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Hydro-mechanical behaviour of a cement-bentonite mixture along evaporation and water-uptake controlled paths / Musso, Guido; Vespo, VINCENZO SERGIO; Guida, Giulia; Della Vecchia, Gabriele. - In: GEOMECHANICS FOR ENERGY AND THE ENVIRONMENT. - ISSN 2352-3808. - 33:(2023), p. 100413. [10.1016/j.gete.2022.100413]

Availability:

This version is available at: 11583/2973420 since: 2022-11-28T09:27:22Z

Publisher:

Elsevier

Published

DOI:10.1016/j.gete.2022.100413

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<http://dx.doi.org/10.1016/j.gete.2022.100413>

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Hydro-mechanical behaviour of a cement-bentonite mixture along evaporation and water-uptake controlled paths

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Abstract

The results of a laboratory investigation on the effects of drying and wetting on the hydro-mechanical behaviour of a cement-bentonite mixture are presented. A large hysteresis along the hydraulic paths was found for what concerns both the water retention and the volume strains. This might be related to the shrinkage of cement bentonite clusters and to the development of micro-cracks, that were observed with a microscope along drying. Macroscopic cracks developed in a squat sample which had been cured for 28 days and was then left to dry in the laboratory environment, while this did not occur in an analogous sample cured for 60 days, possibly in virtue of its higher strength as detected through a pocket penetrometer. Water uptake after partial desaturation of squat and column samples took place in short times with respect to drying, but it did not allow for a full recovery of the initial water content. Electrical Resistivity Tomography (ERT) monitoring provided reliable water content determinations in the high range and correct indication of crack patterns at low water contents. A good reproduction of the experimental results was obtained with a coupled Thermo-Hydraulic model formulated on purpose.

1 Introduction

Cement-bentonite mixtures (CB) are widely employed in many geotechnical and geo-environmental applications such as jet grouting, pile systems and slurry trench cutoff walls. The latter aim at controlling groundwater flow and at confining both dissolved and non-aqueous pollutants (Cermak *et al.*, 2012; Evans and Ruffing, 2017). Cutoff walls are generally built excavating trenches enclosing the polluted site, and filling them up with the CB slurry (Ryan, 1985; Evans, 1994; Malusis *et al.*, 2017). The curing process, that occurs on site, conveys to the mixture target properties in terms of hydro-mechanical behaviour and durability (Fratolocchi *et al.*, 2006; Guida *et al.*, 2021; Flessati *et al.*, 2021).

In many site applications, the water level inside or outside the encapsulated area may fluctuate over time due to natural and anthropogenic causes. Furthermore, the top of the wall is exposed to the atmosphere, being subjected to evaporation (whose effects will be increasingly important due to climate change) or infiltration processes. As a consequence, a significant part of the barrier may be subjected to drying/wetting cycles (Wu *et al.*, 2019). This may lead to changes in the hydraulic state of the material and to a reduction of the overall effectiveness of the barrier (Malusis *et al.*, 2011).

In particular, the shallower portions of cutoff walls often show signs of damage due to drying soon after the construction, especially if they are left uncapped (Joshi *et al.*, 2009). According to Jefferis (2012), CB slurry trench cutoff walls develop desiccation cracks because of drying induced shrinkage. The formation of cracks affects the transport properties required for a good performance of the barrier, leading *e.g.* to an increase of hydraulic conductivity of about 2-3 orders of magnitude, to a degradation of material strength and to an increase in heterogeneity of the wall on site (Joshi *et al.*, 2009; Wu *et al.*, 2019). Despite the engineering relevance of the changes in hydraulic properties that might be triggered by desiccation, a limited number of studies has been focusing on these aspects. Soga *et al.* (2013) found that CB samples cracked when exposed to laboratory air, but remained intact when they were immersed in a box filled with sand and then exposed to drying-wetting cycles. (Royal *et al.*, 2018) have reported experimental evidences about microstructural changes of enriched cement-bentonite specimens exposed to air, including discolouration (changing from dark green to a very pale blue tinged light grey), formation of irreversible drying induced shrinkage cracks on the surface, widening (and intersection) of cracks with prolonged drying upon complete disgregation. A few studies dealt with the effects of additives on the tendency to cracking of the cement bentonite mixtures. The ash of slag enrichment allows to improve the low permeability requirements for the CB in saturated conditions, but it seems to induce worse performances upon drying (Jefferis, 2012), whereas according to the experimental evidence in Cao *et al.* (2021) adding super-absorbent poly-

mers increases the resistance to cracking of the mixture.

In general, a full understanding of the impact of evaporation and infiltration, and therefore of drying and wetting cycles, on the hydro-mechanical behaviour of CB mixtures is still missing. As an attempt to contribute to this aspect, obtained through the material characterization aimed at thermo-hydraulic modelling, this paper presents the results of a laboratory study on the hydro-mechanical behaviour of CB samples in unsaturated conditions, with a focus on the water retention properties and on the volumetric and strength changes induced by suction variations. No additives were used in the preparation of the mixtures used in this study, as this can be considered the 'baseline' material with respect to whom the enriched mixtures might be compared. Royal *et al.* (2018) have reported experimental evidences about microstructural changes of enriched cement-bentonite specimens exposed to air, including discolouration (changing from dark green to a very pale blue tinged light grey), formation of irreversible drying-induced shrinkage cracks on the surface, widening (and intersection) of cracks with prolonged drying upon complete disgregation.

The effects of suction changes on the water retention and on the volumetric behaviour were investigated first. A preliminary insight on the effects of water content on strength was obtained through pocket penetrometer measurements, while the development of cracks along drying was checked with visual inspection and Electrical Resistivity Tomography (ERT) on squat samples. Evaporation and water uptake from a column sample was investigated assessing local changes in the water content through ERT. A good reproduction of the results of the drying and water uptake experiments was obtained through simulations with a Thermo-Hydraulic coupled model, whose parameters were set up accordingly with the previous water retention and permeability measurements .

2 Material and methods

2.1 Material

The tests were performed at the Geotechnical Engineering Lab of the Department of Structural, Geotechnical and Building Engineering of Politecnico di Torino. CB mixtures were prepared by mixing a commercial cement (CEM I 32.5 N, Portland 325) with a commercial mixed sodium-calcium bentonite provided by Laviosa Chimica Mineraria (Livorno, Italy). The bentonite has a liquid limit $w_L = 616\%$, a plastic limit $w_P = 42\%$, a specific gravity $G_s = 2.95$ and a powder hygroscopic water content at laboratory conditions $w = 14\%$ (Trischitta *et al.*, 2020). The mixing proportions are consistent with the ones used in the construction of the cut-off walls (see *e.g.* Sarsby 2013). The mixture

85 preparation procedure consisted in: (i) mixing tap water with bentonite at a 18:1 water-bentonite
86 mass ratio until a homogeneous plastic slurry was obtained; (ii) leaving the slurry covered with an
87 impermeable sheet for 24 hours to ensure full hydration; (iii) adding dry cement powder in order to
88 obtain a cement-bentonite mass ratio 4:1 and stir again the suspension; (iv) pouring the mixture into
89 plastic cylindrical moulds of different size and leaving it to cure immersed in tap water for 28 or 60
90 days. The specific gravity of the endured mixture was $G_s = 2.68$. The water content of the samples
91 was found to range between $w_0 = 285\%$ and $w_0 = 325\%$, while the saturated hydraulic conductivity at
92 a confinement of 50 kPa measured with flexible and rigid wall permeameters was around $K_w^{sat} = 10^{-8}$
93 m/s.

94 **2.2 Water retention determinations**

95 Four different techniques (axis translation, filter paper, vapour equilibrium VET and measurements
96 with a WP4 dewpoint psychrometer) were employed to determine the water retention properties of
97 the CB mixture. The main drying branch of the water retention was determined exposing specimens
98 extracted from the curing bath to increasingly high suctions, whereas the main wetting branch was
99 determined by exposing specimens that had been previously dried in the laboratory environment
100 (fixed temperature of $T=21$ °C and relative humidity $h_r = 0.38$) to progressively lower suctions.

101 The axis translation (Hilf, 1956) and the filter paper techniques (ASTM, 2016) were used either to
102 impose or measure matric suction s . The axis translation technique was implemented in a suction
103 controlled oedometer and the range of suction explored was $0.050 \text{ MPa} \leq s \leq 0.30 \text{ MPa}$. Different
104 values of air and water pressures were imposed independently while the volume of water leaving or
105 entering the specimen was measured with a volume indicator. The height of the specimen was mon-
106 itored with a LVDT. The filter paper technique was implemented using the Scheicher & Schuell no.
107 589 WH filter paper: the range of suction explored was $0.020 \text{ MPa} \leq s \leq 2 \text{ MPa}$. Two twin specimens
108 were used for each determination: along the drying path, they were partially dried through expo-
109 sure to the laboratory air; along the wetting path, fully dried specimens were partially wetted adding
110 known masses of water. Three sheets of filter paper were sandwiched between the specimens and
111 the whole assemblage was sealed hermetically in plastic containers for 15 days before the mass and
112 suction determinations. Changes in size were measured with a calliper.

