 the first days prevented quick liquid water flow through inter-pellet voids.

147 The burette was regularly filled with synthetic water during the duration of the test. A small quantity  
148 of oil was poured at the top of the water column to prevent evaporation of the water in the burette.  
149 Silicone gel was added, between the glass faces and the metal cell, following the cell closure to prevent  
150 any leakage during imbibition.

## 151 2.6. Measurements of water content and suction

152 After the end of imbibition tests, suction and water content were measured at different elevations in  
153 the samples. Suction was measured using a WP4 dew point potentiometer (Decagon). Water content  
154 was measured by oven-drying at 105°C for 24 h. In the following, the term water content is used for the  
155 gravimetric water content in the sample.

## 156 3. Experimental results

157 This section presents the results obtained after 640 days of imbibition for the 70/0 and 70/15  
158 mixtures. Because of an experimental issue, the 70/30 test was stopped and dismantled after 235 days,  
159 thus results for only 235 days are presented for this mixture. “Final values” reported in the following  
160 thus correspond to values measured after 640 days for the 70/0 and 70/15 samples and 235 days for the  
161 70/30 sample.

### 162 3.1. Imbibed volume of water

163 The volume of imbibed water as a function of elapsed time is presented in Figure 4a for the three  
164 mixtures, along with horizontal lines corresponding to the estimated total void volume in the samples  
165 (including pellet porosity + powder grain porosity + inter-pellet porosity). These values, summarised in  
166 Table 6, were estimated from data shown in Table 3 and Table 4. Figure 4b presents the evolution of the  
167 water volumetric flow rate during the three tests. It can be observed that the water uptake as well as the  
168 water uptake rate were very close between the three tests, despite notable differences in total dry density.

169 Table 6. Information regarding estimated void volumes in the three tested mixtures.

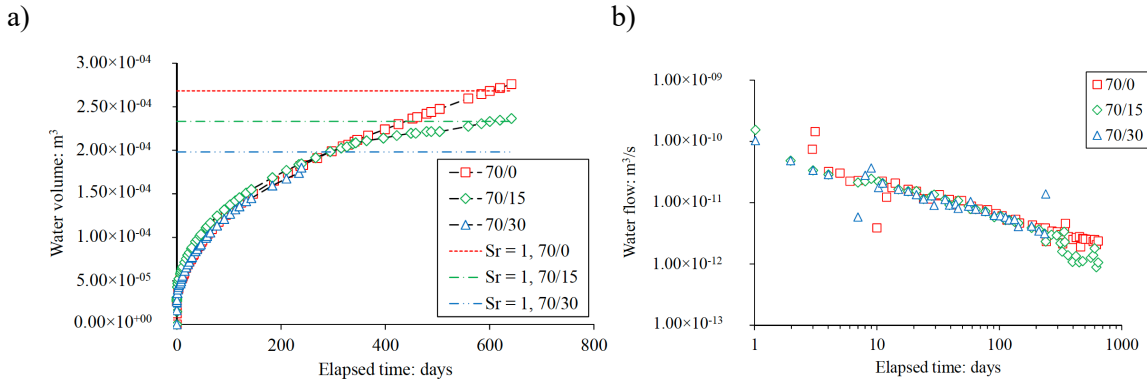
Mixture	70/30	70/15	70/0
Total porosity: dimensionless	0.458	0.534	0.621
Total void volume: $\times 10^{-4}$ m <sup>3</sup>	1.98	2.33	2.68
Saturated water content: dimensionless	0.31	0.42	0.59

170

171 The evolution of the imbibed water volume in the three cells followed close trends. The water flow  
172 rate monotonically decreased during imbibition. The water flow rate in the experiments can be related  
173 to the logarithm of elapsed time with a linear function. This function is the same for the three mixtures.  
174 In the 70/15 experiment, progressive clogging of the inlet tube by rust from a screw located at the bottom

175 of the sample induced a decrease in the water flow (between 350 – 500 days). After changing the tube,  
 176 the water flow increased again to values comparable to the global trend.

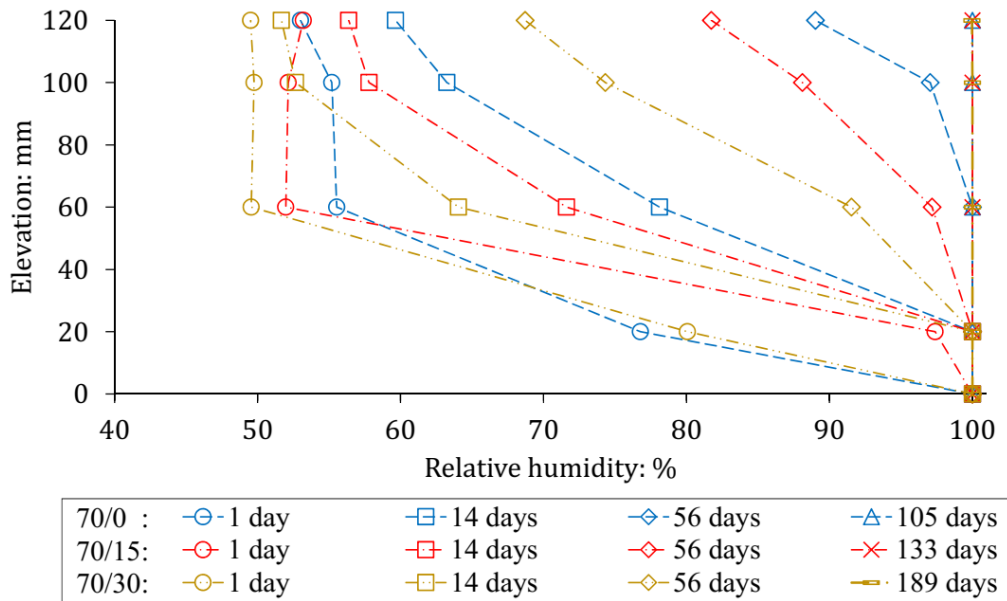
177 After ~600 days of imbibition, the estimated volume of voids in the sample was reached in samples 70/0  
 178 and 70/15. Water intake continued to increase, while the water flow rate continued to decrease.



