



HAL
open science

Modelling the behaviour of bentonite pellet-powder mixtures upon hydration from dry granular state to saturated homogeneous state

Benjamin Darde, Patrick Dangla, Jean-Noël Roux, Jean-Michel Pereira, Jean Talandier, Minh Ngoc Vu, Anh Minh A.M. Tang

► To cite this version:

Benjamin Darde, Patrick Dangla, Jean-Noël Roux, Jean-Michel Pereira, Jean Talandier, et al.. Modelling the behaviour of bentonite pellet-powder mixtures upon hydration from dry granular state to saturated homogeneous state. *Engineering Geology*, 2020, 278, pp.105847. <10.1016/j.enggeo.2020.105847>. <hal-03053646>

HAL Id: hal-03053646

<https://enpc.hal.science/hal-03053646v1>

Submitted on 11 Dec 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



HAL Authorization

Title:

Modelling the behaviour of bentonite pellet-powder mixtures upon hydration from dry granular state to saturated homogeneous state

Authors:

Benjamin Darde^{1,2}: benjamin.darde@enpc.fr

Patrick Dangla¹: patrick.dangla@univ-eiffel.fr

Jean-Noël Roux¹: jean-noel.roux@univ-eiffel.fr

Jean-Michel Pereira¹: jean-michel.pereira@enpc.fr

Jean Talandier²: jean.talandier@andra.fr

Minh Ngoc Vu²: minh-ngoc.vu@andra.fr

Anh Minh Tang¹: anh-minh.tang@enpc.fr

Affiliations:

¹: Navier, Ecole des Ponts, Univ Gustave Eiffel, CNRS, Marne-la-Vallée, France

²: French national radioactive waste management agency (Andra), France

Corresponding author:

Anh Minh Tang

Ecole des Ponts ParisTech

6-8 avenue Blaise Pascal

Cité Descartes – Champs-sur-Marne

77455 Marne-la-Vallée cedex 2

anh-minh.tang@enpc.fr

Abstract:

Bentonite pellet-powder mixtures are candidate materials for sealing the galleries in deep geological repositories for radioactive waste. In the present work, swelling pressure tests are performed on pellet-powder mixtures with different powder contents. Results highlight the influence of the initial granular structure on the mechanical behaviour of pellet-powder mixtures. In mixtures with a low density powder in the inter-pellet porosity, the macroscopic response of pellet-powder mixtures is identical to that of a pellet assembly with no powder. A model is proposed to describe the hydromechanical behaviour of pellet-powder mixtures. The formulation considers two distinct states of the material, Granular and Continuous. In the Granular state, the pellets control the mechanical behaviour of the mixture. In the Continuous state, both pellets and powder contribute to the mechanical behaviour of the mixture. In the Granular domain, the material behaviour is described by constitutive laws proposed after a numerical study using Discrete Element Method. In the Continuous domain, the material behaviour is described by a modified Barcelona Basic Model. Transition between the two domains depends on the density of the powder phase and suction. The model is implemented in a Finite Element Method code, and the swelling pressure tests performed in the laboratory are simulated with a single set of parameters. These results improve the knowledge on the behaviour of bentonite pellet-powder mixtures during hydration in repository conditions.

Keywords:

Expansive soil, Pellets, Swelling pressure, Discrete Element Method, Finite Element Method, Constitutive modelling

List of notations:

(e)	Elastic (superscript)
(p)	Plastic (superscript)
$\dot{\epsilon}$	Strain rate of the periodic cell in DEM
a	Sphere diameter in DEM
a_0	Initial sphere diameter in DEM
C_A	Model parameter
dX	Increment of X
D	Pellet diameter
D_{cell}	Diameter of the isochoric cell
D_{sensor}	Diameter of the sensor
e	Total void ratio
e_0	Initial value of e
E_I	Pellet Young modulus
e_1	Pellet void ratio
e_{10}	Initial value of e_1
e_2	Powder grains void ratio
e_{20}	Initial value of e_2
e_M	Macrostructural void ratio
e_m	Microstructural void ratio
e_{m0}	Initial value of e_m
e_{M0}	Initial value of e_M
$f_{m a}$	Model parameter related to contact plasticity
$f_{m b}$	Model parameter related to contact plasticity
f_{Mm}	Model parameter
F_N	Normal contact force

F_{N0}	Initial value of F_N
$F_{N\alpha}$	α -coordinates of the contact force F_N
F_N^*	Mean contact normal force in the granular assembly
F_T	Tangential reaction vector
f_ε	Dimensionless stiffness parameter for the granular assembly
$f_{\varepsilon a}$	Value of f_ε if contacts are considered elastic
$f_{\varepsilon p}$	Value of f_ε if contacts are considered plastic
h	Height of the cylinder part of the pellet
H	Height of the pellet
H_{cell}	Height of the isochoric cell
I	Dimensionless inertia parameter
k_s	Model parameter
M	Slope of the critical state line
m_l	Mass of the pellet
m_g	Dimensionless pressure parameter
m_g^*	Limit value of m_g
N_l	Number of spheres in DEM
N_c	Number of contacts
n_p	Model parameter related to the initial elastic limit
p	Total mean stress
p^*	Elastic limit
p'	Effective mean stress
p'_0	Initial effective mean stress
p'_1	Effective mean stress in pellets
p'_2	Effective mean stress in powder grains
p_0	Initial value of p
p_{atm}	Atmospheric pressure

p_c	BBM reference stress
p_r	Reference stress
q	Deviatoric stress
\mathbf{r}	Vector joining the centre of particles in contact in DEM
r	Norm of the \mathbf{r} vector
r_0	Initial value r
R_I	Pellet strength
R_c	Curvature radius of the pellet
r_{vf}	Ratio of volume fraction
r_{vf}^*	Threshold value of r_{vf}
r_α	α -coordinates of vector \mathbf{r}
s	Total suction
s^*	Threshold suction
s_0	Initial suction
s_1	Suction in pellets
s_2	Suction in powder
s_M	Suction in macropores
\mathbf{u}	Displacement field
x_{plas}	Proportion of plastic contacts
x_s	Proportion of powder
Z	Coordination number
α_m	Model parameter related to microstructural stiffness
β_m	Model parameter related to microstructural stiffness
δ_N	Normal deflection
δ_N^*	Average normal deflection
ε_V	Total volumetric strain
$\varepsilon_{V I}$	Volumetric strain of the pellet

ε_{V2}	Volumetric strains of the powder grains
ε_{VM}	Volumetric strain of the macrostructure
κ	Dimensionless elastic stiffness parameter of the macrostructure related to changes in mean stress
κ_m	Dimensionless elastic stiffness parameter of the microstructure
κ_s	Dimensionless elastic stiffness parameter of the macrostructure related to changes in suction
λ	Dimensionless elastoplastic stiffness parameter of the macrostructure
λ_0	Value of λ in fully saturated conditions
ν_1	Poisson ratio of the pellet
ρ	Ratio $\lambda(s \rightarrow \infty)/\lambda_0$
ρ_d	Dry density of the mixture
ρ_{d10}	Initial pellet dry density
ρ_s	Density of solid particles
σ_{aa}	Diagonal components of the Cauchy stress tensor
σ_{xx0}	Initial xx components of the Cauchy stress tensor
σ_{yy0}	Initial yy components of the Cauchy stress tensor
σ_{zz0}	Initial zz components of the Cauchy stress tensor
Φ_1	Pellet volume fraction in experiments
ϕ_1	Pellet volume fraction in the model
Φ_{10}	Initial pellet volume fraction in experiments
ϕ_{10}	Initial pellet volume fraction in the model
Φ_2	Powder volume fraction in experiments
ϕ_2	Powder volume fraction in the model
Φ_{20}	Initial powder volume fraction in experiment
ϕ_M	Volume fraction of macrostructural voids
ϕ_{mat}	Volume fraction of the powder in inter-pellet voids

$\phi_{mat\ 0}$	Initial volume fraction of the powder in inter-pellet voids
ω	Model parameter related to the increase in λ with suction
Ω	Total volume of the granular assembly
Ω_0	Initial value of Ω
Ω_1	Volume of pellet
Ω_s	Total volume of solid in the mixture
$\Omega_{s\ 1}$	Volume of solid in the pellets
$\Omega_{s\ 2}$	Volume of solid in the powder grains
Ω_v	Total volume of voids in the mixture
$\Omega_{v\ 1}$	Volume of void in the pellets
$\Omega_{v\ 2}$	Volume of void in the powder grains
$\Omega_{v\ M}$	Volume of void between pellets and powder grains

Funding:

This work was funded by École des Ponts ParisTech and the French National Agency for Radioactive Waste Management (Andra).

1 **1. Introduction**

2 Bentonite materials are considered as sealing materials in concepts of deep geological disposal for
3 radioactive waste. These materials are characterised by a low permeability, good radionuclide
4 retention capacity and ability to swell upon hydration. The latter feature allows these materials to fill
5 technological voids and to exert a swelling pressure against the host rock, which confines the
6 excavation damaged zone and thus reduces the fluid flow along the gallery via this zone.

7 Owing to operational convenience, pellet-based materials have been considered as an alternative to
8 compacted blocks (Bernachy-barbe et al., 2020; Darde et al., 2018; Molinero-Guerra et al., 2017;
9 Navarro et al., 2020a, 2020b; Sánchez et al., 2016). The material is installed in the galleries in a dry
10 state as a granular assembly. Upon hydration by the pore water of the host rock, the granular material
11 progressively becomes homogeneous. Before homogenisation, the mechanical behaviour of the
12 material is controlled by its granular nature. In fully saturated conditions, the material has totally lost
13 its initial granular structure. Its structure in these conditions is similar to fully saturated compacted
14 blocks (Hoffmann et al., 2007; Imbert and Villar, 2006; van Geet et al., 2005). The behaviour of
15 pellet-based materials is thus controlled by different mechanisms depending on the hydration state.

16 In mixtures of pellet and crushed pellets (powder), powder fills inter-pellet voids, increasing the
17 total dry density of the sealing material thus its final swelling pressure (Imbert and Villar, 2006;
18 Kaufhold et al., 2015; Lloret et al., 2003; Wang et al., 2012). Considering these mixtures as sealing
19 materials involves challenges. For instance, powder in the inter-pellet porosity may migrate during
20 installation and induce local heterogeneities of density. In addition, depending on the density of the
21 powder phase, this latter can be considered to either participate in the macroscopic mechanical
22 response, or to leave the mechanical behaviour controlled by the pellet assembly.

23 Existing modelling frameworks do not account for these particular features of pellet-powder
24 mixtures. It is proposed in the present study to: *(i)* provide experimental evidence of the influence of
25 the granular structure on the macroscopic response of the mixture upon hydration; *(ii)* use Discrete
26 Element Method (DEM) to study the intrinsic behaviour of pellet assemblies; *(iii)* propose a

27 conceptual model, based on DEM results and modified existing modelling frameworks, able to
28 satisfactorily reproduce the main features of pellet-powder mixtures upon hydration.

29 In this respect, the present study is organised as follows. First, swelling pressure tests are carried
30 out in the laboratory on three pellet-powder mixtures, with identical pellet volume fraction and various
31 powder volume fractions, to highlight the influence of the granular structure on the material behaviour.
32 Experimental results are discussed and a conceptual interpretation of the behaviour of pellet mixtures
33 is proposed. Then, DEM simulations are performed to model large pellet assemblies and determine
34 constitutive laws. An elasto-plastic model, based on DEM results and a modified formulation of the
35 Barcelona Basic Model (BBM) (Alonso et al., 1990) is then proposed to describe the hydromechanical
36 behaviour of pellet-powder mixtures in granular and continuous states. The swelling pressure tests are
37 finally simulated using the proposed model.

38

39 2. Swelling pressure tests

40

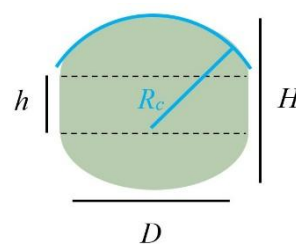
41 2.1. Tested materials

42 Pellets are made of MX80 bentonite. Pellets are composed of a cylinder-shaped part with two
43 spherical caps at both ends (Figure 1). Pellet geometrical properties and physical properties are
44 presented in Table 1.

45 a)

b)

46



47

47 Figure 1: a) Picture of a pellet; b) Schematic view of a pellet.

Table 1: Initial properties of the pellets.

Pellet properties	Value
<i>Geometrical properties</i>	
Diameter, D	7 mm
Height of the cylinder part, h	5 mm
Height, H	7 mm
Curvature radius, R_c	6.5 mm
<i>Physical properties</i>	
Density of solid particles, ρ_s (Saba et al., 2014)	2.77 Mg/m ³
Initial dry density, ρ_{d10}	1.91 Mg/m ³
Initial void ratio, e_{10}	0.45
Initial suction, s_0	89 MPa

49 The material referred to as “powder” is obtained by crushing pellets. Initial suction of the powder is
50 180 MPa. The average grain diameter of the powder is 0.65 mm (Molinero-Guerra et al., 2017).