113 Vapour Equilibrium technique (VET) and a WP4 dewpoint psychrometer (Decagon Devices) were
114 adopted either to impose or measure total suction ψ . The VET was used over a range $9.60 \text{ MPa} \leq$
115 $\psi \leq 55 \text{ MPa}$, suspending the specimens in hermetically closed jars above saturated salt solutions.

116 Changes in the mass of the specimens were measured with a high precision balance and occurred
117 in about 2-3 weeks. Measurements of ψ with the WP4 were taken on specimens that, after exposure
118 to the laboratory environment or addition of water, had been sealed in plastic covers and suspended
119 above pure water for 48 hours to allow for suction homogenization. The mass of the specimens was
120 measured with a high precision balance and their size was measured with a calliper. However, for
121 suctions $\psi > 14$ MPa the specimens became very fragile and use of the calliper impossible. Some of
122 the specimens cracked at high suctions. In this case, where possible, measurements were performed
123 on their larger surviving portions. In this sense, the provided water retention characterization refers
124 to the hydraulic properties of the cement bentonite matrix.

125 The matric suction of the VET and WP4 measurements was determined as:

$$s = \psi - \pi \quad (1)$$

126 where π is osmotic suction, which in diluted solutions is linearly proportional to the concen-
127 tration of species dissolved in the pore water. The osmotic suction at different water contents was
128 evaluated as:

$$\pi = \pi_0 \frac{w_0}{w} \quad (2)$$

129 w_0 and w being the initial and the current water content and π_0 being the osmotic suction of the
130 curing bath, approximately equal to 285 kPa according to WP4 measurements. Note that the relatively
131 high value of π_0 has to be related, at least partially, to the dissolved species present in the bentonite
132 powder and to the reaction products of the hydration of the cement.

133 **2.3 Microstructural determinations**

134 Microstructural effects induced by relatively high suctions on specimens cured for 28 days were in-
135 vestigated by means of Scanning Electron Microscopy (SEM) analyses. Three conditions were in-
136 vestigated: fully saturated ($s = 0$ MPa, specimen as extracted by the curing bath), dry ($s \approx 129$ MPa,
137 induced by drying at the laboratory conditions) and at an intermediate suction ($s \approx 4$ MPa, obtained
138 conditioning the specimen with the VET). The specimens, with the exception of the dry one, were
139 preliminarily freeze-dried to avoid undesired changes in the microstructure due to oven drying (see
140 *e.g.* Delage and Pellerin 1984).

2.4 Strength determinations

A preliminary characterization of the effects of suction/water content on the strength of the CB samples was pursued measuring the tip resistance of a pocket penetrometer during sample drying. The values of resistance reported in the following are the average of three determinations. A commercial pocket penetrometer from Controls with a range 0-5 MPa was used.

2.5 Electrical conductivity and ERT reconstructions

Electrical Resistivity Tomography (ERT) is a technique that allows the reconstruction of the electrical conductivity field within an object, based on the inversion of electrical measurements performed at the boundaries of the object itself. Injecting and measuring electrodes are needed, and their position is changed until a number of measurements sufficient for the tomographic reconstruction is performed (Borsic *et al.*, 2005). The information provided by the distribution in space and time of the electrical conductivity is very useful for geo-environmental applications: it has been in fact successfully used as a proxy to detect heterogeneities (Borsic *et al.*, 2005), porosity changes, distribution of contaminant in soil samples (Comina *et al.*, 2011), also allowing estimating the water retention and hydraulic properties of different soils (Cosentini *et al.*, 2012). It also has been used to investigate moisture movement in cement, both undamaged (Smyl *et al.*, 2016) – thus allowing to study the homogeneous behaviour of the material matrix – and cracked (Smyl *et al.*, 2017). A 2D reconstruction protocol (as in Musso *et al.* 2020) was set up, relying on measurements taken from 16 electrodes equally spaced along the circumference of the CB samples. Electrodes were thin silver squared foils of approximately 5 mm sides, welded to silver wires. The electrodes were placed inside the moulds before pouring the mixture for curing.

In order to provide a reliable map of water content distribution, a relationship between electrical conductivity and the amount of water in the porous material is needed. Theoretical relationships are available relating the electrical conductivity of rocks and soils to water content, porosity and mineralogy: however, to the authors' knowledge, their validity has not been checked for cement bentonite mixtures and in this study a phenomenological law based on a preliminary characterization was preferred. Such a characterization was performed on cylindrical specimens of height $H = 80$ mm and cross section $S = 507 \text{ mm}^2$. Two cylindrical electrodes, A and B, were applied at the ends of the cylinders, while two 'needle' electrodes (M and N) were inserted along the side, at a distance $L = 40$ mm one from the other and equally spaced from the ends (scheme of the apparatus in Figure 1).

The electrical conductivity χ was evaluated as:

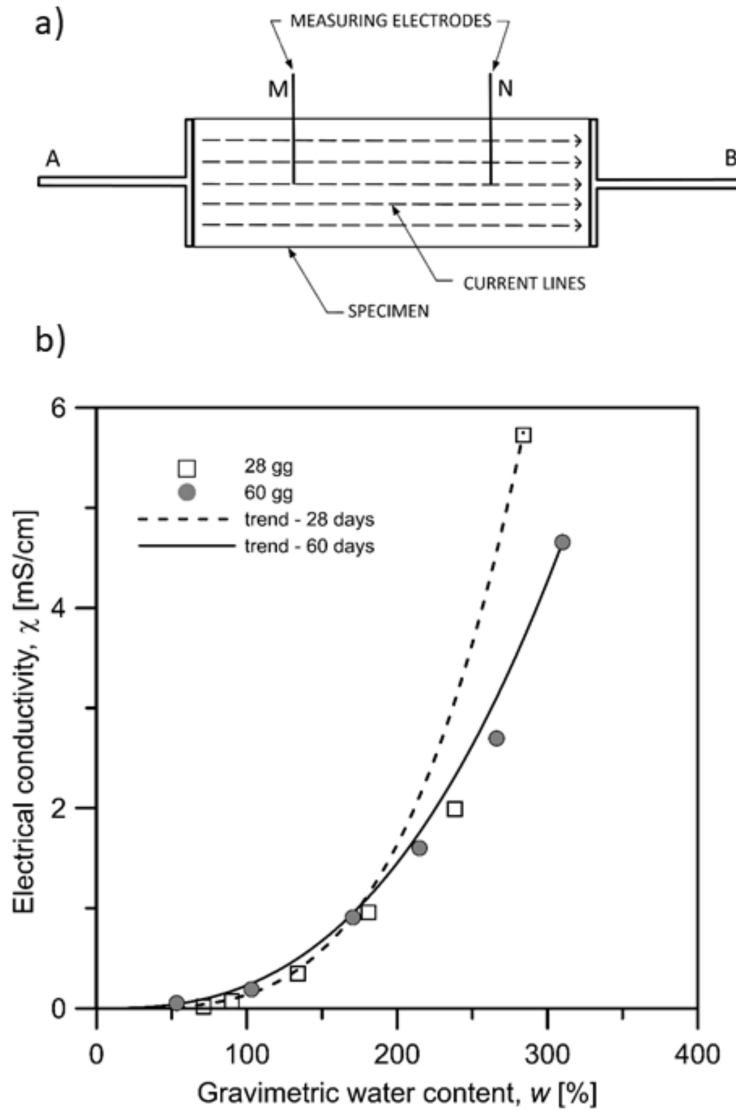


Figure 1: a) scheme of the equipment used for determining the relationship between water content and electrical conductivity b) relationship between water content and electrical conductivity

$$\chi = \frac{\Delta V L}{I S} \quad (3)$$

172 where I is the electrical current applied by electrodes A and B and ΔV is the drop of electrical poten-
 173 tial between M and N.

174 The first measurement was performed immediately after removing the specimens from the wa-
 175 ter bath where they were cured, starting from the initial water content. Other measurements were
 176 performed at decreasing water contents after a controlled air drying procedure. The specimens were
 177 exposed to the laboratory air for a few hours, then wrapped up in a plastic cover and left to equili-
 178 brate in a humid environment for 24 hours (Zibisco, 2019). They were then weighted before a new
 179 electrical test. Volume changes were determined with a caliper. The procedure was repeated down

to a water content $w = 53\%$. Lower water contents were not investigated, due to the progressive increase in sample fragility and the large reduction in electrical conductivity. Experimental evolution of electrical conductivity with water content is shown in Figure 1 b. Experimental data are fitted with equation (4):

$$\chi = \chi_0 \left(\frac{w}{w_0} \right)^\beta \quad (4)$$

where χ_0 and w_0 are the electrical conductivity and the gravimetric water content of the saturated specimens and β is a parameter that depends on the curing time, equal to 3.59 for 28 days of curing and to 2.66 for 60 days of curing. The electrical conductivity and the water content of the specimens as extracted from the curing bath were respectively $\chi_0 = 5.75$ mS/cm and $w_0 = 284$ % (28 days of curing), $\chi_0 = 4.66$ mS/cm and $w_0 = 310$ % (60 days of curing).