179  
 180 Figure 4. Evolution of a) imbibed water volume and b) water flow rate as functions of elapsed time for the three imbibition  
 181 tests. Horizontal lines in a) correspond to the estimated total void volume in each sample (Table 6).

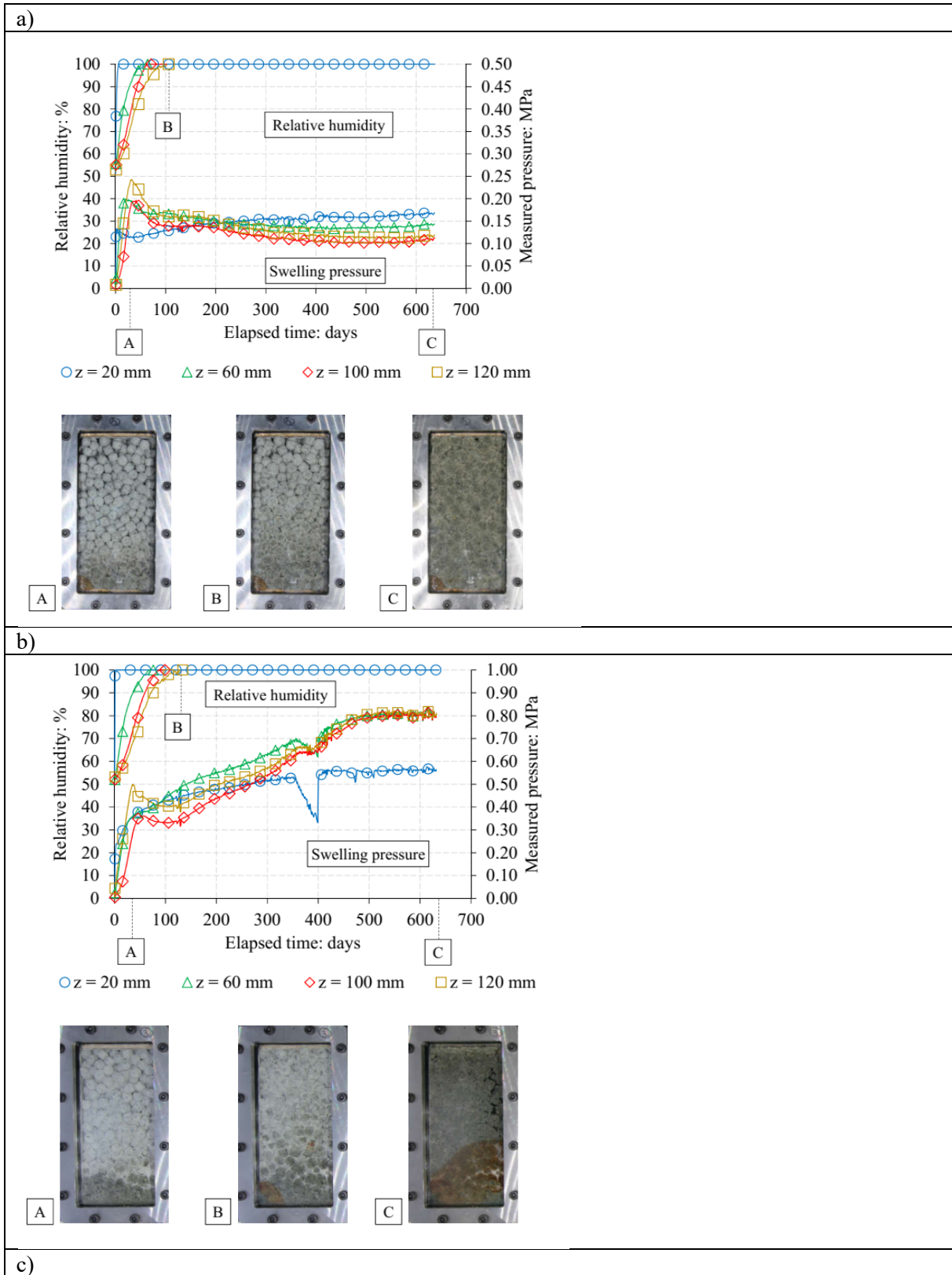
182 **3.2. Evolution of the relative humidity**