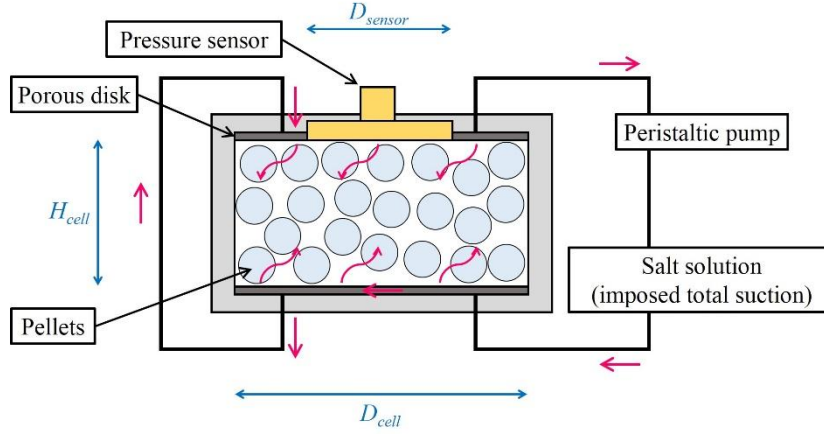
51 Three pellet-powder mixtures are prepared at the same pellet volume fraction and three different
52 powder volume fractions. Pellet initial volume fraction, Φ_{10} (volume of pellets / total volume), is
53 0.553 in all samples. The three powder initial volume fractions, Φ_{20} , are 0.236, 0.118, and 0. The
54 highest value of Φ_{20} corresponds to pellet/powder proportions of 70/30 in dry mass. To highlight that
55 Φ_{10} remains identical in all samples, the three mixtures are referred to as 70/30, 70/15, and 70/0 in the
56 following. The dry densities (ρ_d) of the 70/30, 70/15, and 70/0 materials are 1.50 Mg/m³, 1.275 Mg/m³,
57 and 1.05 Mg/m³, respectively.

58

59 2.2. Method

60 Four suction-controlled swelling pressure tests are carried out in isochoric cells. Cylindrical
61 isochoric cells made of stainless steel are used to perform the tests (see Figure 2). Swelling pressure is

62 determined as the vertical pressure measured by a sensor on the top wall of the cell. Two porous disks
 63 are placed at the top and bottom of the cell. The cell dimensions are: height, $H_{cell} = 30$ mm and
 64 diameter, $D_{cell} = 60$ mm. The sensor diameter is $D_{sensor} = 30$ mm.



65

66 Figure 2: Schematic view of the experimental setup. Red arrows represent humid air circulation.

67 Samples are prepared by placing pellets one by one in the isochoric cells to form a layer. Then, the
 68 proportion of powder associated to the laid proportion of pellets is poured in the inter-pellet voids.
 69 This procedure is followed to reduce heterogeneity of the samples (Molinero-Guerra et al., 2017).
 70 Four samples are prepared (Table 2).

71 Table 2: Properties of the pellet-powder mixtures.

Sample	Mixture	Φ_{10}	Φ_{20}	ρ_d : Mg/m ³
SP30	70/30		0.236	1.50
SP15	70/15	0.553	0.118	1.275
SP0a	70/0		0	1.05
SP0b	70/0		0	1.05

72

73 Hydration is performed step by step using the vapour equilibrium technique (Hoffmann et al.,
 74 2007; Lloret et al., 2003; Tang and Cui, 2005; Zhang et al., 2020) from s_0 to a suction $s = 4$ MPa. A
 75 peristaltic pump makes air circulate through the saturated salt solution to maintain a constant target
 76 relative humidity, then through the isochoric cell (Figure 2). Humid air is allowed to circulate directly

77 from the bottom to the top of the cell through a side tube. Thus, no excessive air pressure is developed
78 when using the peristaltic pump and humid air is considered to diffuse freely inside the inter-pellet
79 porosity. When the pressure measured by the sensor reaches a plateau, the subsequent suction step is
80 imposed. Following the suction step at $s = 4$ MPa, the sample is wetted using synthetic water of
81 similar chemical composition as the pore water of the host rock of Meuse/Haute-Marne Underground
82 Research Laboratory (France). Room temperature is controlled and remains constant ($20^{\circ}\text{C} \pm 1^{\circ}\text{C}$).
83 Hydration paths are summarised in Table 3. It is worth noting that hydration paths described as zero
84 suction are performed using synthetic water, which may have an osmotic suction higher than zero as
85 suggested by (Ferrari et al., 2014).

86 Table 3: Hydration paths followed by the samples. Suctions are given in MPa.

Sample	I	II	III	IV	V	VI	VII	VIII
SP0a	82	59	40	38	25	13	9	-
SP0b	82	59	40	25	9	4	0*	-
SP15	82	59	40	25	9	4	0*	-
SP30	82	59	40	25	13	9	4	0*

87 *: samples wetted using synthetic liquid water

88

89 2.3. Experimental results and discussion

90

91 2.3.1. Evolution of swelling pressure upon hydration

92 Figure 3 presents the evolution of swelling pressure as a function of elapsed time for all hydration
93 steps of the four swelling pressure tests (Table 3).

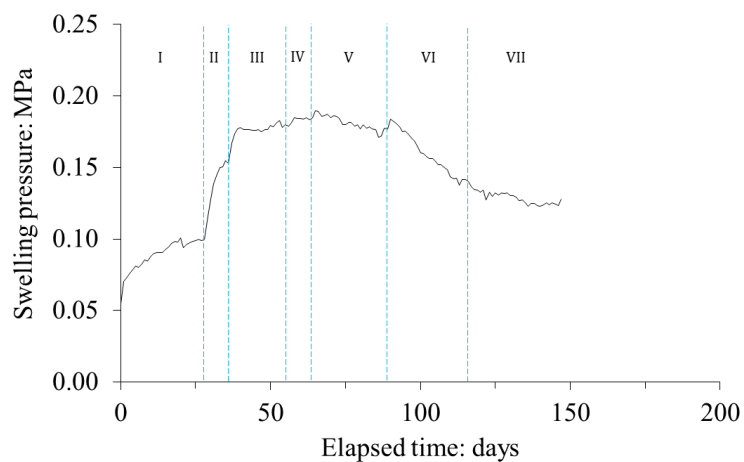
94 The swelling pressure in SP0a increases from 0.055 MPa (pressure following the closure of the
95 cell) to 0.173 MPa, from initial state to $s = 38$ MPa. Upon hydration from $s = 38$ MPa to $s = 9$ MPa,
96 the swelling pressure decreases to 0.128 MPa.

97 The swelling pressure in SP0b increases from 0.010 MPa to 0.153 MPa, from initial state to
98 $s = 9$ MPa. Upon hydration from $s = 9$ MPa to $s = 4$ MPa, the swelling pressure decreases to 0.135
99 MPa. Following liquid water flooding, the swelling pressure increases, reaches a peak at 0.250 MPa,
100 then stabilises at 0.247 MPa after 607 days.

101 The swelling pressure in SP15 increases from 0.015 MPa to 0.290 MPa from initial state to $s = 40$
102 MPa. Upon hydration from $s = 40$ MPa to $s = 4$ MPa, the swelling pressure decreases to 0.200 MPa.
103 Following liquid water flooding, the swelling pressure increases and stabilises at 0.910 MPa after 622
104 days.

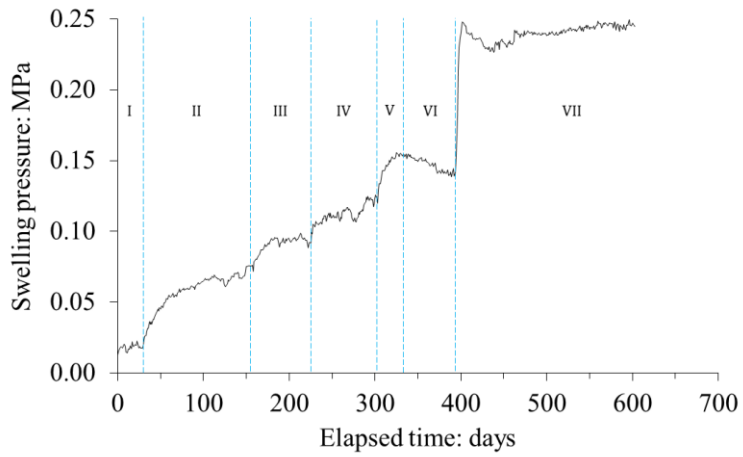
105 The swelling pressure in SP30 increases from 0.004 MPa to 1.22 MPa, from initial state to
106 $s = 9$ MPa. Upon hydration from $s = 9$ MPa to $s = 4$ MPa, the swelling pressure decreases to
107 0.650 MPa, followed by an increase to 1.36 MPa. Following liquid water flooding, the swelling
108 pressure increases and stabilises at 4.45 MPa after 755 days.

109 a)



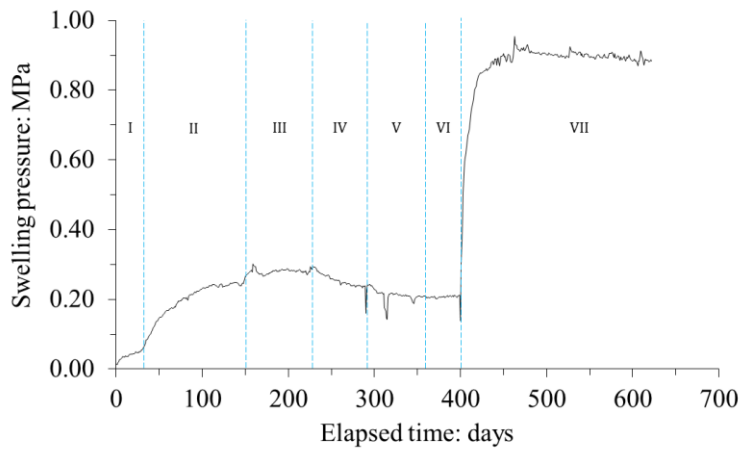
110

111 b)



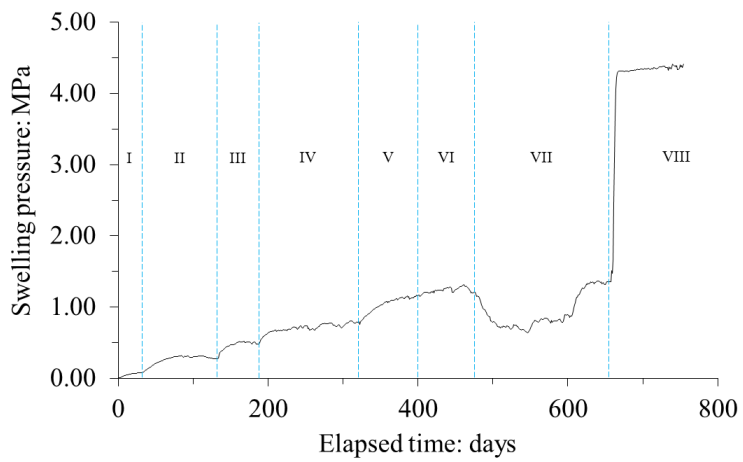
112

113 c)



114

115 d)



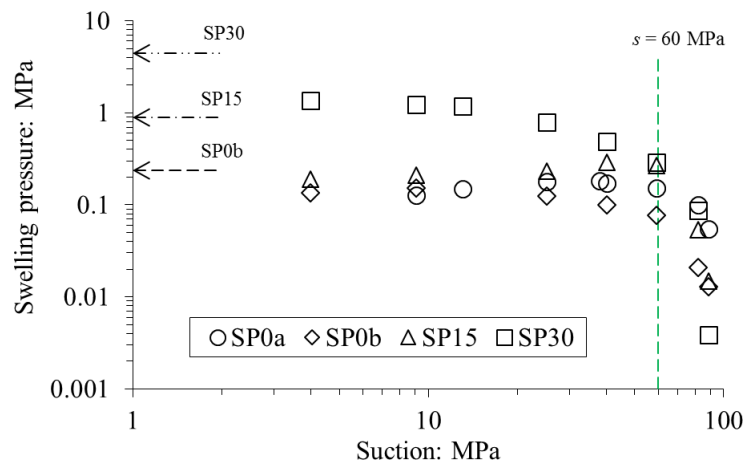
116

117 Figure 3: Evolution of swelling pressure as a function of elapsed time. a) SP0a test, b) SP0b test, c) SP15 test, d) SP30

118 test.

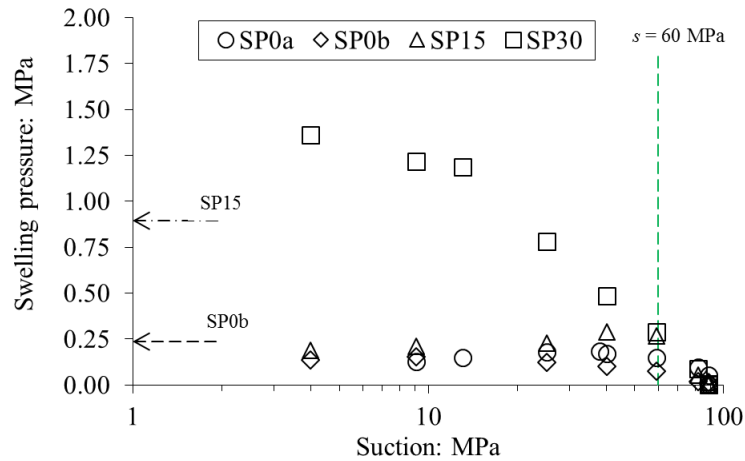
119 In Figure 4 the swelling pressure at the end of each step is plotted versus the imposed suction
 120 corresponding to the step. Because of the wide range of swelling pressure measured upon hydration,
 121 results of all tests are shown using a logarithmic scale in Figure 4a while a linear scale is used in
 122 Figure 4b, but limited to swelling pressures lower than 2 MPa. It is highlighted that during the first
 123 two hydration steps, the swelling pressure developed in 70/0 (SP0a and SP0b tests), 70/15 (SP15 test),
 124 and 70/30 (SP30 test) materials remains within the same order of magnitude. For $4 \text{ MPa} < s < 60 \text{ MPa}$,
 125 the swelling pressure developed in the 70/30 material is significantly higher than in the 70/15 and 70/0
 126 materials while those developed in 70/15 and 70/0 materials are still very close. Following water
 127 flooding, the final value of swelling pressure is higher for a higher dry density of the mixture, as
 128 previously observed in the literature (Bian et al., 2019; Imbert and Villar, 2006; Jia et al., 2019;
 129 Kaufhold et al., 2015; Lloret et al., 2003; Wang et al., 2013, 2012).