3 Water retention and volume changes along drying and wetting

The measured water retention curve for the CB mixture is shown in Figure 2, both in terms of liquid degree of saturation S_l and volumetric liquid content w . The wetting and drying branches of the water retention were fitted by means of the van Genuchten equation (Eq.5):

$$S_l = S_{res} + (1 - S_{res}) \left[1 + (\alpha s)^n \right]^{-m} \quad (5)$$

where S_{res} is the residual degree of saturation and α , n and m are model parameters whose values are reported in Table 1.

Table 1: van Genuchten water retention curve parameters

	main drying	main wetting
α [1/MPa]	0.38	30
n [-]	2.05	0.70
m [-]	0.73	0.60
S_{res} [-]	0.04	0.04

A large hydraulic hysteresis is appreciated in the (S_l, s) plane: along the main drying path the mixture remains nearly saturated up to matric suctions of about $s = 800$ kPa while along the main wetting path the degree of saturation is as low as $S_l = 0.60$ for $s = 50$ kPa. Differences between the main drying and the wetting branches are even larger in the (w, s) plane. This is caused by the volumetric

200 behaviour of the mixture, shown in Figure 3. Upon drying, changes in the void ratio are very small up to
 201 a threshold matric suction of about 4-5 MPa, while a strong shrinkage occurs at higher suctions.
 202 Upon wetting from dry conditions swelling is very limited and the volume remains almost constant.

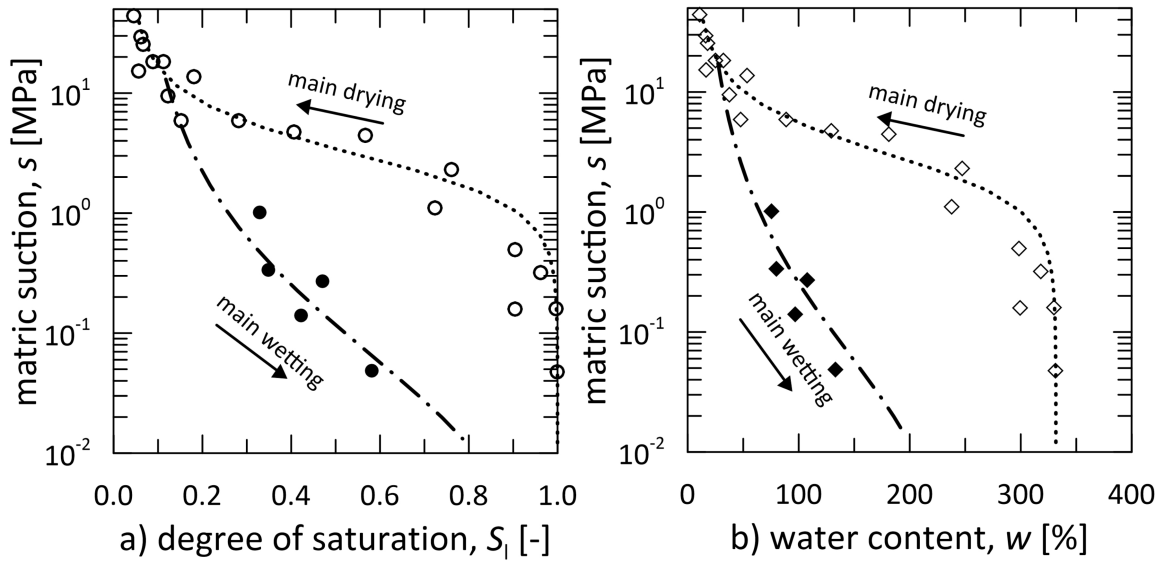


Figure 2: Water retention curves in terms of: a) degree of saturation; b) gravimetric water content.

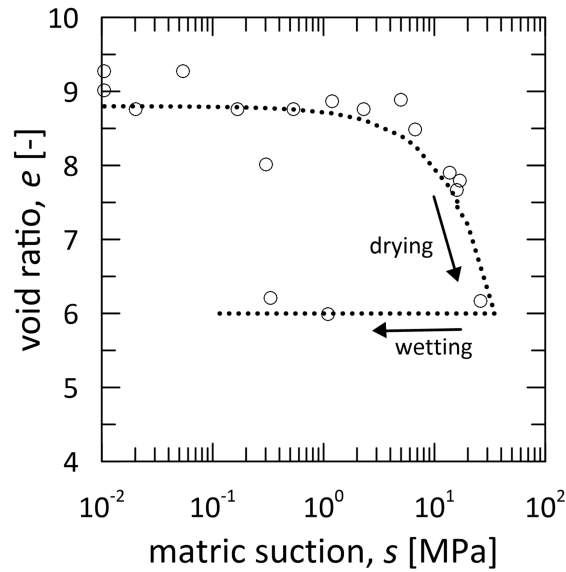


Figure 3: Void ratio evolution along drying and wetting paths

203 An insight on the microstructural effects which cause such large hydraulic hysteresis and non
 204 reversible volumetric behaviour was given by SEM investigations. Figure 4 provides images obtained
 205 with a 500 x magnification for a saturated specimen, for a specimen dried to $s \approx 4$ MPa and for a
 206 fully dried specimen. The saturated specimen (left) has a quite homogeneous matrix structure, with
 207 a uniform distribution of cement-bentonite clusters. The size of these clusters is reduced at $s \approx 4$
 208 MPa, which causes the onset of micro-cracks with a very small aperture (elongated 'pores' which

209 run diagonally from the bottom left to the top right of the central image in Figure 4). In fully dry
210 conditions (right image in Figure 4) the CB clusters largely shrank and the micro-cracks enlarged.

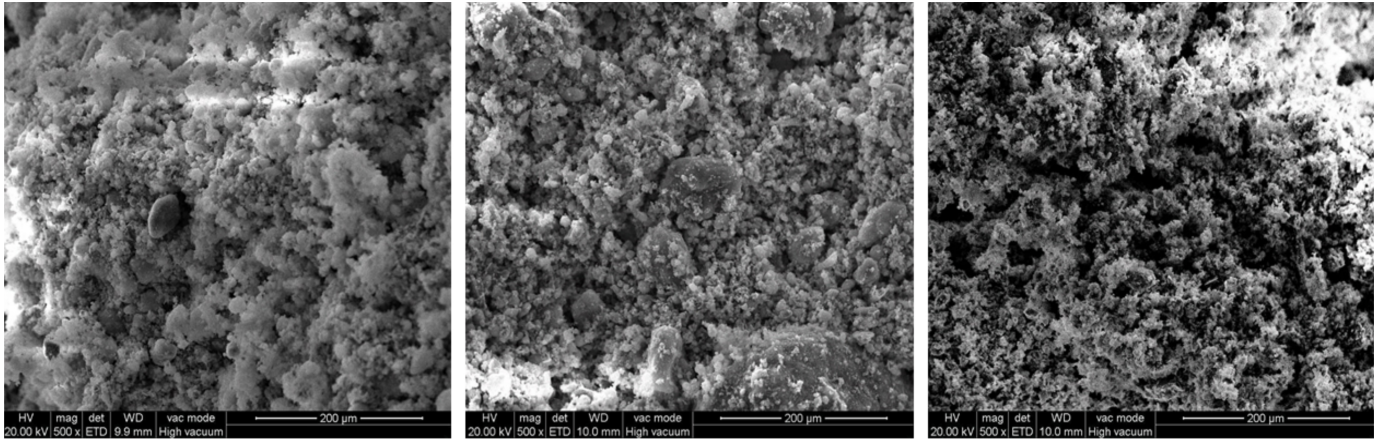


Figure 4: SEM images of: a) saturated specimen; b) specimen equalised at $s \approx 4$ MPa; c) dry specimen. Bar length is $200 \mu\text{m}$ in all images

211 4 Evaporation and water uptake tests

212 An experimental investigation on the processes related to evaporation and water uptake in CB mix-
213 tures was carried out. Tests were run on squat samples with initial height $H = 40$ mm and diameter D
214 $= 110$ mm and on a bench-scale column sample with initial height $H = 300$ mm and diameter $D = 130$
215 mm.

216 The samples were taken out of the curing bath whereas they were kept inside the mould into
217 which they were cured. Evaporation was induced by exposing their top surface to the laboratory
218 environment (temperature $T = 21^\circ\text{C}$ and relative humidity $h_r = 0.38$, total suction $\psi \approx 131$ MPa).
219 During the drying stage, stage 1 in the following, they were placed on a plastic surface to which their
220 bottom was sealed. During the water uptake stage (see Figure 5), stage 2 in the following, they were
221 immersed in water for two days (water height 10 mm height for the squat samples and 40 mm for the
222 column sample). Test details are reported in Table 2.