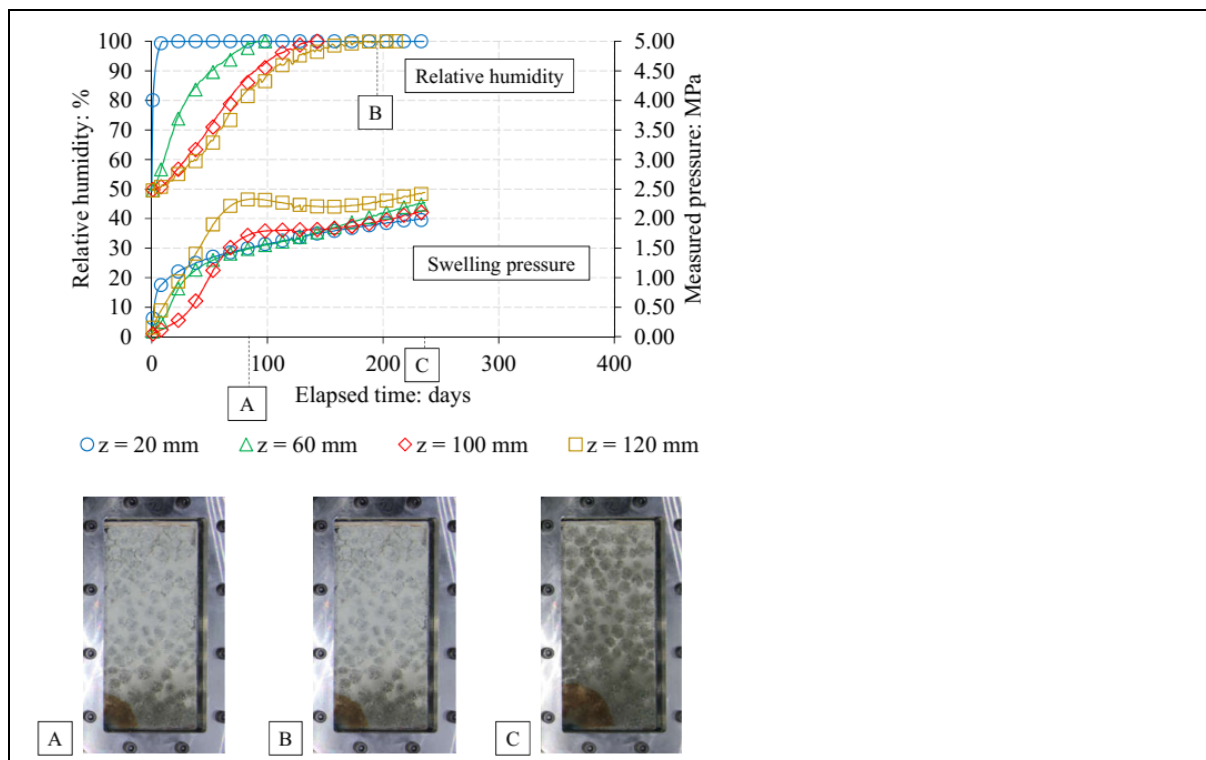
183 In all three samples, the relative humidity monotonically increased with time during the first 200  
 184 days (Figures 5 & 6). The increase in relative humidity at each value of  $z$  was slower for samples with  
 185 higher powder content.



186  
 187 Figure 5. Profiles of relative humidity in the three samples at different times.

188 The time required for all sensors to obtain a relative humidity of 100% was 105 days for the 70/0  
 189 sample, 133 days for the 70/15 sample, and 189 days for the 70/30 sample. Afterwards, the measured  
 190 relative humidity remained 100% for all the sensors (Figure 6). In addition, the initial suction of the  
 191 mixtures can be estimated from the measurement of relative humidity. For instant, initial relative  
 192 humidity was equal to 52-54%, 51-53% and 50% for the samples 70/0, 70/15, and 70/30, respectively.  
 193 Note that the initial suction of the powder (180 MPa) was higher than that of pellet (89 MPa). Higher  
 194 powder content should induce thus higher suction (or lower relative humidity) of the mixture.





196 Figure 6. Evolution of the measured pressure, relative humidity, and pictures of the materials at various elapsed times: a)  
 197 70/0; b) 70/15; c) 70/30.

### 198 3.3. Evolution of the measured pressure

199 In the 70/0 sample (Figure 6a), lateral pressure increased first close to the water inlet (sensor  
 200  $z = 20$  mm), at the bottom of the sample. A peak was reached after few days. Afterward, the pressure  
 201 decreased, stabilised and started to increase from  $\sim 50$  days. At higher elevations, the measured pressure  
 202 evolved differently. Axial and lateral pressures increased to higher values and reached sharper peak.  
 203 Following the peaks, these pressures decreased progressively and slow increase in pressure was  
 204 observed only from  $\sim 500$  days.

205 In the 70/15 sample (Figure 6b), pressures at  $z = 20$  mm and  $z = 40$  mm increased continuously from  
 206 the beginning. At higher elevations, the trend was close to that observed in the 70/0 sample, but the  
 207 increase in the measured pressure following the peak was more significant and occurred earlier (from  
 208  $\sim 100$  days). A transient decrease of pressure was observed before  $\sim 400$  days by all sensors. This decrease  
 209 corresponded to a progressive clogging of the water inlet tube identified in the analysis of water intake  
 210 (Figure 4). After changing the tube, the measured pressures increased again at all elevations.

211 In the 70/30 sample (Figure 6c), the evolution of pressure close to the water inlet followed a trend  
 212 comparable to the 70/15 sample. However, at higher elevations, the evolution of the measured pressures  
 213 was different. Increase of measured pressures was slower and peaks were not as sharp. As in other tests,  
 214 the highest value of peak pressure was obtained at the top of the sample.

215 In all samples, the measured pressure continued to increase after relative humidity at all elevations  
 216 had reached 100% (Figure 6)

### 217 3.4. Evolution of the texture of the mixtures

218 Pictures of the samples, taken during hydration (Figure 6), allowed the evolution of the material  
 219 textures to be observed and analysed. Three pictures are shown corresponding to the moments when the  
 220 peak axial pressure was measured at the top of the samples (A), when 100% RH was measured by all  
 221 sensors (B), and at the end of the test before dismantling (C).