130 a)



131

132 b)



133

134 Figure 4: Evolution of swelling pressure as a function of suction upon hydration. a) Swelling pressure on a logarithmic
 135 scale, b) Swelling pressure on a linear scale, limited to swelling pressures lower than 2 MPa. Horizontal arrows represent the
 136 final swelling pressure of the tested materials.

137

138 2.3.2. Influence of the granular structure and the powder volume fraction

139 In SP0a and SP0b tests, no powder is filling the inter-pellet voids and the material behaviour is
 140 controlled by the granular assembly of pellets as long as the material is granular. In the course of
 141 hydration, pellet swelling in constant-volume conditions of the sample induces an increase of inter-
 142 granular forces and swelling pressure. As inter-granular forces reach pellet strength, swelling pressure
 143 no longer increases and a plateau/decrease of swelling pressure is observed as suction decreases. The
 144 influence of initial granular structure can be divided in two phases: (i) increase of inter-granular
 145 forces, controlled by pellet stiffness; (ii) plateau/decrease of swelling pressure controlled by pellet
 146 strength and stiffness decreasing upon wetting (Alonso et al., 2010; Darde et al., 2020b; Hoffmann et
 147 al., 2007).

148 Comparison of the suction-swelling pressure relationships of the three materials upon hydration
 149 suggests that the powder has a variable influence on the macroscopic response of the material. All
 150 samples have the same pellet solid fraction but different powder contents. Differences between results
 151 of the different tests is thus attributed to the presence of powder in the inter-pellet porosity.

152 For $s > 60$ MPa, the influence of powder is not significant. Then, for $4 \text{ MPa} < s < 60 \text{ MPa}$, the
153 response of the 70/30 material, characterised by a denser powder phase, displays a clear influence of
154 the powder in the inter-pellet porosity whereas that of the 70/15 material, characterised by a loose
155 powder phase, is not influenced by powder. These results suggest that powder can freely swell in the
156 inter-pellet porosity until it reaches a volume fraction allowing powder grains to participate in the
157 mechanical behaviour of the mixture.

158 In fully saturated state, an estimation of the final swelling pressure of MX80 bentonite-based
159 materials from their dry density is proposed by Wang et al. (Wang et al., 2012). For the three studied
160 materials, this estimation, obtained from empirical results on compacted powder samples, yields
161 swelling pressures of 4.44 MPa, 0.970 MPa, and 0.210 MPa for the 70/30, 70/15, and 70/0 materials.
162 The measured values are 4.45 MPa, 0.910 MPa, and 0.247 MPa, respectively. The close values
163 compared to experimental results obtained on pellet mixtures suggest that a high degree of
164 homogenisation is reached in the final state at the laboratory scale (Hoffmann et al., 2007; Imbert and
165 Villar, 2006; van Geet et al., 2005), which is confirmed by sample dismantling (Figure 5).

a)



b)



166 Figure 5: Pictures of SP0 samples. a) SP0a at 9 MPa of suction, b) SP0b following liquid water saturation. The cell
167 diameter is 60 mm.

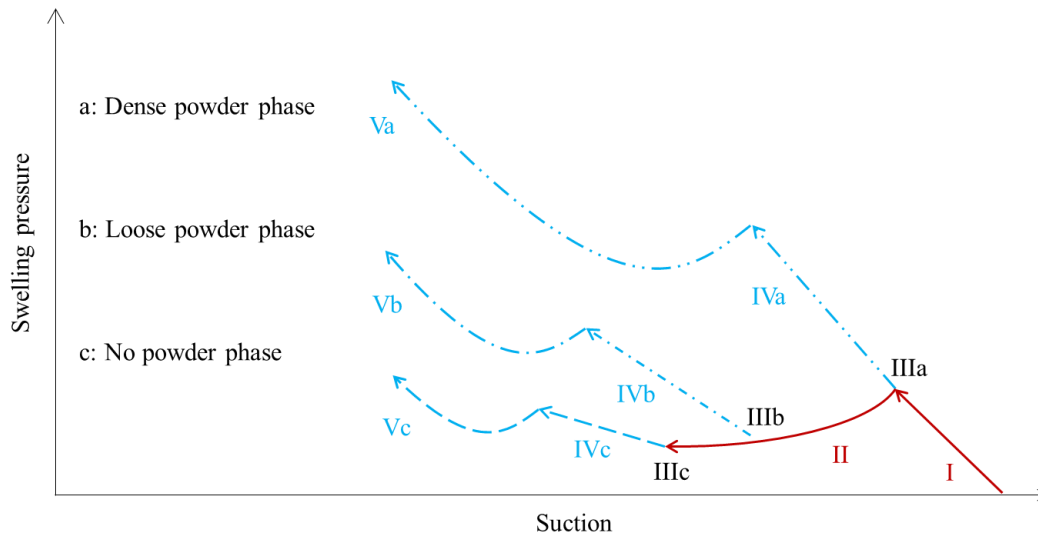
168 2.3.3. Conceptual interpretation of the material behaviour

169 Experimental results highlight that the material behaviour is controlled by the initial granular
170 structure until powder volume fraction Φ_2 reaches a maximum value. For the 70/30 material (with
171 $\Phi_{I0} = 0.553$), it occurs at $s \approx 60$ MPa. Besides, all materials, including the 70/0 material, appear to
172 have reached a high degree of homogenisation in the fully saturated state. In pellet assemblies, the
173 influence of initial granular structure is lost between $s = 4$ MPa and $s = 0$ MPa. In this suction range, it
174 has been evidenced that a significant reorganisation of the material fabric occurs (Saiyouri et al., 2004,
175 2000). Aggregates exfoliation in the macroporosity occurs, the microstructure and hydration
176 mechanisms change, and it is suggested that the initial granular structure no longer controls the
177 macroscopic behaviour. The key phenomena controlling the behaviour of pellet-powder materials are
178 thus: (i) the pellet stiffness; (ii) the pellet strength; (iii) a threshold value of Φ_2 ; (iv) a threshold value
179 of s ; and (v) the behaviour of the material when the influence of the initial granular structure is lost.

180 A conceptual diagram describing the understanding of the material behaviour in the light of
181 experimental results is presented in Figure 6. In this Figure, a wetting phase (decrease of suction) is
182 represented for three types of pellet-powder mixtures (no powder, loose powder phase, dense powder
183 phase).

184 Upon suction decrease, the swelling pressure evolution is first controlled by the assembly of pellets
185 only (Phases I and II in Figure 6). During this phase, the material behaves as a granular assembly.
186 Then, a “granular-continuous” transition is observed: the material response is not totally controlled by
187 the granular structure anymore but still is not that of a continuous bentonite material (Phase III).
188 Finally, the initial granular structure has lost its influence on the macroscopic response (Phases IV and
189 V). During this “continuous phase”, the macroscopic response is controlled by the hydration of the
190 bentonite regardless of the initial granular nature of the material.

191 Peak phenomena can be observed in the granular phase, as a consequence of the contact force
192 reaching pellet strength, or in the continuous phase, as a result of the collapse of the macrostructure of
193 the bentonite following microstructural rearrangement (Lloret et al., 2003).



194

Granular domain	I	Increase of contact forces
	II	Contact forces reach pellet strength
Transition	III	Loss of influence of the initial granular structure
Continuous domain	IV	Microstructural rearrangements and subsequent collapse of the macrostructure of the bentonite
	V	Increase of swelling pressure to the final value

195

196 Figure 6: Conceptual interpretation of the behaviour of pellet-powder materials upon hydration in constant-volume
 197 conditions. The red solid lines represent the influence of the initial granular structure. The blue dashed lines represent the
 198 behaviour after the transition to the continuous domain.

199

200 3. Insight into the behaviour of the granular material using DEM

201

202 3.1. Objectives

203 Existing modelling framework used to describe the behaviour of compacted bentonite materials do
 204 not account for the features described in the previous section. Since the pellet assembly can control the
 205 mechanical response of the material if Φ_2 is below a threshold value, it is proposed in the present
 206 section to study the mechanical behaviour of pellet assemblies with no powder.

207 In this respect, DEM is used to address the mechanical behaviour of pellet assemblies from the
 208 mechanical properties of the pellet (Darde et al., 2018).

209

210 3.2. Method

211 DEM is used to perform isotropic compression on large pellet assemblies unaffected by cell walls
212 to address the intrinsic behaviour of the material. Each pellet is modelled by a spherical particle. The
213 behaviour of each particle, interaction at contacts and details concerning the simulation procedure are
214 given in the following sections.

215

216 3.2.1. Model for a pellet at high suctions

217 (Darde et al., 2018) proposed, from experimental characterisation, the following expressions to
218 describe the evolution of the pellet Young modulus, E_1 , volumetric strain, ε_{V1} , and compressive
219 strength, R_1 :

$$E_1 = 3(1 - 2\nu_1) \frac{1}{\beta_m} \exp(\alpha_m p') \quad (1)$$

$$\varepsilon_{V1} = \int_{p'_0}^{p'} \beta_m \exp(-\alpha_m p') dp' \quad (2)$$

$$R_1 = C_A E_1 = 3(1 - 2\nu_1) \frac{C_A}{\beta_m} \exp(\alpha_m p') \quad (3)$$

$$p' = p + s \quad (4)$$

220 where ν_1 is the pellet Poisson ratio, p' is the effective mean stress, p'_0 is the initial effective mean
221 stress, C_A , α_m and β_m are model parameters, p is the total mean stress and s the total suction.

222 The model is based on the following assumptions: the pellet is considered saturated, its behaviour
 223 is elastic and controlled by the behaviour of the aggregates. Further information and discussions
 224 concerning the model can be found in (Darde et al., 2018). The values of the model parameters are
 225 presented in Table 4.

226 Table 4: Parameters of the model for a pellet.

Parameter	Value
α_m	0.024 MPa ⁻¹
β_m	0.016 MPa ⁻¹
ν_1	0.3
C_A	1.206 x10 ⁻⁷ m ⁻²
s_0	89 MPa

227

228 3.2.2. Contact laws

229 In DEM simulations, each pellet is modelled as a sphere. Interactions at contact between spheres
 230 are described by (i) the Hertz law (equation (5)) for normal force with perfect plasticity as the normal
 231 force F_N reaches the pellet strength R_f (Darde et al., 2020a) and (ii) a simplified form of the Cattaneo-
 232 Mindlin-Deresiewicz law (Johnson, 1985) for tangential elasticity with sliding as the tangential force
 233 reaches the Coulomb condition (Agnolin and Roux, 2007). Damping in contacts is considered as in
 234 (Agnolin and Roux, 2007).

235 For two spheres of same diameter a in contact, the Hertz law is expressed as follows:

$$F_N = \frac{1}{3} \frac{E_1}{1 - \nu_1^2} a^{\frac{1}{2}} \delta_N^{\frac{3}{2}} \quad (5)$$

236 Where δ_N is the normal deflection.

237

238 3.2.3. Study of the behaviour of large pellet assemblies under isotropic compression

239 (Darde et al., 2020a) highlighted that wall effects in the laboratory affect the material behaviour, as
240 suggested by (Bernachy-barbe et al., 2020) in an experimental study. Thus, it is proposed to model
241 large periodic pellet assemblies (with no wall) to address the intrinsic behaviour of these materials.
242 The main objective is to determine constitutive laws describing the macroscopic response of pellet
243 assemblies under variations of mean stress and suction. The macroscopic response of the material
244 depends on pellet stiffness, pellet strength, pellet diameter (varying upon pellet swelling/shrinkage),
245 deflection in contacts. All of them depend on both mean stress and suction. It is proposed to study: (i)
246 the behaviour of the granular assembly under isotropic compression at constant sphere diameter; and
247 (ii) the behaviour of the granular assembly under particle swelling at constant mean stress, pellet
248 stiffness and pellet strength.

249 The behaviour of the granular assembly under isotropic compression at constant sphere diameter is
250 studied using DEM. The behaviour of the granular assembly under particle swelling at constant mean
251 stress, pellet stiffness and pellet strength is addressed through a theoretical approach, discussed in a
252 forthcoming section.

253 Two dimensionless control parameters are used in DEM simulations. A dimensionless pressure
254 parameter, m_g , is used to describe the compression state of the granular assembly. m_g is defined as:

$$m_g = \left(\frac{p}{\frac{E_1}{1 - \nu_1^2}} \right)^{\frac{2}{3}} \quad (6)$$

255 Where p is the mean stress in the granular assembly. Details concerning the expression of m_g can
256 be found in Appendix A.

257 m_g is a convenient parameter to study the behaviour of the pellet assembly, since a single term
258 depending on both mean stress and pellet stiffness can account for features such as deflection in
259 contacts, creation/loss of contacts between pellets, evolution of the total volume (see Appendix A).

260 The strain rate of the periodic cell, $\dot{\epsilon}$, is controlled by a dimensionless inertia parameter, I , defined
 261 as (Agnolin and Roux, 2007):

$$I = \dot{\epsilon} \left(\frac{m_1}{a p} \right)^{\frac{1}{2}} \quad (7)$$

262 where m_1 is the mass of the pellet.

263 A constant value of I is imposed to apply a constant strain rate of the cell. In the present work, I is
 264 set to 10^{-5} to model an isotropic compression at constant slow rate. The evolution of m_g is determined
 265 from equation (6) during compression.