223 4.1 Squat samples

224 These tests were aimed at checking the use of ERT as a tool for indirect measurements of the water
225 content of CB mixtures, at gaining a qualitative observation of cracking and at obtaining a prelim-
226 inary evaluation of the relationship between strength and water content, determined with pocket
227 penetrometer measurements during drying. Two different curing times were considered: in the fol-

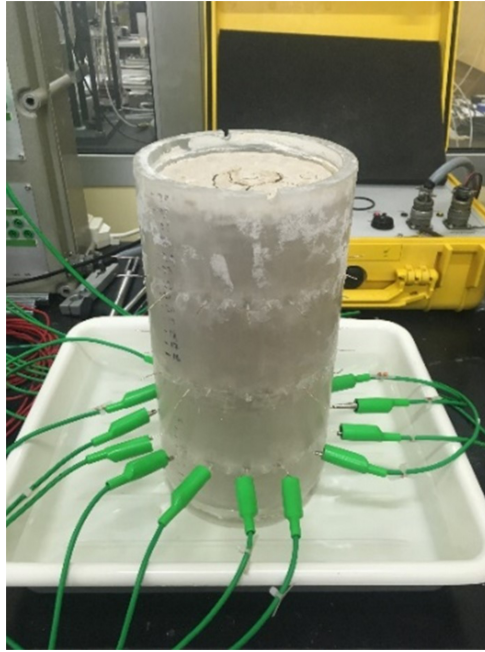


Figure 5: Stage 2 (water uptake) of the experiment on the columnar sample.

Table 2: Details of the evaporation and water uptake tests

	Squat samples		Column sample
	test A	test B	
height [mm]	40	40	300
diameter [mm]	110	110	130
curing [day]	28	60	60
duration of stage 1 [days]	10	8	13
duration of stage 2 [day]	2	2	2
initial void ratio, e [-]	8.05	7.80	6.60 – 8.50

lowing 'test A' stands for 28 days of curing and 'test B' for 60 days of curing. Since pocket penetrometer are destructive measurements, two samples were used for each curing time: one of them for crack detection and ERT, the other one for the penetrometer tests.

The average water content was physically measured by weighting the samples on a high precision balance, while it was also reconstructed by coupling the electrical conductivity maps from the ERT with the equation (4). As shown in Figure 6, loss of water content proceeded quite linearly with time, at a rate that was slightly higher in test B with respect to test A. ERT provided a very good estimate of the average water content for the initial stages of the tests, during which cracking was not detected through naked eye observations and homogeneous maps of water content were obtained (Figure 7).

At longer times (more than 4 days for test A and 7 days for test B) ERT estimations diverged from physical measurements. This mainly occurred because of an increasing number of errors in the electrical measurements due to poor contact between the electrodes and the samples and to crack oc-

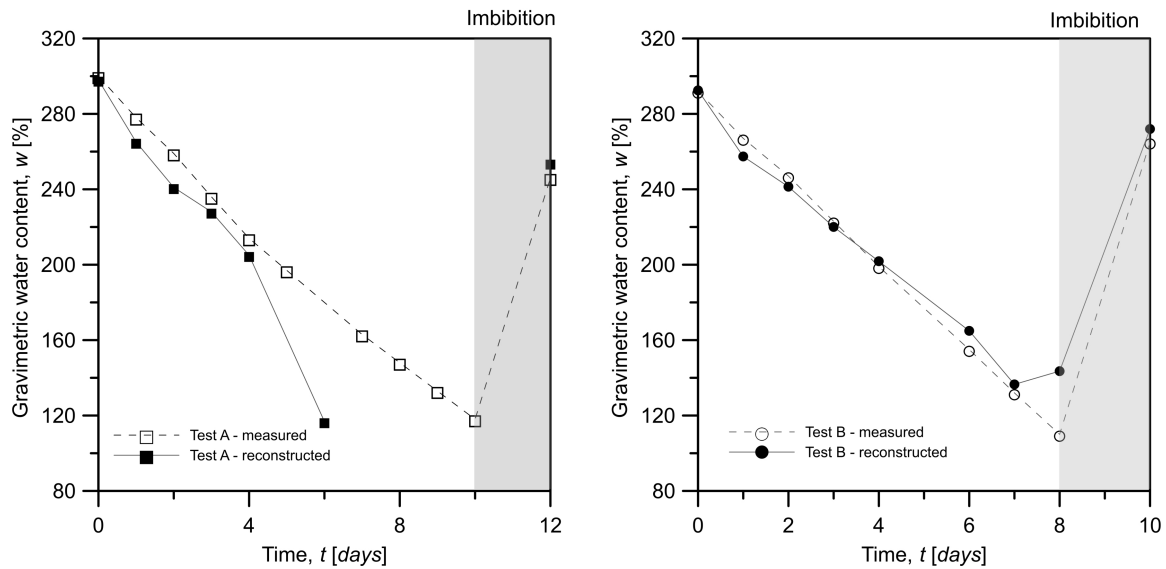


Figure 6: Water content evolution of squat samples during the evaporation and water uptake tests. a) Test A; b) Test B

240 currence, which started to develop in Test A after about 7 days. The water content was then $w \approx 160$
 241 % , which according to the water retention in Figure 2 corresponds to a suction $s \approx 4$ MPa. The state
 242 of such sample at the end of the evaporation phase, *i.e.* after 10 days, is presented in Figure 8. While
 243 at this point water content estimation proved to be totally unreliable, ERT was still able to detect the
 244 crack pattern that had developed as areas of very low electrical conductivity, although the thickness
 245 of the discontinuities is not matched. In general, ERT reconstructions were able to detect cracks hav-
 246 ing a thickness of the order of 2-3 mm. Notice that the thickness of the inclusion that can be matched
 247 with the electrical tomography strongly depends on the Finite Element mesh used in the numerical
 248 inversion procedure (see Borsic *et al.* 2005). As for test B, no evident cracks were detected through
 249 visual inspection or ERT monitoring.

250 The water content increased quickly during the water uptake stage, although in both tests it did
 251 not recover its initial value. Interestingly, errors in the electrical measurements decreased with water
 252 uptake and the estimate of the average water content based on the ERT matched again very well with
 253 the physical one (Figure 6).

254 Figure 9 shows the evolution with the water content of the tip resistance detected with the pocket
 255 penetrometer on the samples matrix. For both curing times, the tip resistance increases as the water
 256 content decreases, *i.e.* as the mixture desaturates and suction increases. At any water content, the
 257 resistance of the test B is higher than the one of the test A, which might contribute explaining why
 258 significant cracks did not develop during test B.

259 Some complementary Unconsolidated Undrained triaxial tests were carried out on specimens
 260 cured for 28 and 60 days. The uniaxial compressive strength was 38 kPa after 28 days and 100 kPa after