222 When the peaks of axial pressure were measured at the top of samples 70/0 and 70/15 (moment A in  
 223 Figure 6a,b), both characterised by sharp decrease of axial pressure (compared to that of sample 70/30),

224 inter-pellet voids were still clearly observable. The powder grains did not fill all the inter-pellet voids at  
 225 the top of the 70/15 sample and the top of both 70/0 and 70/15 samples had a clear granular structure.  
 226 In the 70/30 sample (Figure 6c), inter-pellet voids were completely filled with powder grains when the  
 227 peak axial pressure was reached. In the three samples, the liquid water front was still at the bottom of  
 228 the sample. At the bottom of the samples the structure was more homogeneous.

229 When 100% of relative humidity was reached at all elevations (moment B in Figure 6), large inter-  
 230 pellet voids were still present at the top of the 70/0 sample (Figure 6a). Conversely, volume of inter-  
 231 pellet voids decreased because of pellet swelling and a significant part of the inter-pellet voids was filled  
 232 with powder in the 70/15 sample (Figure 6b). It corresponded to the moment when measured pressures  
 233 increased again in the 70/15 sample, while they continued to decrease but with a much smaller rate in  
 234 the 70/0 sample. The texture of the 70/30 material had not significantly evolved when 100% of relative  
 235 humidity was measured. In all three samples, the liquid water front was still at a low elevation at this  
 236 moment.

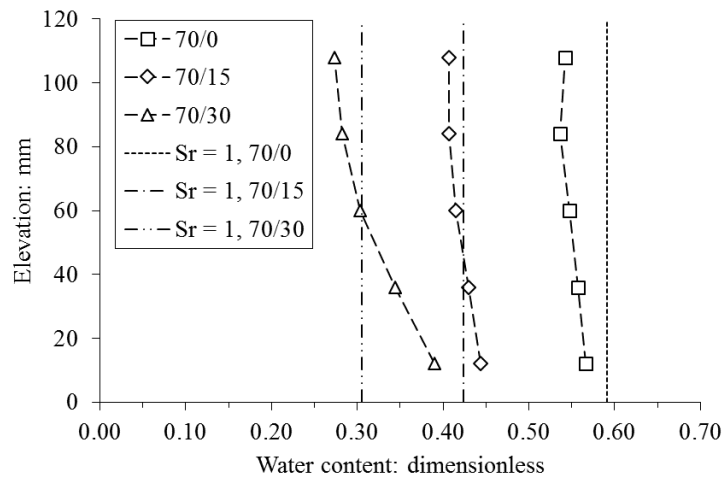
237 At the end of the test (moment C in Figure 6), the closure of inter-pellet voids was nearly total in the  
 238 70/0 sample and total in the other tests. In all samples, the material had lost the initial granular structure  
 239 but was not completely homogeneous. Besides, some dry pores could be identified in the 70/0 sample,  
 240 demonstrating that full saturation was not reached.

241 A dark area spreading from the bottom of the 70/15 sample can be observed on the picture. This was  
 242 due to rust coming from a screw located at the bottom of the cell, as already mentioned.

### 243 3.5. Sample dismantling

244 At the end of the test, the cells were opened by removing the glass side facing the camera. Following  
 245 the opening, a utility knife was used to split samples into small portions, in order to measure suction and  
 246 water content profiles.

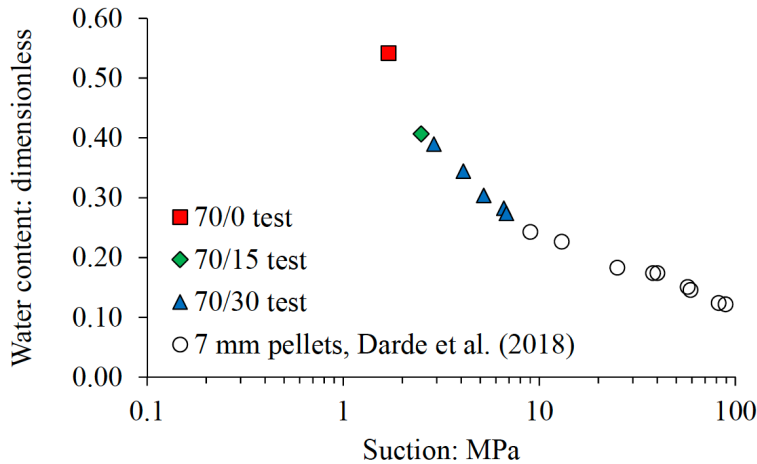
247 The water content profiles of the three samples are presented in Figure 7, along with values of  
 248 estimated saturated water content as given in Table 6. Saturated water content was estimated with a  
 249 water density  $\rho_w = 1.0 \text{ Mg/m}^3$  and a constant value of the mixture dry density. In the three samples, the  
 250 water content following dismantling was higher at lower elevation.



251  
 252 Figure 7. Water content profiles of the three samples following dismantling. “Sr = 1” lines correspond to the estimated  
 253 water content in fully-saturated state for constant dry density and water density of  $1.0 \text{ Mg/m}^3$ .