266 The simulations are performed on different granular assemblies, prepared at different values of Φ_{10}
 267 and initial coordination number Z . All numerical samples are composed of 4000 spheres, in a cubic
 268 cell with periodic boundary conditions (pellets located at one cell boundary are in contact with pellets
 269 located at the opposite boundary), thus no pellet-wall interaction exists (Agnolin and Roux, 2007).
 270 Five types of granular assemblies are prepared and denoted A, B, C, D, and E. For each type of
 271 granular assembly, three samples are prepared to assess the calculation repeatability. A, B, C, and E
 272 samples are prepared under $m_g = 1.19 \times 10^{-4}$; D samples are prepared under $m_g = 1.03 \times 10^{-4}$. Samples
 273 preparation procedure is described in details in (Agnolin and Roux, 2007). Table 5 summarises the
 274 main initial properties of the samples.

275 Table 5: Initial properties of the numerical samples. All samples are cubic and contain 4000 spheres.

Sample		Z	Φ_{10}
A	1	6.0	0.638
	2	6.1	0.639
	3	6.0	0.636
B	1	4.4	0.636
	2	4.2	0.634

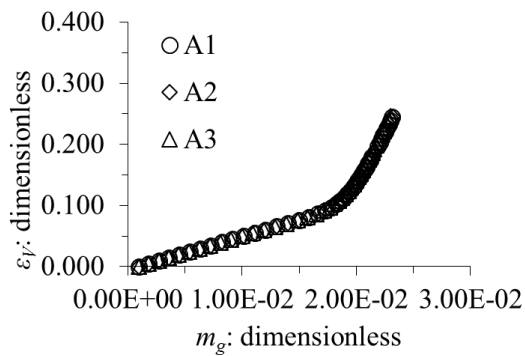
		3	4.3	0.635
	C	1	4.4	0.595
		2	4.3	0.593
		3	4.4	0.594
	D	1	4.5	0.577
		2	4.4	0.575
		3	4.4	0.578
	E	1	5.8	0.628
		2	5.8	0.627
		3	5.8	0.627

276

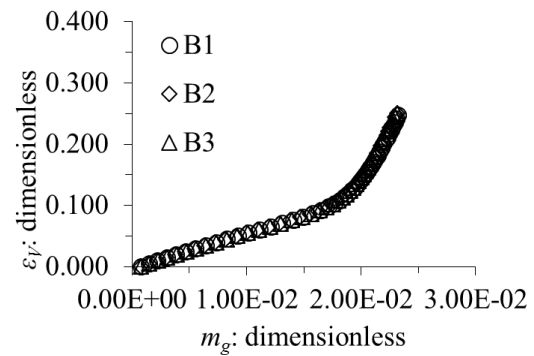
277 3.3. DEM simulation results and discussion

278 The evolution of volumetric strain, denoted by ε_V , during isotropic compression is presented in
 279 Figure 7 as a function of m_g , for all the granular assemblies. Compressive stress are positive and
 280 positive volumetric strains are associated to volume decrease in the following.

a)

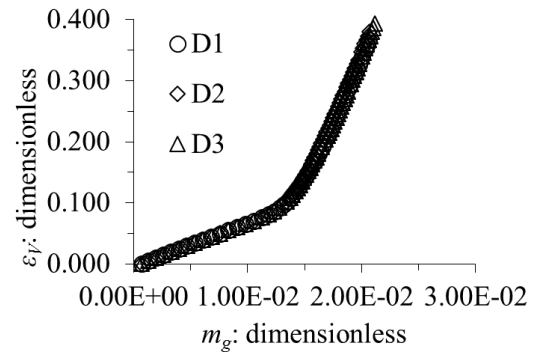
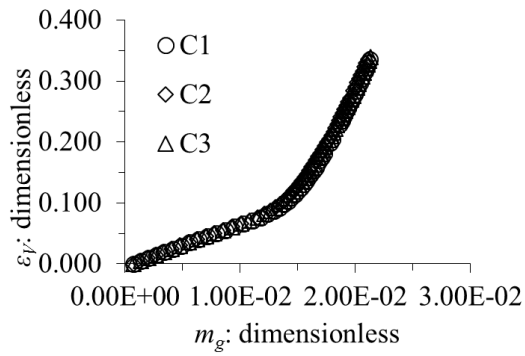


b)

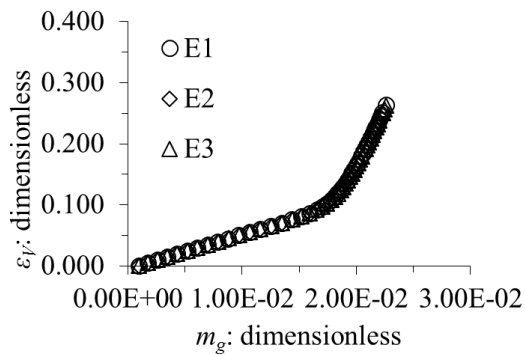


c)

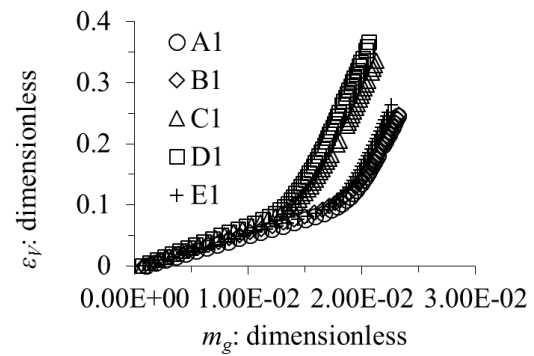
d)



e)



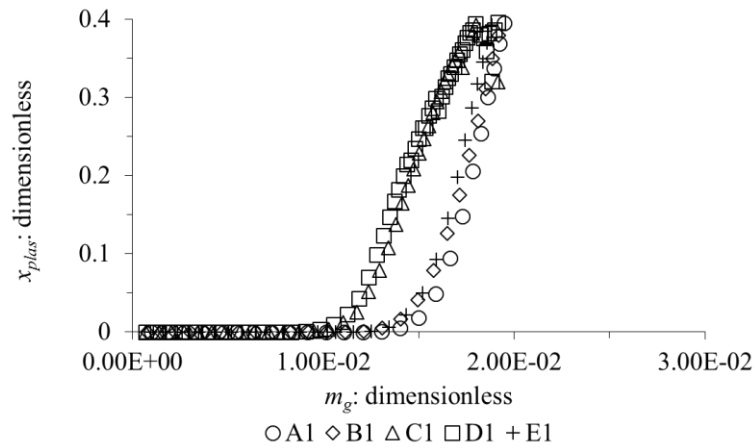
f)



281

282 Figure 7: Evolution of ε_V as a function of m_g during isotropic compression for all granular assemblies. a) A samples, b) B
 283 samples, c) C samples, d) D samples, e) E samples, f) comparison between A1, B1, C1, D1, E1.

284 All granular assemblies display a comparable response during isotropic compression. The relation
 285 between ε_V and m_g can be represented by a piecewise linear function with two pieces. As highlighted
 286 in Figure 7f, the slopes $\Delta\varepsilon_V/\Delta m_g$ do not vary significantly from one sample to another. The threshold
 287 value of m_g varies depending on the sample types, however.



288

289

Figure 8: Comparison of the evolution of x_{plas} as a function of m_g for the five types of granular assemblies.

290

291

292

293

294

From the definition of m_g (equation (6)) and results presented in Figure 7, the influence of pellet stiffness and mean stress is easily determined. Increase in p at constant E_I causes the total volume to decrease. Decrease in E_I at constant p causes the total volume to decrease. For E_I increasing with increasing p (equations (1) and (4)), evolution of p can induce competing effects on the total volumetric strain, which highlights the relevance of m_g to describe the material behaviour.

295

296

297

298

299

300

301

302

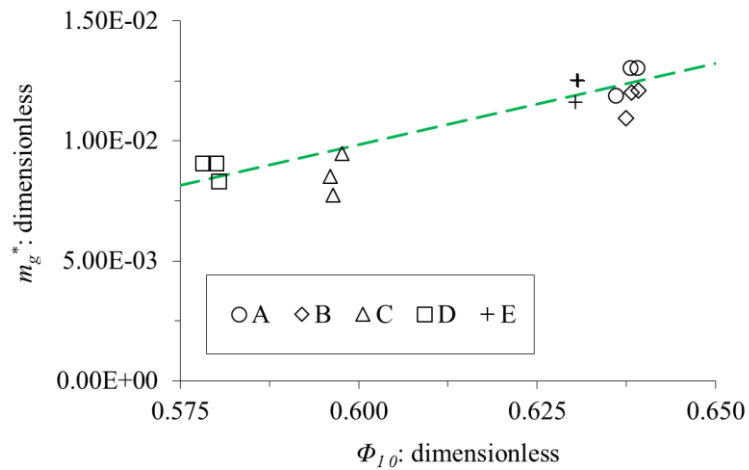
The pellet strength is accounted for by introducing perfect plasticity in the contact law. Influence of the pellet strength is thus highlighted by comparison of the evolutions of ε_V and the proportion of plastic contacts, x_{plas} , as functions of m_g during isotropic compression (Figure 7 and Figure 8). In all samples, all contacts remain elastic until a threshold value of m_g is reached. Contact plasticity then increases continuously. Interestingly, this threshold value is very close to that corresponding to the slope change in the piecewise linear evolution of $\varepsilon_V(m_g)$. This limit value of m_g is denoted by m_g^* . The slope change in the ε_V - m_g relationship is interpreted as a consequence of contact forces reaching the pellet strength.

303

304

305

From Figure 7, it is highlighted that Φ_{10} has no significant influence on the values of $\Delta\varepsilon_V/\Delta m_g$. Figure 8 however highlights that m_g^* is higher for higher values of Φ_{10} . The relationship between these two values is plotted in Figure 9 for all samples.



306

307 Figure 9: Relationship between m_g^* and Φ_{10} for all samples. The dashed line represents the fit corresponding to

308 $m_g^* = 0.067 \Phi_{10} - 0.031$.

309

310 4. Proposition of a macroscopic model for pellet-powder mixtures

311

312 4.1. Conceptual approach

313 This section presents a conceptual model able to describe the hydromechanical behaviour of pellet-
 314 powder mixtures, accounting for relevant features such as a free swelling phase for the powder,
 315 influence of granular structure, influence of pellet strength, transition to a continuous state. The
 316 approach is based on the scheme presented in Figure 6.

317 Following this approach, the material is considered to be either “granular” or “continuous”. In the
 318 granular domain, the mechanical behaviour is controlled by the pellet assembly. Constitutive laws are
 319 proposed from DEM results. In the continuous domain, the material behaves as a continuum. Its
 320 mechanical behaviour is described by a modified Barcelona Basic Model (BBM, (Alonso et al.,
 321 1990)). Transition from granular domain to continuous domain is related to a ratio of powder volume
 322 fraction to pellet volume fraction and to suction.

323 Three different pore types are considered: the pellet void ratio is denoted by e_1 ; the powder grains
 324 void ratio is denoted by e_2 ; and the total void ratio is denoted by e :

$$e_1 = \frac{\Omega_{v1}}{\Omega_{s1}} \quad (8)$$

$$e_2 = \frac{\Omega_{v2}}{\Omega_{s2}} \quad (9)$$

$$e = \frac{\Omega_v}{\Omega_s} \quad (10)$$

325 where Ω_{s1} and Ω_{s2} are the volume of solid in the pellets and powder grains, and $\Omega_s = \Omega_{s1} + \Omega_{s2}$ is the
 326 total volume of solid in the mixture. Ω_{v1} and Ω_{v2} are the volume of void in the pellets and powder
 327 grains (microstructural pores), and Ω_v is the total volume of voids in the mixture. Denoting by Ω_{vM} the
 328 volume of void between pellets and powder grains (macrostructural pores, $\Omega_{vM} = \Omega_v - \Omega_{v1} - \Omega_{v2}$), the
 329 macrostructural void ratio, e_M , is defined as:

$$e_M = \frac{\Omega_{vM}}{\Omega_s} \quad (11)$$

330 The microstructural void ratio, e_m , is then defined as:

$$e_m = \frac{\Omega_{s1}}{\Omega_s} e_1 + \frac{\Omega_{s2}}{\Omega_s} e_2 \quad (12)$$

331 The solid phase is considered incompressible. In all the formulation, volumetric strains are defined
 332 relative to the initial volume. Volumetric strains associated to each levels of structure are defined as
 333 follows:

$$\varepsilon_{v1} = -\frac{\Delta e_1}{1 + e_{10}} \quad (13)$$

$$\varepsilon_{V2} = -\frac{\Delta e_2}{1 + e_{20}} \quad (14)$$

$$\varepsilon_V = -\frac{\Delta e}{1 + e_0} \quad (15)$$

334 where ε_{V1} and ε_{V2} are the volumetric strains of the pellets and of the powder grains, respectively. The
 335 initial value of e , e_1 , e_2 , are denoted by e_0 , e_{10} , e_{20} , respectively.

336 Volume fractions of the different levels of structure are defined as follows:

$$\phi_1 = \frac{\Omega_{v1} + \Omega_{s1}}{\Omega_v + \Omega_s} \quad (16)$$

$$\phi_2 = \frac{\Omega_{v2} + \Omega_{s2}}{\Omega_v + \Omega_s} \quad (17)$$

$$\phi_M = 1 - \phi_1 - \phi_2 \quad (18)$$

337 where ϕ_1 , ϕ_2 , ϕ_M are the volume fractions of pellets, powder, and macrostructural voids. For
 338 convenience, an additional volume fraction, ϕ_{mat} , is considered. ϕ_{mat} is the volume fraction of the
 339 granular assembly composed of the powder grains within inter-pellet voids:

$$\phi_{mat} = \frac{\Omega_{v2} + \Omega_{s2}}{\Omega - (\Omega_{v1} + \Omega_{s1})} = \frac{\phi_2}{1 - \phi_1} \quad (19)$$

340 Suction in pellets is denoted by s_1 , suction in powder is denoted by s_2 , suction in macropores is
 341 denoted by s_M . In the conceptual approach presented in this study, hydraulic equilibrium is considered:

$$s_1 = s_2 = s_M = s \quad (20)$$

342 The macroscopic mean stress p is parted into two terms affecting the pellet and powder phases as
 343 follows:

$$p = \sum_i \phi_i p_i \quad (21)$$

344 And the effective mean stress in each phase i , p'_i is defined as follows:

$$p'_i = s_i + p_i \quad (22)$$

345 where ϕ_i , p_i and s_i are the volume fraction of phase i , mean stress in phase i and suction in phase i ,
 346 respectively.