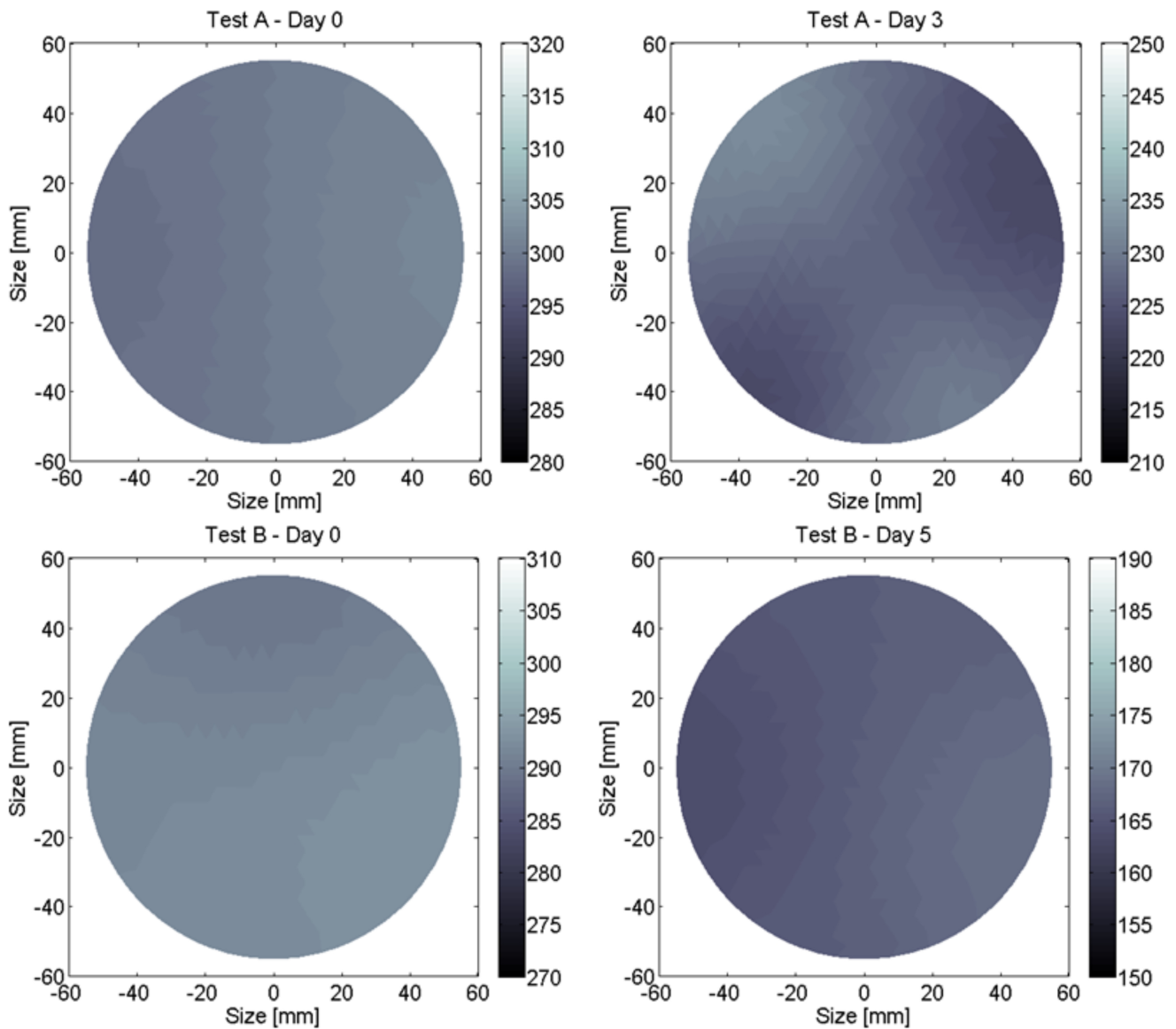


Figure 7: ERT reconstructed water content distribution during drying. Colorbars are gravimetric water contents in %

261 60 days. Compared with Figure 8, these results suggest that the pocket penetrometer measurements
 262 provide a rough estimation of the uniaxial compressive strength, as also pointed out in the literature
 263 (see *e.g.* Blum 1997). The uniaxial compressive strength after one month of curing was also roughly
 264 comparable to the one obtained on a similar mixture for the same curing time by Evans and Opdyke
 265 (2006).

266 4.2 Column sample

267 The main purpose of the column test was to observe local evolutions of the water content during
 268 drying and water uptake, also detecting the possible development of cracks. Relying on the results
 269 obtained with the squat samples, arrays of electrodes were disposed at three different heights from

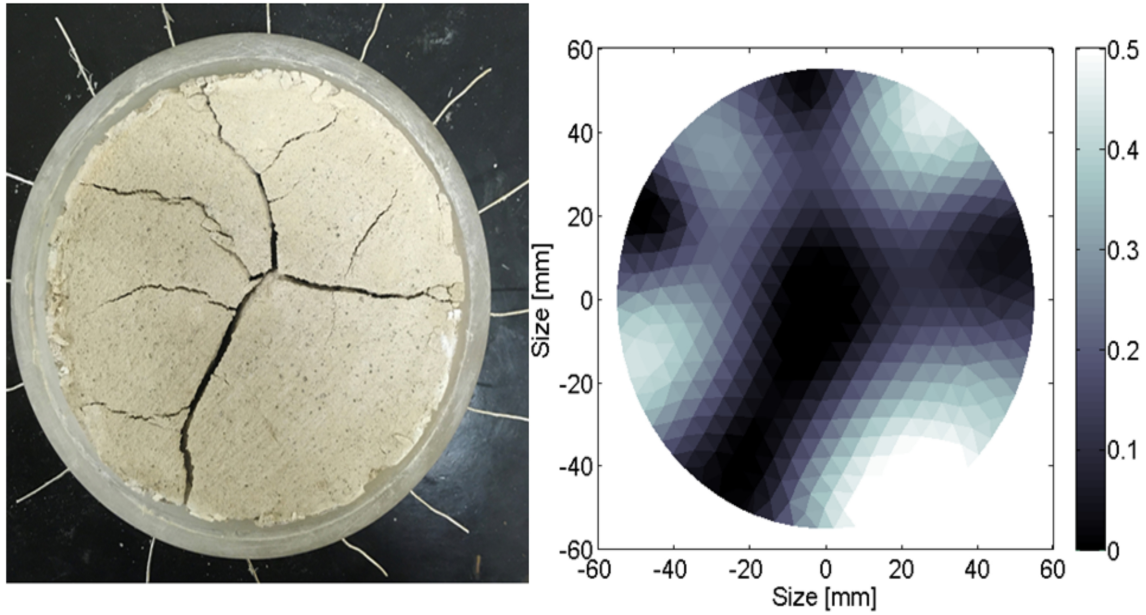


Figure 8: Picture of test A sample sample after 10 days of drying (left) and corresponding map of electrical conductivity (right, values in mS/cm).

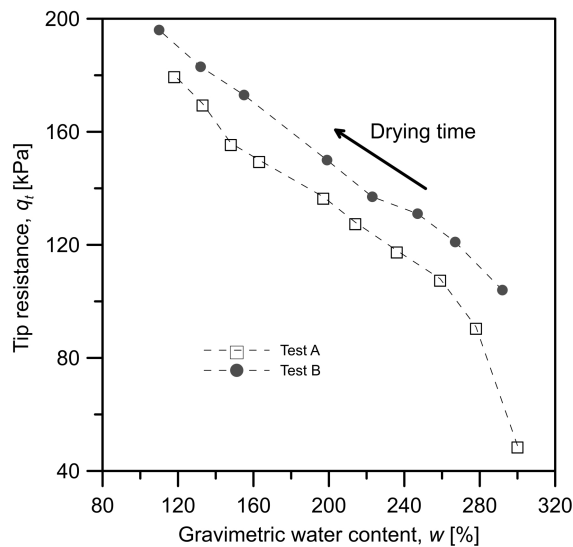


Figure 9: Relationship between water content and tip resistance from pocket penetrometer measurements

270 the bottom of the column (respectively $z_1=75$ mm, $z_2=150$ mm and $z_3=225$ mm) to run ERT recon-
 271 structions from which local evaluations of the water content in time were deduced. The void ratio of
 272 this sample, determined through mass measurements, varied with depth and it was $e = 6.60$ at the
 273 bottom and $e = 8.50$ at its top. Such variation is likely to be due to the self-weight induced consolida-
 274 tion taking place during curing,

275 Figure 10 shows the evolution with time of the water content at the monitored points. Higher
 276 water contents pertain to the upper layers, which have a larger void ratio. The ERT results suggest a
 277 limited loss of water mass during the 13 days of drying (stage 1): the largest decrease in water content

278 corresponded to z_3 ($\Delta w \simeq 40\%$) whereas the smallest corresponded to z_1 ($\Delta w \simeq 25\%$). As already
 279 detected with the squat samples, with the water uptake the water content increased, but it did not
 280 fully recover its initial value. This was surely due to the irreversible reduction in the void ratio and to
 281 the development of microcracks along drying as described in Section 3. A further contribution could
 282 come from the persistence of air bubbles, providing an irreducible air saturation at the pressure given
 283 by the limited water height imposed at sample bottom. The water content increased well within 60
 284 minutes from the beginning of the imbibition stage and then remained about constant.

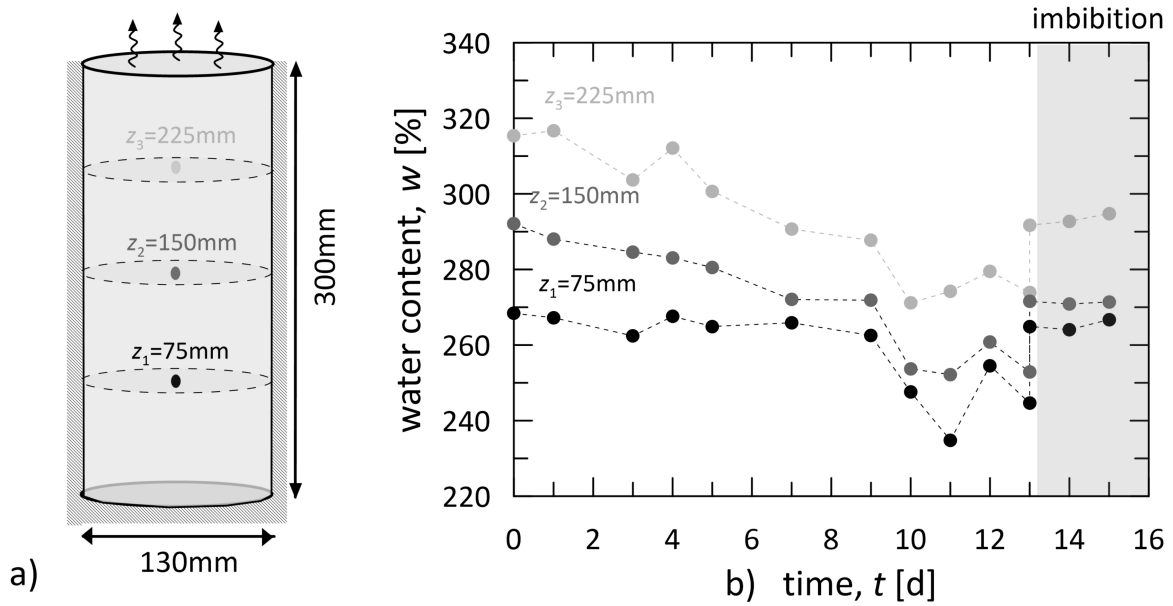


Figure 10: Results of the evaporation and water uptake bench scale test: trends with time of the water content along the column sample as evaluated by ERT