254 Values of suction (measured by WP4) and water content (determined by oven-drying) were obtained  
 255 at  $z \approx 95\text{-}120 \text{ mm}$  for the 70/0 and 70/15 samples, and  $z \approx 0\text{-}25 \text{ mm}$  ;  $25\text{-}50 \text{ mm}$  ;  $50\text{-}70 \text{ mm}$  ;  $70\text{-}$   
 256  $95 \text{ mm}$  ;  $95\text{-}120 \text{ mm}$  for the 70/30 sample. The results of water content-suction relationship for the three  
 257 samples are presented in Figure 8, along with results obtained for a free swelling MX80 7 mm pellet,

258 identical to those used in the present study (Darde et al., 2018). All the experimental data seem show a  
259 unique relationship which could be attributed to the water retention curve of the material.



260

261 Figure 8. Water content – suction relationship obtained by measures performed on the three samples and a single pellet  
262 under free swelling conditions.

263

## 264 4. Discussion

### 265 4.1. Choice of experimental apparatus

266 In this study, imbibition columns were square cuboids with two glass faces. This allowed for the  
267 direct observation of the material using a camera. From the existing studies, cylindrical cells have been  
268 generally used (e.g. Imbert and Villar, 2006). The cylindrical shape reduces the local deformation;  
269 however, it prevents the direct observation of the material. The square cells could induce more  
270 deformation of the material in the centre compared to that at the angles. In the present work, pictures  
271 taken by the camera did not show significant difference on the behaviour of the material at the angles  
272 compared to the other parts.

273 The use of a second glass face should have allowed the observation of both sides of the cell. Because  
274 of space constraint in the laboratory, it was not possible to have cameras on both sides of the cells.  
275 Observation of the material evolution on both sides remains an interesting perspective for future  
276 experiments.

### 277 4.2. Non-measurement of dry density

278 The profiles of dry density were not determined following dismantling in the present work. The  
279 authors considered that experimental constraints (such as the necessity to test a part of sample large  
280 enough to contain pellets and powder, the unavoidable small but still non-negligible deformation during  
281 sampling, the time required for the measurement, etc.) could affect the relevance of this measurement.

### 282 4.3. Slow hydration method

283 Because of the large inter-pellets pores in the initial state, the permeability of pellet mixtures in the  
284 initial state should be high (Hoffmann et al., 2007). Thus, upon fast liquid flooding, water would flow  
285 in the large pores, as discussed by Molinero Guerra et al. (2018a), resulting in early loss of granular  
286 structure and homogenisation of the mixture. In the present study, it was found that upon slow hydration,  
287 the materials could be inhomogeneous even at a relative humidity of 100%. In pellet mixtures, contact  
288 interactions between pellets have an influence on the evolution of swelling pressure (Hoffmann et al.,  
289 2007; Alonso et al., 2010; Darde et al., 2020b).

### 290 4.4. Homogenisation of the mixture upon hydration

291 The homogenisation of pellet-based bentonite materials reaching fully-saturated state was observed  
292 by various authors (Imbert and Villar, 2006; Hoffmann et al., 2007; Molinero Guerra et al., 2018a;  
293 Bernachy-Barbe et al., 2020; Darde et al., 2020a). In the present work, taking pictures allowed to follow  
294 the progressive loss of the initial granular structure. Under constant-volume conditions, the swelling of  
295 the pellet-powder mixture as a whole was prevented. Powder grains thus swelled in inter-pellet voids,  
296 the volume of which progressively decreased also as a consequence of pellet swelling, leading to the  
297 loss of granular structure. As dismantling was performed before full saturation, the texture did not reach  
298 the same degree of homogenisation as in the previously mentioned studies. However, pictures showed  
299 that pellets and powder were no longer distinct from each other at the end of the tests.

300 Dismantling allowed to determine profiles of water content for the three mixtures (Figure 7). In all  
301 three mixtures, the water content was higher at a lower elevation. Even if full saturation was not reached,  
302 it is worth noting that for the 70/30 sample, the final water content at  $z < 60$  mm was higher than the  
303 value estimated for the saturated state corresponding to the initial average void ratio. Upon hydration,  
304 the bentonite swelled close to the water inlet. Consequently, the void volume following saturation was  
305 higher in the zone close to the water inlet. This local swelling close to water inlet could compact the part  
306 of the sample further from the water inlet. Thus, even if the texture of the material is expected to reach  
307 a full homogenisation at full saturation, inhomogeneous profiles of water content and densities are to be  
308 expected in pellet-based bentonite materials as in other bentonite materials.

309 Axial and lateral pressures were not similar in the samples at the end of the tests. Even if further  
310 evolution was still expected because the samples were not fully-saturated, disparity of the measured  
311 pressure has been observed in the literature (Molinero Guerra et al., 2018b; Dueck et al., 2019;  
312 Bernachy-Barbe et al., 2020; Bernachy-Barbe, 2021). Swelling pressure was affected by the initial  
313 heterogeneities of density of the sample (Molinero Guerra et al., 2020), as well as by the non-  
314 homogeneous hydration as previously discussed. In addition, friction between bentonite and the cell  
315 walls may contribute to the difference between axial and lateral pressures (Bernachy-Barbe, 2021). Such  
316 heterogeneities of measured pressure were also obtained in experiments performed on compacted  
317 bentonite block (Saba et al., 2014).