347 In the granular domain, it is assumed that the pellet controls the mechanical behaviour of the
 348 material, as observed in swelling pressure tests. Only pellets are affected by the macroscopic mean
 349 stress. Thus, effective mean stresses in the granular domain are defined as follows:

$$p'_1 = s + \frac{p}{\phi_1} \quad (23)$$

$$p'_2 = s \quad (24)$$

350 In the continuous domain, it is assumed that both pellets and powder contribute to the mechanical
 351 behaviour of the material. Effective mean stresses in the continuous domain are defined as follows:

$$p'_1 = s + \frac{p}{\phi_1 + \phi_2} \quad (25)$$

$$p'_2 = s + \frac{p}{\phi_1 + \phi_2} \quad (26)$$

352 Transition from granular to continuous domain occurs as the pellet assembly no longer controls the
 353 mechanical behaviour of the pellet-powder mixture. From experimental results, it is proposed that this
 354 phenomenon can be related to the density of the granular assembly composed of the powder grains
 355 within inter-pellet voids. The following ratio r_{vf} is proposed to describe the density of the powder
 356 phase:

$$r_{vf} = \frac{\phi_{mat}}{\phi_1} \quad (27)$$

357 A threshold value of r_{vf} , denoted by r_{vf}^* , has to be determined. The material is considered in the
 358 continuous domain if the following condition is verified:

$$r_{vf} \geq r_{vf}^* \quad (28)$$

359 It was discussed in (Darde et al., 2020b, 2018) that the model used to study the behaviour of a
 360 pellet assembly using DEM was no longer relevant at low suction. Indeed, bentonite materials undergo
 361 significant microstructural changes at low suction that have an influence on the macroscopic response
 362 under hydromechanical loadings (Molinero-Guerra et al., 2020; Saiyouri et al., 2004; Villar, 2007).
 363 For instance, the significant increase of swelling pressure observed at suction lower than 4 MPa in the
 364 swelling pressure tests, regardless of the powder content, (Figure 3) cannot be reproduced using the
 365 DEM model. These phenomena are not related to the initial granular structure. Thus, in the proposed
 366 conceptual approach, a threshold value of suction for the domain transition is proposed.

367 The threshold suction, denoted by s^* , is also a material parameter. The material is considered in the
368 continuous domain if the following condition is verified:

$$s \leq s^* \quad (29)$$

369 The transition is related to the loss of the initial structure and is considered an irreversible
370 phenomenon.

371

372 In both granular and continuous domains, the volumetric strain of the microstructure (pellets and
373 powder grains) is considered to remain elastic. Plasticity only affects the macrostructure. In the
374 granular domain, DEM results highlighted that local plasticity in contacts can affect the material
375 behaviour. In the proposed approach, this phenomenon is addressed by considering different elastic
376 stiffness parameters depending on the value of m_g . These points are presented in the following section.

377

378 **4.2. Elastic volumetric strains**

379 In the granular domain, the increment of elastic (superscript (e)) volumetric strains for the
380 microstructure (pellets and powder) grains are written as follows:

$$d\varepsilon_{V1}^{(e)} = d\varepsilon_{V1} = \beta_m \exp(-\alpha_m p'_1) dp'_1 \quad (30)$$

381 and

$$d\varepsilon_{V2}^{(e)} = d\varepsilon_{V2} = \beta_m \exp(-\alpha_m p'_2) dp'_2 \quad (31)$$

382 As previously discussed, in the granular domain the behaviour of the granular assembly is
 383 considered to be dependent on variations of m_g (equation (15)) at constant pellet volume and variations
 384 of pellet volume at constant m_g . Elastic total volumetric strain is thus a function of m_g and ε_{V1} :

$$d\varepsilon_V^{(e)} = \frac{\partial \varepsilon_V^{(e)}}{\partial m_g} dm_g + \frac{\partial \varepsilon_V^{(e)}}{\partial \varepsilon_{V1}} d\varepsilon_{V1} \quad (32)$$

385 Denoting $\frac{\partial \varepsilon_V^{(e)}}{\partial m_g}$ by f_ε and $\frac{\partial \varepsilon_V^{(e)}}{\partial \varepsilon_{V1}}$ by f_{Mm} , equation (32) is written as follows:

$$d\varepsilon_V^{(e)} = f_\varepsilon dm_g + f_{Mm} d\varepsilon_{V1} \quad (33)$$

386 where f_ε is the slope of the piecewise linear function as discussed in the DEM study:

$$\begin{cases} f_\varepsilon = f_{\varepsilon a} ; m_g \leq m_g^* \\ f_\varepsilon = f_{\varepsilon b} ; m_g > m_g^* \end{cases} \quad (34)$$

387 in which $f_{\varepsilon a}$ and $f_{\varepsilon b}$ are parameters, with $f_{\varepsilon a} < f_{\varepsilon b}$. m_g^* is a function of the initial volume fraction of
 388 pellets ϕ_{10} (Figure 9):

$$m_g^* = f_{m a} \phi_{10} + f_{m b} \quad (35)$$

389 with $f_{m a}$ and $f_{m b}$ two model parameters. Equations (34) and (35) allow to account for contact plasticity
 390 in the model while still considering the stress state to be elastic.

391 f_{Mm} is obtained from equations (5) and (56) (Appendix A). For elastic contacts, its value is 1 in a
 392 granular assembly of spherical particles. See details in Appendix B.

393 Constitutive models for unsaturated soils generally consider two independent stress variables. In
 394 the present work, this approach is adopted and the two stress variables used to describe the volumetric

395 behaviour are the mean stress p and suction s . In this respect, an alternative expression for equation
 396 (33) is:

$$d\varepsilon_V^{(e)} = \left(f_\varepsilon \frac{\partial m_g}{\partial p} + f_{Mm} \frac{\partial \varepsilon_{V1}}{\partial p} \right) dp + \left(f_\varepsilon \frac{\partial m_g}{\partial s} + f_{Mm} \frac{\partial \varepsilon_{V1}}{\partial s} \right) ds \quad (36)$$

397

398 with

$$\frac{\partial m_g}{\partial p} = \frac{2}{3} \left[\frac{1}{p} - \frac{\alpha_m}{\phi_1} \right] \left(\frac{\beta_m}{3} \frac{1 - \nu^2}{1 - 2\nu} \right)^{\frac{2}{3}} p^{\frac{2}{3}} \exp\left(-\frac{2}{3} \alpha_m p'_1\right) \quad (37)$$

$$\frac{\partial m_g}{\partial s} = -\frac{2}{3} \alpha_m \left(\frac{\beta_m}{3} \frac{1 - \nu^2}{1 - 2\nu} \right)^{\frac{2}{3}} p^{\frac{2}{3}} \exp\left(-\frac{2}{3} \alpha_m p'_1\right) \quad (38)$$

$$\frac{\partial \varepsilon_{V1}}{\partial p} = \frac{\beta_m}{\phi_1} \exp(-\alpha_m p'_1) \quad (39)$$

$$\frac{\partial \varepsilon_{V1}}{\partial s} = \beta_m \exp(-\alpha_m p'_1) \quad (40)$$

399

400 In the continuous domain, the BBM (Alonso et al., 1990) is modified to account for three distinct
 401 porosities. Increment of elastic volumetric strains are written as follows:

$$d\varepsilon_{V1}^{(e)} = d\varepsilon_{V1} = \frac{\kappa_m}{1 + e_{10}} \frac{dp'_1}{p'_1} \quad (41)$$

402 and

$$d\varepsilon_{V2}^{(e)} = d\varepsilon_{V2} = \frac{\kappa_m}{1 + e_{20}} \frac{dp_2'}{p_2'} \quad (42)$$

403 where κ_m is a dimensionless stiffness parameter.

404 Elastic total volumetric strain is a function of the volumetric strains of each level of structure:

$$d\varepsilon_V^{(e)} = \frac{1}{1 + e_0} \left[(1 + e_{M0}) d\varepsilon_{VM}^{(e)} + (1 - x_s)(1 + e_{10}) d\varepsilon_{V1} + x_s (1 + e_{20}) d\varepsilon_{V2} \right] \quad (43)$$

405 where e_{M0} is the initial value of e_M , $\varepsilon_{VM}^{(e)}$ is the elastic volumetric strain of the macrostructure
 406 (macropores between pellets and powder grains) and x_s is defined as:

$$x_s = \frac{\Omega_s}{\Omega_s} \quad (44)$$

407 The elastic increment of macrostructure volumetric strain is defined as follows:

$$d\varepsilon_{VM}^{(e)} = \frac{1}{1 + e_{M0}} \left(\kappa \frac{dp}{p} + \kappa_s \frac{ds}{s + p_{atm}} \right) \quad (45)$$

408 where κ and κ_s are dimensionless stiffness parameters related to variations of p and s , respectively, and
 409 p_{atm} is the atmospheric pressure.

410 The dependency of the macrostructural swelling potential on the material density is accounted for
 411 by assuming a linear relationship between κ_s and κ_m :

$$\kappa_s = \frac{e_{m0}}{e_0} \kappa_m \quad (46)$$

412 where e_{m0} is the initial value of e_m .

413

414 **4.3. Plasticity and hardening**

415 A single plastic flow rule is used for the granular and continuous domains. Plasticity and hardening
416 are addressed using the concept of the Loading-Collapse curve (Alonso et al., 1990). The elastic limit
417 p^* evolves as a function of suction as:

$$p^*(s) = p_c \left(\frac{p^*(0)}{p_c} \right)^{\frac{\lambda(0)-\kappa}{\lambda(s)-\kappa}} \quad (47)$$

418 where p_c is the BBM reference stress (Alonso et al., 1990) and $\lambda(s)$ is a dimensionless elastoplastic
419 stiffness parameter, function of s . The hardening law is written as follows:

$$\frac{dp^*(0)}{p^*(0)} = \frac{1 + e_0}{\lambda(0) - \kappa} d\varepsilon_v^{(p)} \quad (48)$$

420 where the superscript (p) denotes the plastic part of the volumetric strain.

421 The evolution of the dimensionless elastoplastic stiffness parameter λ as a function of suction is
422 described as follows:

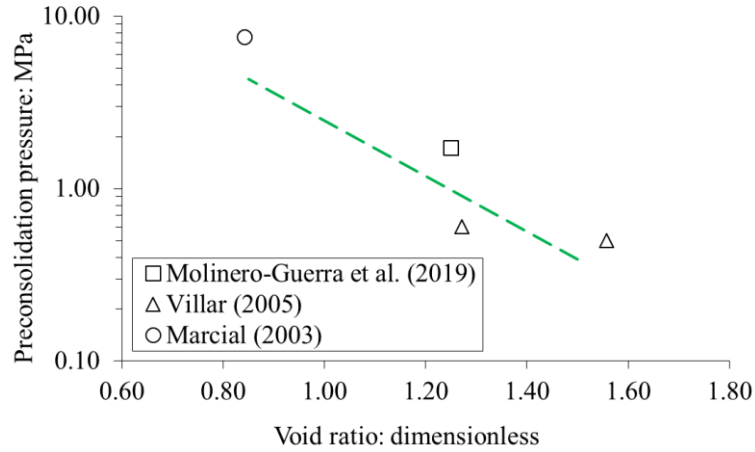
$$\lambda(s) = \lambda_0 [\rho + (1 - \rho) \exp(-\omega s)] \quad (49)$$

423 where λ_0 , ρ , and ω are material parameters respectively related to the saturated elastoplastic stiffness,
424 ratio of $\lambda(s \rightarrow \infty)$ to $\lambda(0)$, and evolution of stiffness with suction.

425 From oedometer tests carried out on saturated MX80 bentonite materials by (Marcial, 2003;
426 Molinero-Guerra et al., 2019a; Villar, 2005), an empirical relationship between the initial value of
427 $p^*(0)$ and e_0 is proposed (Figure 10):

$$p^*(0) = p_r \exp(-n_p e_0) \quad (50)$$

428 where p_r is a reference stress and n_p a material parameter.



429

430 Figure 10: Preconsolidation pressure as a function of the void ratio in oedometer tests performed on bentonite materials
 431 by Molinero-Guerra et al. (2019) (Molinero-Guerra et al., 2019a), Villar (2005) (Villar, 2005), and Marcial (2003) (Marcial,
 432 2003). The green dashed line is obtained using equation (50) with $p_r = 80$ MPa and $n_p = 3.7$.

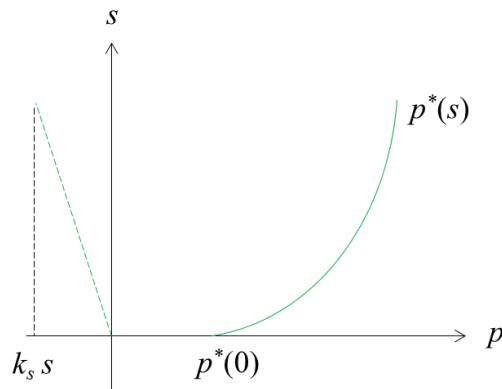
433 In triaxial stress states, the yield surface is written as in the original BBM:

$$q^2 - M^2(p + k_s s)(p^*(s) - p) = 0 \quad (51)$$

434 where q is the deviatoric stress, M is the slope of the critical state line, and k_s a parameter related to the
 435 increase in cohesion with increasing suction (Alonso et al., 1990). An associated flow rule is
 436 considered in the model.