285 5 Modelling of evaporation and water uptake

286 Numerical simulations were run to achieve a further understanding of the evaporation and water
 287 uptake processes. The balance of water mass as a chemical species and of the thermal energy of the
 288 mixture in unsaturated conditions were implemented and solved with the Finite Element commer-
 289 cial code Comsol Multiphysics[®]. 1D models were set up to reproduce the laboratory tests discussed
 290 in the previous sections. A full description of the Thermo-Hydraulic (TH) model and of the related
 291 equations is provided in the Appendix. A continuum approach was adopted, *i.e.* the presence of
 292 fractures was not introduced. Changes in porosity with suction were accounted for introducing an
 293 empirical equation based on the results presented in figure 3. The hydraulic parameters obtained
 294 from the experimental results described in Section 3 were used, together with the van Genuchten
 295 relationship for relative permeability function and a linear scanning equation $ds_l = -k_s ds$ with

$k_s = 4 \cdot 10^{-5} \text{ 1/kPa}$ for the transition from the main wetting to the main drying retention curve. Thermal parameters were estimated using relationships from the literature accounting for the properties of the single phases.

The initial and boundary conditions for the balance equations of water mass and energy, summarised in Table 3, were chosen to reproduce laboratory conditions. A hydrostatic water pressure distribution and a uniform temperature, equal to the laboratory one $T = 21^\circ\text{C}$, were adopted as initial conditions. The hydraulic and heat fluxes induced by evaporation and briefly discussed in the following (Eqs.6 and 9, respectively) were imposed at the top boundary. At the bottom boundary either a no hydraulic flow or a water pressure corresponding to the imposed water height h_w were imposed, so to reproduce the drying and the water uptake stage respectively. A constant temperature of $T_{amb} = 21^\circ\text{C}$ was applied for both stages.

Table 3: Initial and boundary conditions

	Water mass balance	Energy balance
Initial conditions		
$t = 0$	$u_w = \rho_w g(z - h)$	$T = T_{amb}$
Boundary conditions: evaporation stage		
top ($z = h$)	evaporative flux Eq.(6)	heat flux Eq.(9)
bottom ($z = 0$)	no flux	$T = T_{amb}$
Boundary conditions: imbibition stage		
top ($z = h$)	evaporative flux Eq.(6)	heat flux Eq.(9)
bottom ($z = 0$)	$u_w = \rho_w g h_w$	$T = T_{amb}$

The outflow of water mass q_{evap} due to evaporation was evaluated as a function of the actual rate of evaporation AE , defined as (Penman, 1956):

$$q_{evap} = \rho_l AE; \quad AE = PE \frac{p_{v,soil} - p_{v,amb}}{p_{v,sat} - p_{v,amb}}; \quad (6)$$

where ρ_l is the water mass density, PE is the potential rate of evaporation, $p_{v,soil} = p_{v,sat}(T)h_r(T_{soil})$ is the current vapour pressure in the soil at soil temperature T_{soil} , $p_{v,amb} = p_{v,sat}(T_{amb})h_r(T_{amb})$ is the vapour pressure in the environment, and $p_{v,sat}$ is the saturated vapour pressure, related to temperature according to Kendall and Caldwell (1998)

$$p_{v,sat} [\text{kPa}] = 0.6108 \cdot 10^{\frac{7.5(T[^\circ\text{C}]-273.15)}{T[^\circ\text{C}]}}. \quad (7)$$

while the relative humidity is linked to total suction ψ through the psychrometric law (8):

$$h_r = \exp\left(-\frac{\psi M_w}{\rho_l RT}\right), \quad (8)$$

where M_w is the molar mass of the water, R is the constant of perfect gases.

In testing conditions, PE was determined placing a mold filled of water in the laboratory environment for seven days and monitoring its weight loss in time with a balance. The measured potential evaporation was $PE = 2.58$ mm/d.

Due to the thermo-hydraulic coupling, the evaporation process also acts as a heat sink that causes a reduction of temperature. On the other hand, the difference of temperature between the soil and the environment generates a heat flux which aims at re-establishing equilibrium. The thermal flux that was imposed at the upper boundary, subjected to evaporation, was thus (Ralaizafisolariovony *et al.*, 2020):

$$q_{heat} = -L_w q_{evap} + \epsilon_s \sigma (T_{amb}^4 - T^4); \quad (9)$$

where L_w is the latent heat of vaporization, $\epsilon_s = 0.9 + 0.18w$ is soil emissivity, linked with gravimetric water content w according to Van Bavel and Hillel (1976), and $\sigma = 5.670367 \cdot 10^{-8}$ kg s⁻³K⁻⁴ is Stefan-Boltzmann constant.

5.1 Squat samples

Figure 11 shows the model predictions in terms of evolution of the hydraulic state at centre of the squat samples (height 20 mm). At the end of the drying stage, the predicted average degree of saturation for the two samples is around $S_l = 0.3$ and 0.5 respectively (see Figure 11.a), thus far from the residual degree of saturation. When the imbibition begins, suction starts to decrease due to the imposed water head at the bottom boundary, and the samples increase their water content in accordance to the scanning retention path shown in Figure 11. The scanning path of the water retention curve is followed until reaching the main wetting branch.

Figure 12 reports the predicted evolution of the average water content with time, compared with experimental data. Model predictions are satisfactory and in the simulations the initial water content was not completely recovered after the imbibition phase being the final degree of saturation lower than 1. The variation of the void ratio due to the shrinkage induced by drying is not affecting much the hydraulic behaviour of the material, as the maximum values of suctions achieved are around 5 MPa (see Figure 11), that corresponds -according to Figure 2- to a negligible void ratio change.

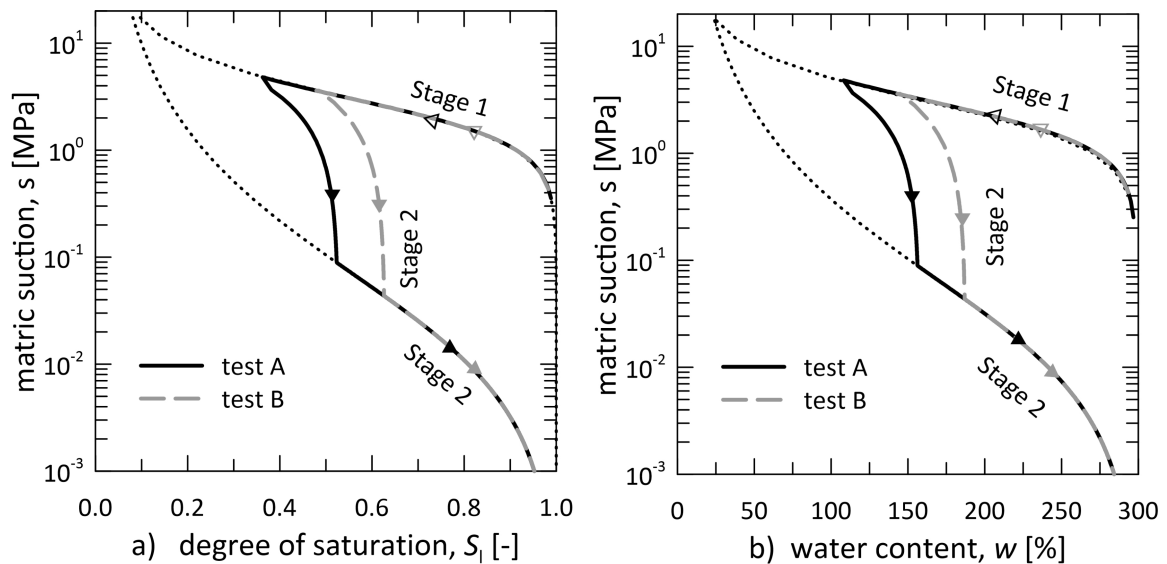


Figure 11: Drying and water uptake paths at the centre of the squat samples a) in the degree of saturation -suction plane; b) in the water content -suction plane

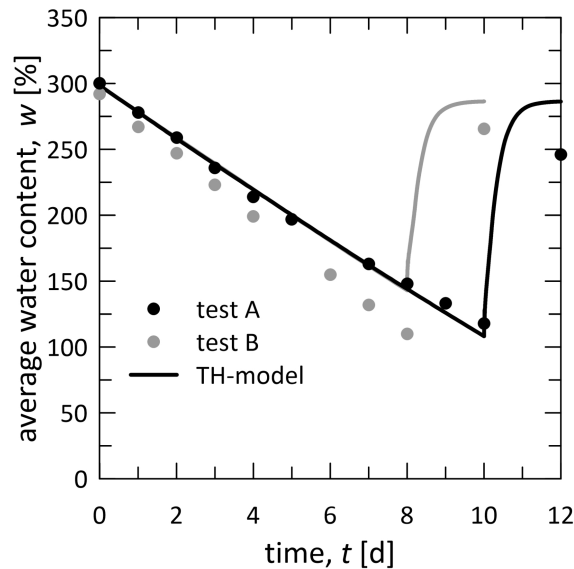


Figure 12: Average water content with time: models prediction *versus* experimental data

340 However, the analysis slightly overestimates the final water content. This seems to suggest that mass
 341 of water retained along the main wetting path at low suctions is less than what predicted through eq.
 342 5, being the parameter reported in Table 1 determined at suctions $s > 50$ kPa.