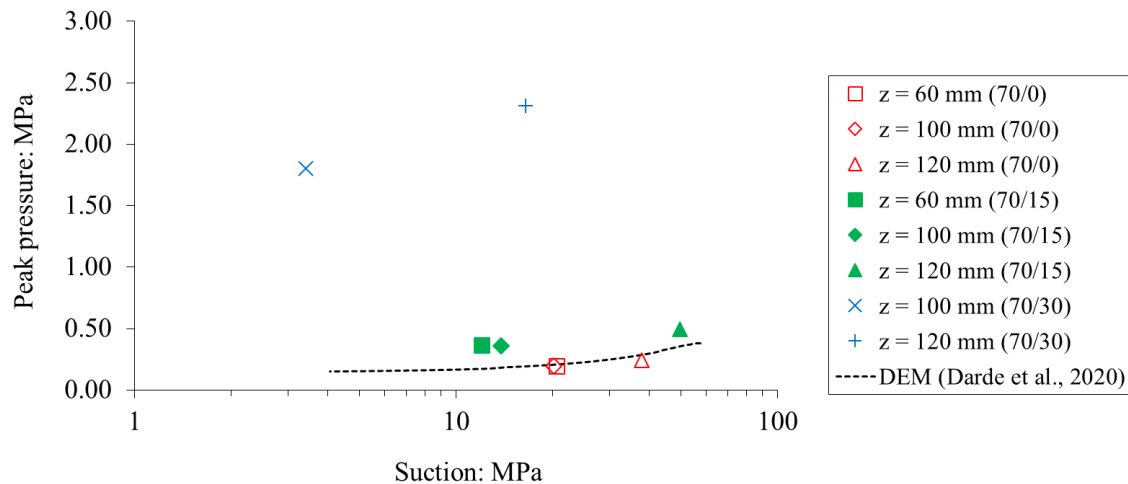
#### 318 **4.5. Influence of the granular structure**

319 The use of cells with glass sides allows to observe the progressive evolution of the material structure  
320 from the initial state. It provides a complementary “qualitative” information to sensor “quantitative”  
321 measurements like values of measured pressure and relative humidity, which brings insight into the  
322 mechanical behaviour of pellet-based bentonite materials under repository conditions.

323 Pictures taken by the camera (Figure 6) showed that the peak axial pressure measured at the top of  
324 the sample for 70/0 and 70/15 was reached when the material structure was characterised by large inter-  
325 pellet pores. Conversely, the inter-pellet pores in the 70/30 sample at peak axial pressure measured at  
326 the top of the sample were completely filled with powder. Besides, after 133 days for the 70/15 sample  
327 and 189 days for the 70/30 sample, when measured axial and lateral pressures were increasing following  
328 the peak in both samples, inter-pellet pores were filled with powder.

329 Figure 9 presents the peak pressures measured during hydration in all samples as functions of suction,  
330 converted from relative humidity measured at the same height using the Kelvin law. Two distinct trends  
331 can be observed: high pressure peaks were obtained in the 70/30 sample; and low swelling pressure  
332 peaks were obtained in the 70/0 and 70/15 samples. Along with experimental results, the mean stress  
333 corresponding to pellet failure in a pellet assembly without powder, determined by Darde et al. (2021),  
334 was plotted in a dashed line. The low swelling pressure peaks, obtained in samples with an observable  
335 granular structure (which were considered in the numerical simulation performed by Darde et al. 2021  
336 using DEM), were close to this line. That confirmed the dominant role of granular structure on swelling  
337 pressure for these samples. Note that the variability of the measurement of suction was expected to be  
338 lower than 5 MPa, such that these trends are not considered affected by sensor accuracy. In addition, the  
339 diameter of the pressure sensors was taken four times as large as the pellet so that measured pressures  
340 are considered accurate.





341

342 Figure 9. Peak pressure measured in the three samples plotted as a function of suction, determined from relative humidity  
 343 measurement and the Kelvin law. The dashed line is the mean stress corresponding to pellet strength in a pellet assembly as  
 344 determined by Darde et al. (2021) using Discrete Element Method.

345 Measurement of a peak of pressure was described in other studies about hydration of bentonite-based  
 346 materials under constant volume conditions. In clayey materials compacted on the dry side of optimum  
 347 compaction (i.e. water content smaller than optimum water content), clay particles (basically stacks of  
 348 lamellae) form aggregates, resulting in a well-documented double structure (Pusch, 1982; Delage et al.,  
 349 1996; Lloret et al., 2003; Cuisinier and Laloui, 2004; Romero and Simms, 2008). Bentonite hydration  
 350 results from the progressive adsorption of water molecules around the cations in the interlayer (Norrish,  
 351 1954; Sposito and Prost, 1982; Torikai et al., 1996; Saiyouri et al., 2004; Jacinto et al., 2012), inducing  
 352 clay particles swelling and decrease of aggregate stability. At high water contents, the aggregate  
 353 structure is no longer stable, collapses, and macropores are filled with exfoliated clay lamellae (Keller  
 354 et al., 2015; Massat et al., 2016). The hydration mechanism then becomes related to the development of  
 355 diffuse double layers (Saiyouri et al., 2000; Delage et al., 2006).

356 Under constant-volume conditions, these mechanisms can result in the development of a peak  
 357 swelling pressure related to the collapse of the aggregated structure, followed by another increase to a  
 358 final value (Lloret et al., 2003). As reported by Lloret et al., (2003), the decrease of swelling pressure  
 359 resulting from this phenomenon was observed if the level of stress was sufficiently high, and thus may  
 360 not be observed in low dry density samples. Peaks of pressure were also measured in pellet-based  
 361 bentonite materials as discussed by others studies (Imbert and Villar, 2006; Alonso et al., 2011; Molinera  
 362 Guerra et al., 2018b; Benarchy-Barbe et al., 2020).