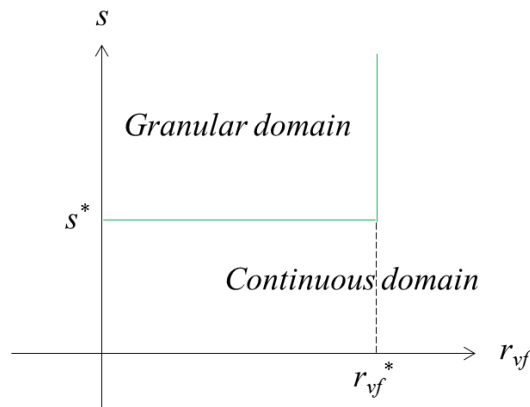
437 A sketch of the elastic domain in the p - s plane is presented in Figure 11. A summary of the model
 438 equations for volumetric strains is presented in Table 6.

439 a)



440

441 b)



442

443 Figure 11: Sketch of the model elastic domain: a) in the p - s plane, b) partition of the elastic domain in the granular and
 444 continuous domains.

445 Table 6: Summary of the model equations related to the volumetric strains.

			Granular domain	Continuous domain
Pellet	ε_{V1}	elastic	Equation (30)	Equation (41)
		plastic	only elastic	only elastic
Powder grains	ε_{V2}	elastic	Equation (31)	Equation (42)
		plastic	only elastic	only elastic
Granular assembly	ε_V	elastic	Equation (33)	Equation (43)
		plastic	Equations (47) to (51)	Equations (47) to (51)

446

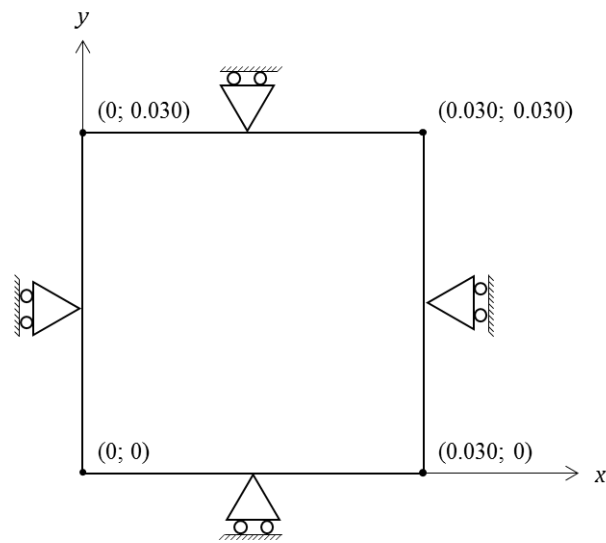
447 5. Simulations of swelling pressure tests

448 The model is implemented in the open source finite element method (FEM) code Bil developed by
449 Dangla (Dangla, 2018). The swelling pressure tests carried out in the laboratory are simulated using
450 the model to assess its ability to reproduce the behaviour of pellet-powder mixture.

451

452 5.1. Method

453 The swelling pressure tests are modelled using two-dimensional axisymmetric conditions. The y
454 axis is the axis of symmetry. The mesh consists in one Q4 element. This latter is presented in Figure
455 12.



456

457 Figure 12: Geometry of the FEM simulations. Lengths are given in m.

458 The following initial conditions for the displacement field, \mathbf{u} , initial stress state and suction are
459 used in all the simulations, for the three swelling pressure tests:

460 - $\mathbf{u} = 0$;

461 - $s_0 = 89$ MPa;

462 - $\sigma_{xx 0} = 10^{-5}$ MPa;

463 - $\sigma_{yy 0} = 10^{-5}$ MPa;

464 - $\sigma_{zz 0} = 10^{-5}$ MPa.

465 where $\sigma_{xx 0}$, $\sigma_{yy 0}$ and $\sigma_{zz 0}$ are the initial diagonal components of the Cauchy stress tensor. Initial mean
466 stress is taken higher than zero to avoid numerical issues associated to zero stress. Gravity is not
467 considered in the simulations. The initial suction in the simulations is taken equal to the pellet initial
468 suction, considered more representative of the whole system than that of the powder.

469 The boundary conditions used in the simulations are:

470 - $u_y = 0$ on the top boundary ($y = 0.030$ m);

471 - $u_y = 0$ on the bottom boundary ($y = 0$);

472 - $u_x = 0$ on the left boundary ($x = 0$);

473 - $u_x = 0$ on the right boundary ($x = 0.030$ m).

474 For simplicity, p_{atm} is taken equal to zero and s is progressively decreased from 89 MPa to 0.1 MPa
475 such that $s + p_{atm}$ is still equal to atmospheric pressure in the final state (equation (45)).

476 The parameters required for the simulations are mixture properties, elastic parameters in the
477 granular domain, elastic parameters in the continuous domain, domain transition parameters, and
478 plasticity parameters. These latter are obtained from the laboratory tests, DEM periodic simulations,
479 literature or estimation if not available.

480 Mixture properties obtained from experimental characterisation are shown in Table 7. Parameters
481 related to the elastic behaviour are recapitulated in Table 8. Parameters related to the granular domain
482 are from (Darde et al., 2018) and DEM simulations presented in a previous section. κ is chosen in the
483 same range as the dimensionless elastic stiffness parameter proposed by (Mokni et al., 2020) for a
484 MX80 bentonite pellet-powder mixture using the same pellets and powder. Parameters related to the
485 granular to continuous transition are presented in Table 9. The threshold suction s^* is chosen according
486 to experimental observations by (Saiyouri et al., 2004) and (Molinero-Guerra et al., 2020) that a
487 significant microstructural rearrangement occurs in MX80 bentonite in the range 3-7 MPa of suction.

488 The value of r_{vf}^* is taken equal to 1. Parameters related to the plastic behaviour are presented in Table
 489 10. λ_0 , ρ and ω are taken in the same range as in (Mokni et al., 2020). p_r and n_p are obtained from
 490 experimental results (Figure 10). The values of κ_m and p_c are estimated so that all swelling pressure
 491 tests are satisfactorily simulated with a single set of parameters.

492 Table 7: Model parameters related to the mixture properties.

Property	70/0	70/15	70/30
x_s	0	0.18	0.30
ϕ_{10}	0.553	0.553	0.553
ϕ_{mat0}	0	0.275	0.529
s_0 : MPa	89	89	89
e_{10}	0.450	0.450	0.450
e_{20}	0.450	0.450	0.450
e_{M0}	1.19	0.722	0.396
e_0	1.64	1.17	0.847

493

494 Table 8: Model parameters related to the elastic behaviour of the material.

Elasticity parameters	All mixtures
<i>Granular domain</i>	
α_m : MPa ⁻¹	0.024
β_m : MPa ⁻¹	0.016
f_{Mm}	1
$f_{\varepsilon a}$	6
$f_{\varepsilon b}$	40
$f_{m a}$	0.067
$f_{m b}$	-0.031

<i>Continuous domain</i>	
κ	0.025
κ_m	0.015

495

496

Table 9: Model parameters related to the granular-continuous transition.

Transition parameters	All mixtures
s^* : MPa	3
r_{vf}^*	1

497

498

Table 10: Model parameters related to the plastic behaviour of the material.

Plasticity parameters	All mixtures
λ_0	0.20
p_c : MPa	0.050
ρ	0.8
ω : MPa ⁻¹	0.1
p_r : MPa	80
n_p	3.7

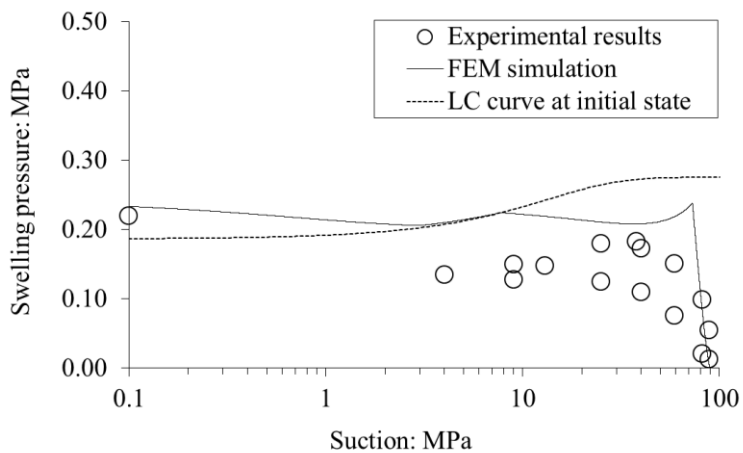
499

500 5.2. FEM simulation results

501 Figure 13a presents the comparison between experimental and numerical results for SP0a and SP0b
502 tests. In the simulation, the swelling pressure increases then reaches a first peak corresponding to
503 pellet strength. Following the peak, swelling pressure remains nearly constant until LC yielding occurs
504 while in the granular domain. Following transition to continuous domain, the swelling pressure
505 increases again to its final value as elastic swelling overcomes the plastic closure of macro-porosity.
506 The general trend of the swelling pressure evolution as well as its order of magnitude are satisfactorily
507 reproduced. The final swelling pressure in the simulation is close to the final swelling pressure in

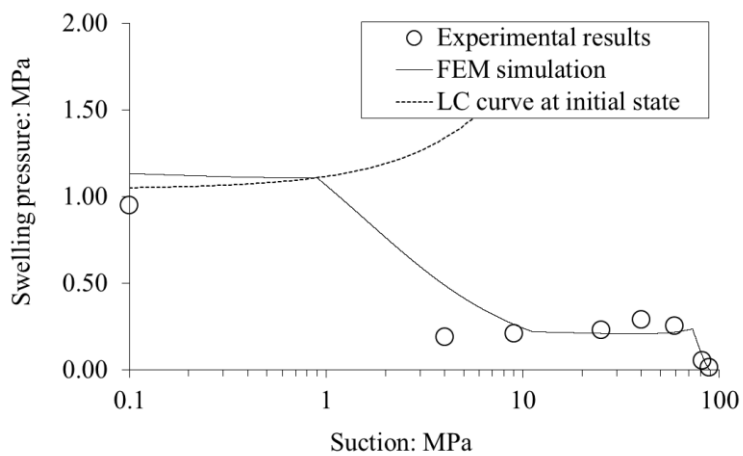
508 experiments. Some differences between experimental and numerical results still can be observed and
509 are discussed in the next section.

510 a)



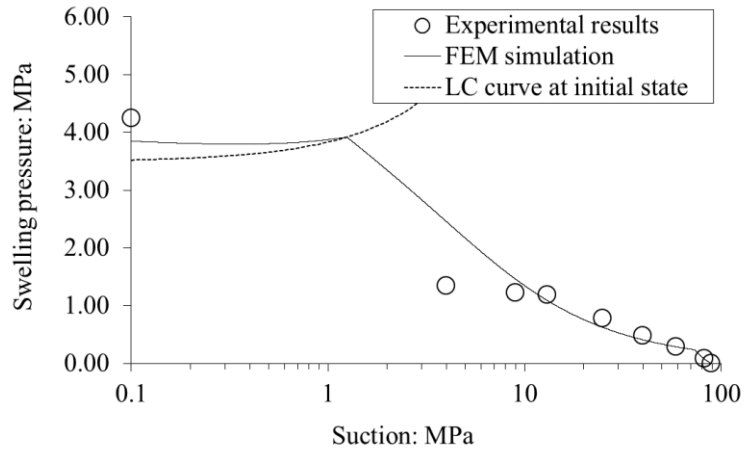
511

512 b)



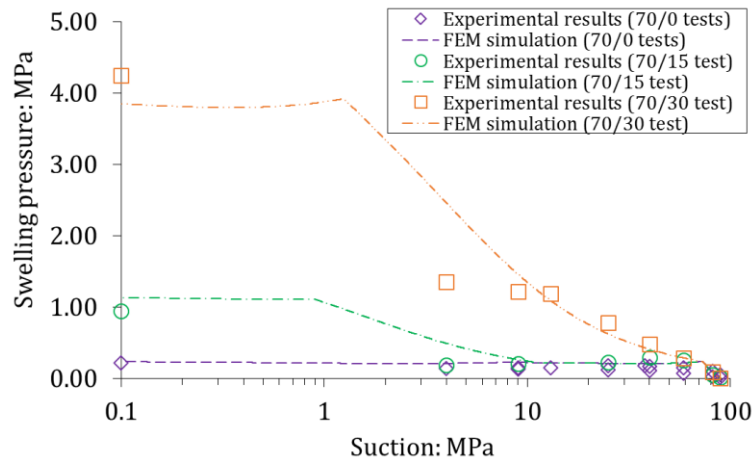
513

514 c)



515

516 d)



517

518 Figure 13: Comparison between FEM simulation and experimental results of swelling pressure tests for a) 70/0 material;
 519 b) 70/15 material; c) 70/30 material, d) comparison of all FEM simulation and experimental results.

520 Figure 13b presents the comparison between experimental and numerical results for SP15 test. In
 521 the simulation, the swelling pressure increases and reaches a peak as pellet strength is reached, then
 522 remains nearly constant until suction reaches ~11 MPa. At this point r_{vf} reaches 1 and the material is
 523 considered continuous. Afterward, swelling pressure increases until reaching the LC yield locus. Then,
 524 the swelling pressure remains nearly constant. The final swelling pressure in the simulation is
 525 overestimated but still close to experimental results.

526 Figure 13c presents the comparison between experimental and numerical results for the 70/30
 527 mixture. Transition occurs as $r_{vf} = 1$ at high suction (~70 MPa in the simulation) and most of the

528 hydration occurs while in the continuous domain. Swelling pressure increases, then LC yielding
529 occurs and the swelling pressure remains nearly constant and reaches a final value close to
530 experimental results. It is interesting to note that, even simplified, the conceptual approach proposed
531 for the continuous domain is able to reproduce the evolution of swelling pressure upon suction
532 decrease.