343 5.2 Column sample

344 Figure 13 compares the predicted and the experimental evolution of water content at the column
 345 monitoring points. A quite good match between the experimental data and the simulations is found,
 346 although the model anticipates and slightly overestimates the decrease in water content in corre-
 347 spondence of z_1 . The partial recovery of water content caused by the water uptake is nicely repro-

348 deduced, both in terms of water amount and of time evolution.

349 Figure 14 shows the predicted hydraulic paths: suction values at the different depths are quite ho-
 350 mogeneous, and the differences in the water content is predominantly due to the depth-dependency
 351 of the initial void ratio. At the end of the drying stage, suction ranges around 1 MPa and the sample
 352 is very far from full desaturation, so that the water uptake stage is governed by the scanning water
 353 retention curve path, that allows the sample to recover about the 20% of water content.

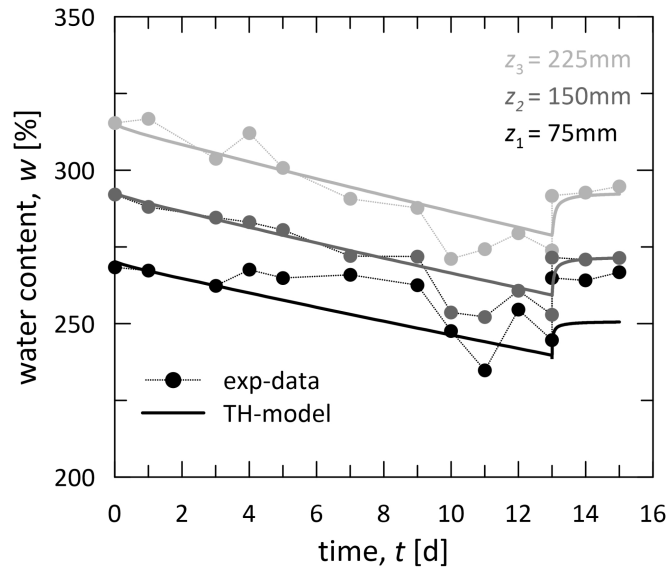


Figure 13: a) Evolution in time of the water content in the column sample. Model predictions *versus* experimental data.

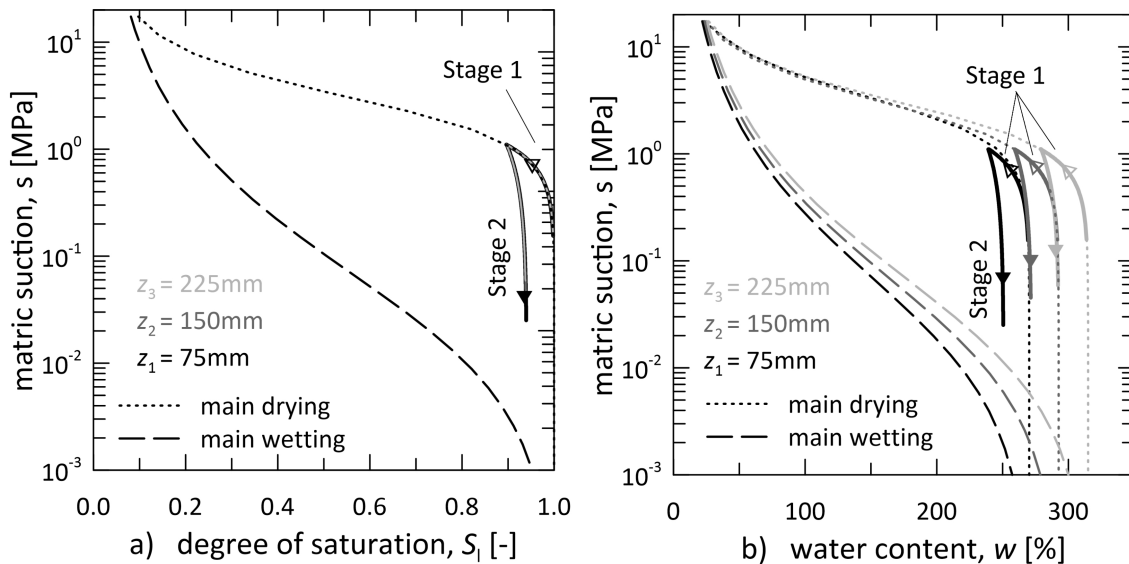


Figure 14: Drying and water uptake paths of the column test a) in the degree of saturation -suction plane; b) in the water content -suction plane.

6 Conclusions

The results of this laboratory investigation allow drawing some conclusions on the behaviour of a CB mixture upon drying and wetting. Although the water retention properties along the main drying path were quite good, with an Air Entry Value of around 800 kPa, a large hydraulic hysteresis was found and the main wetting curve had significantly lower retention capabilities. According to SEM evidence, this might be due to the retraction of cement-bentonite clusters, which leads to the opening of small microcracks: these are well evident at a threshold suction of about 4 MPa and they still enlarge as drying proceeds. Retraction might be only partially reversible, as it is also confirmed by the plastic volumetric strains that develop once that the mixture is exposed to high suctions.

In the laboratory environment, evaporation was able to cause an appreciable water content decrease only when the bottom of squat and column samples was sealed (*i.e.* no water recharge was made available). In this case, the mixtures dried. Cracking of squat samples upon drying occurred above a suction of about 4 MPa for samples cured for 28 days, while it was not evident over the same range of suctions for samples cured for 60 days. A possible reason for this difference might be the increase of the material strength with the curing time. Tests were also monitored with Electrical Resistivity Tomography (ERT), which proved to be effective in determining the water content of the mixture at relatively high water contents. Although at lower water content ERT estimates might be erroneous, it can still provide a reasonable detection of crack patterns when they arise, which suggests that ERT might be fruitfully used for on-site integrity inspections.

Results of an evaporation and water uptake bench scale test on a CB column showed that, while water loss occurs at a relatively slow pace, water uptake occurs in very short time. The initial water content was not recovered in any test, even if the samples had experienced relatively modest suctions (order of a few MPas). For all the laboratory experiments, the transient processes of evaporation process and water uptake were adequately reproduced by continuous models accounting for water and heat transport in unsaturated state, which were built upon the material properties deduced from water retention and permeability testing.

Field evidences discussed in the introduction (Jefferis, 2012; Soga *et al.*, 2013) are inconclusive with respect to the question whether cement-bentonite cutoff walls might crack or not during their service life. The experimental results of this study suggest that, besides on the mechanical constraints acting in situ, this is clearly related to the suction acting in the barrier. On its turn, suction in the wall would not only depend on the interaction with the atmosphere of the site, but also on the amount of water made available by the surrounding soils. In fact, in the experimental data shown in the paper

386 water content changes were very small when a free drainage condition was imposed at the bottom of
387 the column sample, while they were more pronounced when the bottom was sealed. The method-
388 ology here presented (water retention and mechanical characterization, modelling based on mass
389 and energy balance equations) could help the practitioners to identify the risk of cracking account-
390 ing for the actual stratigraphy and the environmental conditions of the site. In this case, however, the
391 definition of the soil-atmosphere boundary conditions should be updated to account for environ-
392 mental features such as wind velocity, solar radiations, daily and seasonal variation of temperature
393 and relative humidity.

394 **Acknowledgements**

395 Tests on squat and column samples were run by A. Zibisco, whose contribution is gratefully acknowl-
396 edged. The activity of one of Dr. Giulia Guida was financed in the context of the public administra-
397 tion agreement between Politecnico di Milano - Department of Civil and Environmental Engineering
398 and the Italian Ministry of Economic Development, Direzione Generale per la Sicurezza anche Am-
399 bientale delle Attività Minerarie ed Energetiche–Ufficio Nazionale Minerario per gli Idrocarburi e le
400 Georisorse–Programme Clypea, which is here gratefully acknowledged.