363 Because of the initial granular structure, the evolution of the measured pressure of pellet materials  
 364 can be different from that of compacted blocks. Before losing the initial granular structure upon  
 365 hydration, the macroscopic pressure developed in a pellet assembly results from an increase of number  
 366 of contacts between grains and the pressure sensor and the magnitude of force at these contacts. If  
 367 contact forces reach pellet strength, they remain constant (perfect plasticity behaviour), while the pellet  
 368 stiffness continues to decrease upon hydration, inducing swelling pressure decrease (Darde et al., 2021).  
 369 Results of the present study suggest that, in mixtures of pellet and powder, the cause of the transient  
 370 decrease of swelling pressure depends on the powder content.

371 In low powder content mixtures, the powder does not completely fill the inter-pellet pores. Its initial  
 372 contribution to the mechanical behaviour is negligible (Darde et al., 2020a). Pressure development is  
 373 related to the increase in contact forces between pellets, and a peak of pressure occurs if contact forces  
 374 reach the pellet strength. Following the peak, pellets and powder continue to swell. The contribution of  
 375 powder progressively becomes significant and overcomes that of contact forces between pellets. The  
 376 material behaviour is then comparable to that of a continuous material. Results obtained for the 70/15  
 377 sample can be explained by this interpretation (Figure 9).

378 In high powder content mixtures, the powder fills the inter-pellet pores. Its contribution to the  
379 mechanical behaviour of the mixture cannot be neglected. Swelling pressure is not directly related to  
380 contact forces between pellets. The pellet assembly does not control the mechanical behaviour. Peak  
381 pressure is thus interpreted as the collapse of the aggregate structure, as continuous bentonite materials.  
382 Results obtained for the 70/30 sample can be explained by this interpretation. Besides, Imbert and Villar  
383 (2006) concluded that a mixture of pellets and powder in 50/50 proportions did not behave differently  
384 than continuous bentonite materials of same dry density.

385

#### 386 4.6. Water migration process

387 The saturated permeability in compacted bentonite was shown to be related to its dry density  
388 (Karnland et al., 2006; Lloret and Villar, 2007). It suggests that under fully-saturated conditions, water  
389 flows through the macropores of the bentonite. In the present study, three different mixtures of pellets  
390 and powder were studied. The initial dry densities of these mixtures were  $1.05 \text{ Mg/m}^3$ ,  $1.275 \text{ Mg/m}^3$ ,  
391 and  $1.50 \text{ Mg/m}^3$ . Under saturated conditions, it was expected that permeability, thus water flow under  
392 comparable gradients of water potential, would be significantly higher for lower dry densities.

393 In Figure 4, for the three samples, the flow rate of water entering the three samples was comparable,  
394 regardless of the mixture dry density. Besides, it was observed that the liquid water front was at a low  
395 elevation when all sensors measured a relative humidity of 100%. Air-filled inter-pellet pores could be  
396 observed in pictures taken by the camera. These observations suggest that the dominant water migration  
397 mechanism under unsaturated conditions was not liquid flow in the macropores (inter-aggregate pores  
398 in the pellets or powder grains). Two other mechanisms can be proposed to explain the saturation of the  
399 material. The first mechanism is liquid water flow through the micropores (intra-aggregate pores in the  
400 pellets or powder grains). The second mechanism is vapour water flow in the macropores and inter-  
401 pellet pores. The flow rate of water entering the sample was thus controlled by: (i) liquid flux in the  
402 micropores, affected by strong solid-water interactions (Komine, 2008); (ii) the rate at which the liquid  
403 water undergoes phase change to vapour at the interface between the saturated and unsaturated zones  
404 (Lozano et al., 2008; Ouedraogo et al., 2013). Both phenomena can be considered independent of dry  
405 density.