533 A comparison of all numerical and experimental results is presented in Figure 13d. It is highlighted
534 that the proposed conceptual approach is able to satisfactorily reproduce the mechanical behaviour of
535 pellet-based bentonite materials upon suction decrease in constant-volume conditions, either with no
536 powder or with powder of various densities in the inter-pellet voids, over a wide range of average dry
537 densities (1.05 Mg/m^3 to 1.50 Mg/m^3). Some differences were obtained between experimental results
538 and prediction of the model, as previously mentioned. However, Figure 13d shows that both the
539 general trends and amplitude of the swelling pressure evolution upon suction decrease are still
540 comparable to the experimental results.

541

542 **6. Discussion**

543

544 **6.1. Hypotheses of the conceptual model**

545

546 6.1.1. Distinction of two domains and transition criteria

547 A hypothesis of the model is the consideration of two distinct domains. The mixture is considered
548 either granular, with powder having no contribution to the mechanical behaviour, or continuous, with
549 pellet and powder equally contributing to the mechanical behaviour.

550 The swelling pressure in SP15 test was very close to, but still slightly higher than, SP0a and SP0b
551 swelling pressure. Even if moderate, powder may have an influence in “loose powder phase” mixtures
552 in the granular domain. The eventual remaining influence of the initial granular structure is not

553 considered when the material is in the continuous domain. It is not known how the evolution of the
554 swelling pressure in SP30 test would compare to that of a compacted powder sample of same dry
555 density. The final state, following water saturation, was homogeneous. No influence of the initial
556 granular structure could be observed (Figure 5). This observation has also been made in other
557 experimental works at laboratory scale (Hoffmann et al., 2007; Imbert and Villar, 2006; van Geet et
558 al., 2005).

559 In the light of experimental results, considering two distinct domains in a conceptual approach is
560 relevant, but attention must be drawn on the fact that distinction of two domains is a simplification of
561 the material behaviour. Transition from the initial granular state to the final continuous state is likely
562 to be more progressive. Nevertheless, the model was able to reproduce the evolution of swelling
563 pressure in three swelling pressure tests performed on three different mixtures, using a single set of
564 parameters.

565

566 6.1.2. Transition criteria

567 The transition criteria have to determine conditions in which the material behaviour can no longer
568 be considered controlled by the pellet assembly. Two criteria have been proposed: (i) The relative
569 volume fraction of powder and pellets granular assemblies; (ii) suction at which the microstructure of
570 bentonite rearrange, inducing changes of the macroscopic response under hydromechanical loadings.

571 For binary granular mixtures, the conceptual approach of the dominant phase (de Larrard, 2014;
572 Roquier, 2015; Roux et al., 2007; Ueda et al., 2011; Westman and Hugill, 1930; Yerazunis et al.,
573 1965) can estimate the volumetric proportion of fine particles corresponding to a transition between a
574 macroscopic behaviour controlled by the assembly of coarse grains to a macroscopic behaviour
575 controlled by the assembly of fine particles. However, in bentonite pellet and powder mixtures, pellets
576 and powder grains swell, their volumetric strains are comparable, and the observed transition can
577 occur at relative volumetric proportions close to the initial state. It is the reason why the r_{vf} parameter
578 is introduced. Since pellets swell upon hydration, the available volume for powder grains to swell

579 decreases and the powder phase becomes progressively denser, even if the volumetric proportions are
580 the same as the initial state. However, the determination of the threshold value r_{vf}^* is tricky. In the
581 present work, a value of $r_{vf}^* = 1$ was assumed. Since data was lacking, it was assumed that the pellet
582 assembly can no longer control the macroscopic behaviour if the density of the powder grains in the
583 inter-pellet porosity reaches the density of the pellet assembly. This choice is considered relevant by
584 comparison with experiment. It was highlighted in the 70/30 test that the influence of powder was
585 non-negligible for suction below 60 MPa. Using equations (30) and (31) assuming that p is negligible
586 compared to s yields $r_{vf}(s = 60 \text{ MPa}) = 1.05$. However, it is likely that a single value of this parameter
587 would not suit all mixtures. The r_{vf}^* parameter should be determined with care and not necessarily
588 taken equal to 1.

589 In bentonite materials, experimental results (Saiyouri et al., 2004) evidence that exfoliation of clay
590 particles occurs upon adsorption of water molecule layers. When the clay particle is no longer stable, a
591 sharp decrease of the number of layers per particle is observed. For MX80 bentonite, this threshold is
592 in the range 3-7 MPa and corresponds to the development of diffuse layers, microstructural
593 rearrangements, modification of the pore size distribution, and change in the water retention behaviour
594 (Keller et al., 2014; Molinero-Guerra et al., 2020; Saiyouri et al., 2004; Villar, 2007). MX80 pellets
595 and powder (crushed pellets) are thus affected by significant microstructural rearrangement in this
596 suction range. It is considered that the initial granular structure is not the main feature controlling the
597 macroscopic response for suction below a threshold suction, s^* .

598

599 6.1.3. Granular material

600 The constitutive equations for the granular domain are based on DEM simulation results of large
601 pellet assemblies. The use of DEM to simulate pellet assemblies from the behaviour of one pellet has
602 been discussed in (Darde et al., 2020a). It was highlighted that modelling pellets by spheres of same
603 diameter, same stiffness and same strength could induce an overestimation of the macroscopic
604 stiffness of the mixture and of the peak pressure attributed to pellet strength, respectively related to $f_{e a}$

605 and m_g^* in the present work. It was also evidenced that wall effects in small samples can influence the
606 material behaviour.

607 In the present work, a value of $f_{\varepsilon a} = 6$ was suggested from DEM results, which can also make the
608 mixture stiffer in FEM simulation than in experiments (Figure 13a). The peak pressure in FEM
609 simulations was also higher than in experiments. Nevertheless, both the macroscopic stiffness and
610 peak pressure appeared to be in the right order of magnitude for all mixtures.

611 The post peak behaviour in the granular domain exhibits a moderate decrease followed by a
612 moderate increase of swelling pressure. After the peak, the material macroscopic stiffness is mainly
613 related to $f_{\varepsilon b}$. It is worthy to mention that in DEM, for $m_g > m_g^*$, $d\varepsilon_v/dm_g$ was increasing even if the ε_v -
614 m_g relationship appears sublinear (Figure 7). Thus, even if $f_{\varepsilon b} = 40$ was suggested (from linear fit),
615 higher values could have also been relevant.

616 It was demonstrated that $\frac{\partial \varepsilon_v^{(e)}}{\partial \varepsilon_{V1}} = 1$ in a granular assembly with elastic contacts. This value has been
617 used in all simulations. For plastic contacts, f_{Mm} progressively becomes smaller than 1. The assumption
618 of a constant “elastic” f_{Mm} induces larger volumetric strain of the mixture upon pellet volume
619 variations. However, as the macroscopic volumetric strain also depends on $f_{\varepsilon b}$ through variations of
620 m_g , a convenient way of handling this problem is to choose a relevant couple $(f_{Mm}, f_{\varepsilon b})$ to reproduce the
621 behaviour of the mixture when $m_g > m_g^*$.

622 Finally, as presented in Figure 7, the repeatability of the results is very good. Variability associated
623 to small size of the sample discussed in (Darde et al., 2020a) is avoided in the present study. The 4000
624 particles periodic sample can be considered representative of a pellet assembly which allows the
625 intrinsic behaviour of pellet assemblies to be addressed with few simulations.

626

627 6.1.4. Continuous material

628 In the continuous domain, a modified BBM (Alonso et al., 1990) was proposed to model the
629 material behaviour. This approach was also adopted to model bentonite-based pellet materials in other

630 studies by various authors (Alonso et al., 2011; Gens et al., 2011; Hoffmann et al., 2007; Mokni et al.,
631 2020; Sánchez et al., 2016), with different modifications of the original modelling framework. In the
632 present work, the BBM approach is used only for the continuous domain because it is not able to
633 reproduce features observed in the laboratory such as the free swelling powder in the 70/15 mixture,
634 the influence of the powder phase in the 70/30 mixture, or the two peaks of swelling pressure observed
635 in the 70/0 mixture.

636 Compared to other numerical studies from the literature, the proposed modified BBM is simpler
637 and does not account for features such as plasticity in the microstructure (Alonso et al., 2011) or
638 damage in the microstructure (Mokni et al., 2020), partial saturation of the microstructure (Alonso et
639 al., 2011; Sánchez et al., 2016), inter-aggregate porosity in the pellets (Navarro et al., 2020a, 2020b),
640 or plastic macroscopic strain resulting from microstructural swelling (Gens et al., 2011; Mokni et al.,
641 2020; Sánchez et al., 2016). Nevertheless, it is able to satisfactorily reproduce the evolution of
642 swelling pressure for all mixtures, notably the 70/30 which was considered continuous in the
643 simulation during most of the hydration path.

644 The simple model for the continuous domain must be regarded as part of the global approach,
645 considering two domains. Even if it is simplified compared to traditional BBM-based modelling
646 approaches, it aims at reproducing only the final part of the hydration path. It allows intrinsic features
647 of the material behaviour to be directly accounted for, for instance change in the macroscopic response
648 depending on the density of the powder phase, evolution of the macroscopic swelling potential with
649 density, homogenisation of the mixture, using a single set of parameters.

650

651 6.1.5. Hydraulic equilibrium

652 Hydraulic equilibrium was assumed during all the simulations for simplicity. In the case of pellet-
653 powder mixture, owing to the low permeability and size difference between pellets and powder grains,
654 it is likely that equation (20) is not realistic. (Gens et al., 2011) and (Sánchez et al., 2016) considered

655 two different suctions for pellets and powder in simulations with a simplified transfer law between the
656 two levels of structure.

657 A consequence of the hydraulic equilibrium assumption is that pellet swelling would be too fast in
658 simulations of hydration paths not performed at equilibrium as in the present experimental study (use
659 of the vapour equilibrium technique).

660

661 **6.2. Ability to reproduce the material behaviour**

662

663 6.2.1. Two peaks in no-powder samples

664 In FEM simulation of the 70/0 tests, a first peak occurs at a suction of ~ 70 MPa as a consequence
665 of contact forces reaching pellet strength. Macroscopic yielding occurs at suction of ~ 7 MPa and a
666 second peak is reached as a result. This latter was also observed in experiments but at lower suction.
667 The overestimation of f_{Mm} for $m_g > m_g^*$ allowed the swelling pressure to reach the LC yield locus in the
668 simulation earlier than expected from experimental results. Yet, the second peak in FEM occurs at a
669 swelling pressure comparable to the second peak in the experiment.

670

671 6.2.2. Influence of the powder phase in loose powder samples

672 In the granular domain, the material response is well reproduced by the model (Figure 13b).
673 Transition to the continuous domain and subsequent increase of swelling pressure in the simulation are
674 found to occur earlier than in experiment. In experiment, swelling pressure remains nearly constant
675 until a suction of 4 MPa. It can be suggested that pellets swell too rapidly in FEM simulation, thus ϕ_{mat}
676 increases faster in the simulation and the transition is computed at higher suction. This would mean
677 that equilibrium in experiment was not perfectly reached after each suction step. Another suggestion to
678 explain this difference between simulation and experiment is that r_{vf}^* is not a constant value and may
679 be higher in 70/15 mixture than in 70/30 mixture. Even if the swelling pressure at $s = 4$ MPa is

680 overestimated, it is interesting to note that the overall response provided by the model is quite close to
681 the experimental one, involving a succession of very different phenomena.

682

683 6.2.3. Swelling pressure evolution

684 In spite of some differences between simulations and experiments, Figure 13d highlights that the
685 overall ability of the model to reproduce the evolution of swelling pressure is good. It predicts the
686 significant influence of the initial granular structure in the 70/15 mixture and the moderate influence of
687 the latter in the 70/30 mixture. It predicts also the trend of the evolution of swelling pressure rather
688 well for all mixtures, either increase or plateau or decrease, at values of swelling pressure and suction
689 close to experimental observation. Macroscopic yielding and peak phenomena are also well
690 reproduced. The final swelling pressure is close to the experimental one in the three simulations.

691 In the model, irreversible strain can arise only if the transition criteria or the yield locus are
692 reached. In experiments, only one hydration path has been performed. More complex hydration paths
693 such as wetting-drying cycles, accumulating plastic strains, would not be properly reproduced by the
694 present model.

695

696 6.2.4. Applicability to field conditions

697 In contact with liquid water, pellets would lose their granular structure (Hoffmann et al., 2007;
698 Molinero-Guerra et al., 2019b) and the evolution of swelling pressure in the granular domain would
699 not be observed. However, in field conditions, vapour diffusion is likely to be the dominant hydration
700 mechanism (Kröhn, 2005). Thus, the influence of the initial granular structure is a relevant feature to
701 take into account. Besides, local heterogeneities of density are likely to arise during installation in the
702 galleries (Molinero-Guerra et al., 2017). Local heterogeneities of density influence the relative
703 proportion of pellets and powder and the local value of the mixture dry density. Powder density has
704 been shown to have a significant influence on the mechanical behaviour. In this regard, the proposed

705 model is relevant because it allows to account for heterogeneities of density with a single set of
706 parameters. In repository conditions, hydration of the mixture will be progressive. Thus, the
707 consideration of two different suctions for pellets and powder grains in the model would be necessary.

708

709 **7. Conclusion**

710 Bentonite pellet-powder mixtures are candidate materials for the sealing of galleries in radioactive
711 waste disposal concepts. The mechanical behaviour of these materials is influenced by their initial
712 granular structure and the density of the powder phase.