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Appendix: Formulation of the Thermo-Hydraulic model for the drying and water uptake problem

Evaporation is a complex process involving simultaneous heat and mass transfer, phase changes, and inter-facial liquid and vapour transfer at pore scales (Saito *et al.*, 2006; Shokri *et al.*, 2010; An *et al.*, 2018). Pore water chemistry and material deformation can play a relevant role too, despite in many application a set of equations including just the coupled transient movement of liquid water, water vapour, and heat in the soil is sufficient (Wilson, 1990). In this work, evaporation was simulated by integrating the coupled water mass balance and the mixture energy balance equation (Thermo-Hydraulic model, T-H). Changes in porosity were accounted for in both balance equations, accordingly with the suction-void ratio relationship of Figure 3.

Water mass balance

It is well recognized that during evaporation processes water may flow to the ground surface as both liquid water and water vapour. If fluid phases are accounted for, molecules of water and dissolved air compose the liquid phase, while molecules of dry air and water vapour compose the gas phase. Trough evaporation, molecules of water are exchanged between the liquid and the gas phase. In order to avoid modelling the transfer of mass between the two phases and the mass balance of the dry air, water liquid and vapour, the mass balance equation was simply imposed on water as a chemical species as:

$$\frac{\partial}{\partial t} \left[\phi (S_l c_l^w + S_g c_g^w) \right] + \nabla \cdot \left(c_l^w \mathbf{q}_l + c_g^w \mathbf{q}_g \right) + \nabla \cdot \phi \left(S_l \mathbf{J}_l^w + S_g \mathbf{J}_g^w \right) = S_l \phi \rho_l \Gamma_l^w + S_g \phi \rho_g \Gamma_g^w \quad (10)$$

where ϕ is porosity, S_l and S_g are the degree of saturation of the liquid and the gas phase respectively ($S_l + S_g = 1$), c_l^w and c_g^w are the water mass concentration in the liquid and gas phase, ρ_l and ρ_g are the liquid and gas mass density, \mathbf{q}_l and \mathbf{q}_g are the advective fluxes of the liquid and gas phase, \mathbf{J}_l^w and \mathbf{J}_g^w are the sum of diffusive and dispersive fluxes of water in liquid and gas phase, Γ_l^w and Γ_g^w are the generation rate of water mass per unit mass of liquid and gas phase. For the simulation of evaporation processes, it was accepted that the water mass balance equation may be simplified by neglecting the source terms, the dispersive flux of water vapour, the presence of air dissolved in the liquid (which implies $c_l^w = \rho_l$, considered constant throughout the paper, and $\mathbf{J}_l^w = 0$) and by assuming that air pressure is constant over space and time (which implies $\mathbf{q}_g = 0$). It follows that:

$$\frac{\partial}{\partial t} \left[\phi(S_l \rho_l + S_g c_g^w) \right] + \nabla \cdot (\rho_l \mathbf{q}_l) + \nabla \cdot \phi(S_g \mathbf{J}_g^w) = 0. \quad (11)$$

The specific discharge of liquid water \mathbf{q}_l in the unsaturated material was expressed by means of a generalized Darcy law as

$$\mathbf{q}_l = -\frac{\mathbf{K}_w(S_l)}{\rho_l g} \nabla(u_l + \rho_l g z), \quad (12)$$

where \mathbf{K}_w is the unsaturated hydraulic conductivity, g is the gravitational acceleration, u_l is the liquid water pressure and z is the elevation head. The unsaturated hydraulic conductivity was estimated as the product between the saturated one \mathbf{K}_w^{sat} and the relative permeability coefficient k_r , which in turn was set to depend on water degree of saturation according to the van Genuchten equation

$$\mathbf{K}_w = \mathbf{K}_w^{sat} \cdot k_r = \mathbf{K}_w^{sat} \cdot S_e^{0.5} \left[1 - (1 - S_e^{1/m})^m \right]^2, \quad (13)$$

being $S_e = \frac{S_l - S_{res}}{1 - S_{res}}$ the effective water degree of saturation, S_{res} the residual degree of saturation and m a material parameter.

The diffusive flux of water vapour in the unsaturated material was expressed through Fick's law as

$$\mathbf{J}_g^w = -\mathbf{D}(S_g) \nabla c_g^w, \quad (14)$$

where $\mathbf{D} = D_v \boldsymbol{\tau}(S_g)$ is the water vapour diffusion tensor of the porous medium, that was set equal to the product of the diffusion coefficient of vapour in free air D_v and the tortuosity tensor $\boldsymbol{\tau}(S_g)$, which accounts for the tortuosity of the path of water vapour molecules through the porous medium. Following Lai *et al.* (1976), it was assumed $\tau = (\phi S_g)^{2/3}$. The vapour diffusivity coefficient in free air depends in turns on temperature, $D_v [\text{m}^2/\text{s}] = 0.229 \cdot 10^{-4} (1 + T[\text{K}]/273)^{1.75}$ (Kimball *et al.*, 1976).

The relation between the degree of saturation of the liquid phase S_l and matric suction $s = u_g - u_l$ is given by the water retention curve, that in this work was expressed by means of the van Genuchten equation (5), with parameters provided in Table 1.

Vapour concentration c_g^w is *viceversa* related to the relative humidity h_r (Eq.8), and then on total suction ψ via the psychometric law:

$$c_g^w = c_{g,sat}^w h_r, \quad (15)$$

where $c_{g,sat}^w$ is the saturated concentration of water vapour. The ideal gas law allows the evaluation

554 of $c_{g,sat}^w$ as a function the saturated vapour pressure of water vapour $p_{v,sat}$:

$$c_{g,sat}^w = \frac{p_{v,sat} M_w}{RT}. \quad (16)$$

555 Finally, the saturated vapour pressure can be linked with temperature according to Kendall and
 556 Caldwell (1998)(7). Under the assumptions that osmotic suction is constant over time and space and
 557 the variation of the gas phase pressure is negligible compared to that of the liquid phase, changes in
 558 total and matric suction are equal and about opposite to changes in water pressure, $\Delta s = \Delta \psi \approx -\Delta u_l$.

559 Energy balance

560 The energy balance equation through porous medium can be expressed as (17):

$$\frac{\partial}{\partial t} (E_s \rho_s (1 - \phi) + E_l \rho_l S_l \phi + E_g \rho_g S_g \phi) + \nabla \cdot (\mathbf{i}_c + \mathbf{j}_{E_s} + \mathbf{j}_{E_l} + \mathbf{j}_{E_g}) = 0, \quad (17)$$

561 where $E_s = c_{p,s} T$, $E_l = c_{p,l} T$, $E_g = c_{p,v} T + L_w$ represent the energy stored at temperature T in the
 562 solid, in the liquid and in the gas phase, respectively, and they depends on the corresponding specific
 563 heat capacity ($c_{p,s}, c_{p,l}, c_{p,v}$) and on the latent heat of vaporization L_w . The specific heat capacity
 564 adopted for the cement bentonite solid (Vespo *et al.*, 2020), liquid and gaseous phase are respectively
 565 $c_{p,s} = 1107 \text{ J/(kg K)}$, $c_{p,w} = 4182 \text{ J/(kg K)}$, $c_{p,v} = 2062 \text{ J/(kg K)}$. The conduction of sensible heat is
 566 accounted for via the term \mathbf{i}_c , that can be linearly linked to the gradient of temperature via the Fourier
 567 law

$$\mathbf{i}_c = -\lambda \nabla T \quad (18)$$

568 where λ is the thermal conductivity of the medium, that was evaluated according to Cosenza *et al.*
 569 (2003) as a function of the different phase contributions: $\lambda = \left(\theta_s \sqrt{\lambda_s} + \theta_w \sqrt{\lambda_w} + \theta_v \sqrt{\lambda_v} \right)^2$, where
 570 $\lambda_s = 1.345 \text{ W/(m K)}$, $\lambda_w = 0.6 \text{ W/(m K)}$, $\lambda_v = 0.024 \text{ W/(m K)}$ are the thermal conductivity of the solid,
 571 water and vapour phase respectively. Energy flux related to the movement of the three phases was
 572 evaluated as follows:

$$\begin{aligned} \mathbf{j}_{E_s} &= 0 \\ \mathbf{j}_{E_l} &= E_l \mathbf{q}_l = c_{p,l} T \left[-\frac{\mathbf{K}_w^{sat} k_r}{g} \nabla (u_w + \rho_l g z) \right] \\ \mathbf{j}_{E_g} &= E_g \mathbf{J}_g^w = (c_{p,v} T + L_w) \left[-\phi (1 - S_l) D_v \tau \frac{c_g^w M_w}{\rho_l R T} \left(\nabla u_l - \frac{u_l}{T} \nabla T \right) \right]. \end{aligned} \quad (19)$$