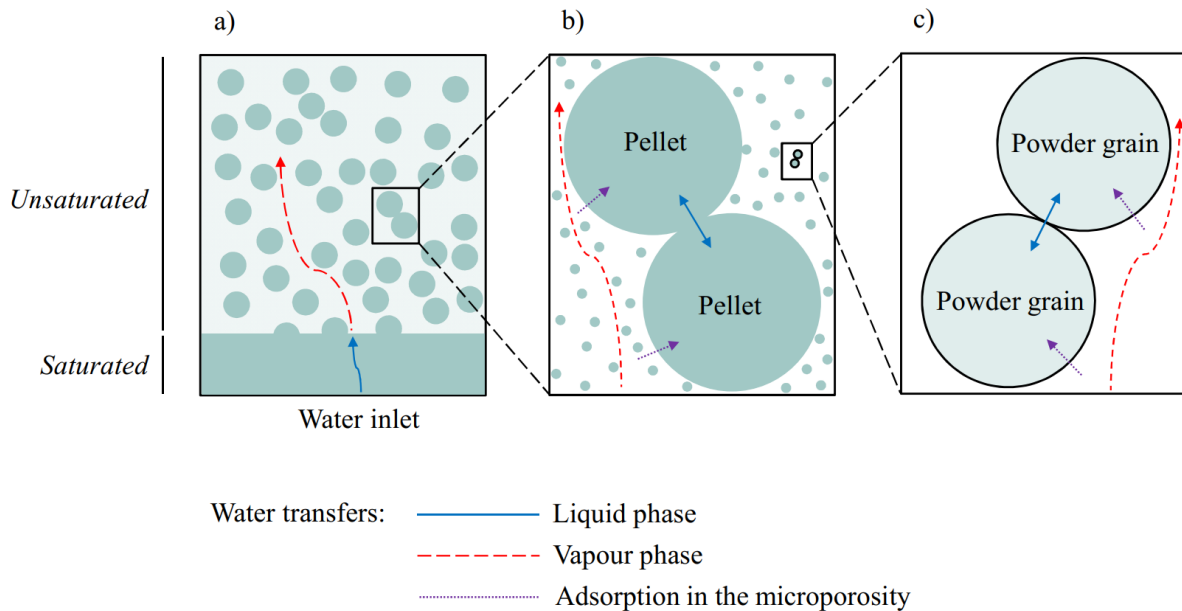
406 Comparison of Figure 4 and Figure 6 **Error! Reference source not found.** provides interesting  
407 additional information about the hydration of the mixtures. The volume of water imbibed by the samples  
408 continued to increase in all samples after all sensors measured a 100% relative humidity in the inter-  
409 pellet pores. Thus, the bentonite continued to take up water. It confirms that diffusive migration of water  
410 vapour was faster in the large inter-pellet space than in the micropores of the pellet. Kröhn (2005) also  
411 proposed vapour migration as the main saturation mechanism in compacted bentonites. Besides, it  
412 means that pellets and inter-pellet pores were not at hydraulic equilibrium during saturation. The  
413 increase in measured pressure following the measurement of 100% relative humidity was also consistent  
414 with this suggestion. On another note, even if the material was not saturated when dismantled, the  
415 imbibed volume of water was higher than the estimated volume of voids (Figure 4). Similar results have  
416 been already observed by Wang et al. (2013). A possible interpretation is the increase of density of the  
417 water adsorbed on the surface of clay lamellae, as already suggested in the literature (Lloret and Villar,  
418 2007; Jacinto et al., 2012; Molinero Guerra et al., 2018b). Another possible explanation would be a leak  
419 in the system, despite the care taken to avoid leakage, or the volume of water required to fill the ducts  
420 and porous plate at the bottom of the column was underestimated.

421 The size of powder grains was smaller than that of pellets. Consequently, a decrease in the relative  
422 humidity of the surrounding air (pore between grains or pellets) would induce a faster decrease of  
423 relative humidity in the whole powder grain than in the whole pellet. In Figure 6, progress of vapour  
424 diffusing in the pore air is shown to be slower in mixtures with higher powder content. It suggests that  
425 hydration of powder grains occurred simultaneously with vapour migration in the inter-pellet pores. This  
426 hydration consumed water, and thus slowed down the apparent rate of vapor diffusion throughout  
427 mixtures containing powder. It is consistent with the profiles of water content following dismantling  
428 (Figure 7): the differences of water contents measured at low and high elevations were less significant

429 in the mixture with no powder, where vapour could diffuse in the large inter-pellet pores with no powder.  
 430 The delay of hydration and hydraulic disequilibrium between micropores and inter-pellet pores is thus  
 431 considered to be mostly related to pellets.

432 Figure 10 presents a summary of the interpretations of experimental results regarding the water  
 433 migration in the samples during the saturation.

434



435

436 Figure 10. Interpretation of the water migration mechanisms in the samples: a) at the imbibition cell scale; b) at the pellet  
 437 scale; c) at the powder grain scale.

438

439 The present discussion suggests that, despite the very high initial permeability of the material, water  
 440 flow in liquid phase through inter-pellet space is very unlikely before full saturation of the buffer under  
 441 repository conditions, in concepts with a low permeability host rock such as claystone.

442

## 443 5. Conclusion

444 The present study investigated the hydromechanical behaviour of pellet-powder mixtures through  
 445 imbibition experiments in the laboratory. A hydration column was specially designed, with glass sides,  
 446 allowing pictures of the sample to be taken during hydration, along with measurements of relative  
 447 humidity and swelling pressure at different elevations. The experiments allowed the transient behaviour  
 448 of pellet-powder mixtures to be studied under conditions close to repository conditions by avoiding  
 449 significant liquid water flow rates in the large inter-pellet voids. Swelling pressure was measured by  
 450 using sensors four times as large as the pellets to decrease the measurement variability. Three mixtures  
 451 with identical pellet volume fractions (with respect to the total volume) and various powder volume  
 452 fractions were tested to study the influence of heterogeneities of density in the repository.

453 In this regard, the present study provides an insight into the behaviour of pellet-powder mixtures  
 454 under repository conditions, the influence of heterogeneities of density, water migration mechanisms,  
 455 and progressive homogenisation of the initial structure. The following conclusions can be drawn from  
 456 the present work:

- 457 - (i) The average dry density of the material did not influence the water flow entering the samples  
 458 under unsaturated conditions;

- 459 - (ii) The evolution of pressure, including the occurrence of a peak, was consistent with an increase  
460 in contact number between pellets (and the forces at these contacts) for samples with low powder  
461 content;  
462 - (iii) The material progressively lost the initial granular structure during hydration;  
463 - (iv) Pellets and inter-pellet pores were not at hydraulic equilibrium during imbibition;  
464 - (v) Vapour migration was expected to be the dominant mechanism of saturation of the pellet-  
465 powder mixtures investigated in this study.

466 In depth analysis of the homogenisation process in pellet mixtures based on the pictures, and relevant  
467 comparison of the swelling pressure development in bentonite pellet mixtures and compacted blocks of  
468 same dry density thanks to the large pressure sensors are two interesting perspectives of the present  
469 work.

#### 470 **Declaration of competing interests**

471 The authors declare that they have no known competing financial interests or personal relationships  
472 that could have appeared to influence the work reported in this paper.

473

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