713 In the present study, swelling pressure tests have been carried out on three mixtures with the same
714 pellet volume fraction and different powder contents. It was highlighted that the initial granular
715 structure has no significant influence on the behaviour of mixtures with dense powder phase.
716 Conversely, the powder phase has no significant influence on the behaviour of mixtures with loose
717 powder phase until a low suction.

718 The behaviour of pellet assemblies has been studied under isotropic compression through DEM
719 simulations. It was highlighted that the initial pellet volume fraction, pellet stiffness and pellet strength
720 have a notable influence on the macroscopic behaviour of the pellet assembly. In addition, it was
721 demonstrated that, upon pellet swelling, the volumetric strain of a pellet assembly is equal to the
722 volumetric strain of the pellet under the assumption of elastic contact reaction.

723 Based on experimental and DEM results, a conceptual model for pellet-powder mixture has been
724 proposed. The model is formulated in the framework of hardening elastoplasticity and considers two
725 distinct domains in elasticity. Depending on suction and the density of the powder phase, the mixture
726 is either considered granular or continuous. In the granular domain, the pellet assembly controls the
727 behaviour of the mixture and constitutive laws are proposed from DEM results. In the continuous
728 domain, pellets and powder contributes to the mechanical behaviour of the mixture and a modified
729 BBM is used to model its behaviour.

730 The model was used to simulate the swelling pressure tests performed in the laboratory. Owing to
731 its conceptual nature, some differences have been noted between numerical and experimental results.
732 However, the numerical results generally reproduced very well the experimental results, using a single
733 set of parameters for all mixtures. Swelling pressures obtained in simulations were very close to
734 experimental ones. These results improve the knowledge on the behaviour of bentonite pellet-powder
735 mixtures during hydration in constant-volume conditions.

736 An interesting perspective arising from the present work is a better characterisation of the transition
737 from the behaviour controlled by the pellet assembly to the continuous behaviour, which is a
738 progressive phenomenon. The influence of the powder density on the macroscopic response can be
739 investigated in this respect. Besides, water transfer laws in pellets should be proposed to avoid the
740 hypothesis of hydraulic equilibrium. The proposed modelling approach can then be used to provide
741 interesting predictive simulations of pellet-powder mixtures in repository conditions, to better assess
742 the long-term safety of these facilities.

743

744 **Declaration of competing interests**

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

747

748 **List of references**

749 Agnolin, I., Roux, J.N., 2007. Internal states of model isotropic granular packings. I. Assembling
750 process, geometry, and contact networks. *Phys. Rev. E - Stat. Nonlinear, Soft Matter Phys.* 76,
751 1–27. <https://doi.org/10.1103/PhysRevE.76.061302>

752 Alonso, E.E., Gens, A., Josa, A., 1990. A constitutive model for partially saturated soils G ”.
753 *Géotechnique* 40, 405–430. <https://doi.org/10.1680/geot.1990.40.3.405>

754 Alonso, E.E., Hoffmann, C., Romero, E., 2010. Pellet mixtures in isolation barriers. *J. Rock Mech.*

755 Geotech. Eng. 2, 12–31. <https://doi.org/10.3724/SP.J.1235.2010.00012>

756 Alonso, E.E., Romero, E., Hoffmann, C., 2011. Hydromechanical behaviour of compacted granular
757 expansive mixtures: experimental and constitutive study. *Géotechnique* 61, 329–344.
758 <https://doi.org/10.1680/geot.2011.61.4.329>

759 Bernachy-barbe, F., Conil, N., Guillot, W., Talandier, J., 2020. Observed heterogeneities after
760 hydration of MX-80 bentonite under pellet/powder form. *Appl. Clay Sci.* 189, 105542.
761 <https://doi.org/10.1016/j.clay.2020.105542>

762 Bian, X., Cui, Y., Li, X., 2019. Voids effect on the swelling behaviour of compacted bentonite.
763 *Géotechnique* 69, 593–605. <https://doi.org/10.1680/jgeot.17.P.283>

764 Christoffersen, J., Mehrabadi, M.M., 1981. A Micromechanical Description of Granular Material
765 Behavior. *J. Appl. Mech.* 48, 339–344.

766 Dangla, P., 2018. Bil-2.6 A modeling platform based on finite volume/element methods (Version
767 v2.6). <https://doi.org/http://doi.org/10.5281/zenodo.1419841>

768 Darde, B., Roux, J.-N., Pereira, J.-M., Dangla, P., Talandier, J., Vu, M.N., Tang, A.M., 2020a.
769 Investigating the hydromechanical behaviour of bentonite pellets by swelling pressure tests and
770 discrete element modelling. *Acta Geotech.* <https://doi.org/10.1007/s11440-020-01040-5>

771 Darde, B., Tang, A.M., Pereira, J.-M., Roux, J.-N., Dangla, P., Talandier, J., Vu, M.N., 2018. Hydro-
772 mechanical behaviour of high-density bentonite pellet on partial hydration. *Géotechnique Lett.* 8,
773 330–335. <https://doi.org/10.1680/jgele.18.00114>

774 Darde, B., Tang, A.M., Roux, J., Dangla, P., Pereira, J., Talandier, J., Vu, M.N., 2020b. Effects of the
775 initial granular structure of clay sealing materials on their swelling properties: experiments and
776 DEM simulations. *Eur. Phys. J. Nucl. Sci. Technol.* 6.
777 <https://doi.org/https://doi.org/10.1051/epjn/2019059>

778 de Larrard, F., 2014. Concrete mixture proportioning: a scientific approach. CRC Press.

779 Ferrari, A., Favero, V., Marschall, P., Laloui, L., 2014. Experimental analysis of the water retention

780 behaviour of shales. *Int. J. Rock Mech. Min. Sci.* 72, 61–70.
781 <https://doi.org/10.1016/j.ijrmms.2014.08.011>

782 Gens, A., Valleján, B., Sánchez, M., Imbert, C., Villar, M.V., Van Geet, M., 2011. Hydromechanical
783 behaviour of a heterogeneous compacted soil: experimental observations and modelling.
784 *Géotechnique* 61, 367–386. <https://doi.org/10.1680/geot.SIP11.P.015>

785 Hoffmann, C., Alonso, E.E., Romero, E., 2007. Hydro-mechanical behaviour of bentonite pellet
786 mixtures. *Phys. Chem. Earth* 32, 832–849. <https://doi.org/10.1016/j.pce.2006.04.037>

787 Imbert, C., Villar, M.V., 2006. Hydro-mechanical response of a bentonite pellets/powder mixture upon
788 infiltration. *Appl. Clay Sci.* 32, 197–209. <https://doi.org/10.1016/j.clay.2006.01.005>

789 Jia, L., Chen, Y., Ye, W., Cui, Y., 2019. Effects of a simulated gap on anisotropic swelling pressure of
790 compacted GMZ bentonite. *Eng. Geol.* 248, 155–163.
791 <https://doi.org/10.1016/j.enggeo.2018.11.018>

792 Johnson, K.L., 1985. *Contact Mechanics*. Cambridge University Press, Cambridge, UK.
793 <https://doi.org/10.1115/1.3261297>

794 Kaufhold, S., Baille, W., Schanz, T., Dohrmann, R., 2015. About differences of swelling pressure -
795 dry density relations of compacted bentonites. *Appl. Clay Sci.* 107, 52–61.
796 <https://doi.org/10.1016/j.clay.2015.02.002>

797 Keller, L.M., Seiphoori, A., Gasser, P., Lucas, F., Holzer, L., Ferrari, A., 2014. The pore structure of
798 compacted and partly saturated MX-80 bentonite at different dry densities. *Clays Clay Miner.* 62,
799 174–187. <https://doi.org/10.1346/CCMN.2014.0620302>

800 Kröhn, K.P., 2005. New evidence for the dominance of vapour diffusion during the re-saturation of
801 compacted bentonite. *Eng. Geol.* 82, 127–132. <https://doi.org/10.1016/j.enggeo.2005.09.015>

802 Lloret, A., Villar, M. V, Sanchez, M., Gens, A., Pintado, X., Alonso, E., 2003. Mechanical behaviour
803 of heavily compacted bentonite under high suction changes. *Geotechnique* 53, 27–40.
804 <https://doi.org/10.1680/geot.53.1.27.37258>

805 Marcial, D., 2003. Comportement hydromécanique et microstructural des matériaux de barrière
806 ouvragée. Doctoral dissertation, Ecole nationale des ponts et chaussées, France.

807 Mokni, N., Molinero-Guerra, A., Cui, Y.-J., Delage, P., Aïmediou, P., Bornert, M., Tang, A.M., 2020.
808 Modelling the long-term hydro-mechanical behaviour of a bentonite pellet/powder mixture with
809 consideration of initial structural heterogeneities. *Géotechnique* 70, 563–580.
810 <https://doi.org/10.1680/jgeot.18.p.110>

811 Molinero-Guerra, A., Cui, Y.J., He, Y., Delage, P., Mokni, N., Tang, A.M., Aïmediou, P., Bornert, M.,
812 Bernier, F., 2019a. Characterization of water retention, compressibility and swelling properties of
813 a pellet/powder bentonite mixture. *Eng. Geol.* 248, 14–21.
814 <https://doi.org/10.1016/j.enggeo.2018.11.005>

815 Molinero-Guerra, A., Delage, P., Cui, Y.-J., Mokni, N., Tang, A.M., Aïmediou, P., Bernier, F.,
816 Bornert, M., 2020. Water retention properties and microstructure changes of a bentonite pellet
817 upon wetting/drying; application to radioactive waste disposal. *Géotechnique* 70, 199–209.
818 <https://doi.org/10.1680/jgeot.17.p.291>

819 Molinero-Guerra, A., Mokni, N., Cui, Y., Delage, P., Tang, A.M., Aïmediou, P., Bornert, M., 2019b.
820 Impact of initial structural heterogeneity on long term swelling behavior of MX80 bentonite
821 pellet/powder mixtures. *Can. Geotech. J.* <https://doi.org/10.1139/cgj-2018-0301>

822 Molinero-Guerra, A., Mokni, N., Delage, P., Cui, Y.J., Tang, A.M., Aïmediou, P., Bernier, F., Bornert,
823 M., 2017. In-depth characterisation of a mixture composed of powder/pellets MX80 bentonite.
824 *Appl. Clay Sci.* 135, 538–546. <https://doi.org/10.1016/j.clay.2016.10.030>

825 Navarro, V., Asensio, L., Gharbieh, H., De, G., Pulkkanen, V., 2020a. A triple porosity hydro-
826 mechanical model for MX-80 bentonite pellet mixtures. *Eng. Geol.* 265.
827 <https://doi.org/10.1016/j.enggeo.2019.105311>

828 Navarro, V., Asensio, L., Morena, G. De, Gharbieh, H., Alonso, J., 2020b. From double to triple
829 porosity modelling of bentonite pellet mixtures. *Eng. Geol.* 274.

- 830 <https://doi.org/10.1016/j.enggeo.2020.105714>
- 831 Roquier, G., 2015. Etude théorique des effets de paroi et de desserrement dans les mélanges
832 granulaires. Doctoral dissertation, Ecole nationale des ponts et chaussées, France.
- 833 Roux, J.-N., Chevoir, F., Toussaint, F., 2007. Etats de compacité maximale pour les mélanges binaires
834 de grains sphériques : étude par simulation numérique. Bull. des Lab. des Ponts Chaussées 268–
835 269, 141–152.
- 836 Saba, S., Cui, Y., Tang, A.M., Barnichon, J., 2014. Investigation of the swelling behaviour of
837 compacted bentonite-sand mixtures by mock-up tests. Can. Geotech. J. 51, 1399–1412.
- 838 Saiyouri, N., Hicher, P.Y., Tessier, D., 2000. Microstructural approach and transfer water modelling in
839 highly compacted unsaturated swelling clays. Mech. Cohesive-frictional Mater. 5, 41–60.
- 840 Saiyouri, N., Tessier, D., Hicher, P.Y., 2004. Experimental study of swelling in unsaturated compacted
841 clays. Clay Miner. 39, 469–479. <https://doi.org/10.1180/0009855043940148>
- 842 Sánchez, M., Gens, A., Villar, M.V., Olivella, S., 2016. Fully Coupled Thermo-Hydro-Mechanical
843 Double-Porosity Formulation for Unsaturated Soils. Int. J. Geomech. D4016015.
844 [https://doi.org/10.1061/\(ASCE\)GM.1943-5622.0000728](https://doi.org/10.1061/(ASCE)GM.1943-5622.0000728)
- 845 Tang, A.-M., Cui, Y.-J., 2005. Controlling suction by the vapour equilibrium technique at different
846 temperatures and its application in determining the water retention properties of MX80 clay.
847 Can. Geotech. J. 42, 287–296. <https://doi.org/10.1139/t04-082>
- 848 Ueda, T., Matsushima, T., Yamada, Y., 2011. Effect of particle size ratio and volume fraction on shear
849 strength of binary granular mixture. Granul. Matter 13, 731–742. [https://doi.org/10.1007/s10035-](https://doi.org/10.1007/s10035-011-0292-1)
850 [011-0292-1](https://doi.org/10.1007/s10035-011-0292-1)
- 851 van Geet, M., Volckaert, G., Roels, S., 2005. The use of microfocus X-ray computed tomography in
852 characterising the hydration of a clay pellet/powder mixture. Appl. Clay Sci. 29, 73–87.
853 <https://doi.org/10.1016/j.clay.2004.12.007>
- 854 Villar, M.V., 2005. MX80 bentonite. Thermo-Hydro-Mechanical Characterisation Performed at

- 855 CIEMAT in the Context of the Prototype Project. Inf. Técnicos Ciemat 1053.
- 856 Villar, M. V., 2007. Water retention of two natural compacted bentonites. *Clays Clay Miner.* 55, 311–
857 322. <https://doi.org/10.1346/CCMN.2007.0550307>
- 858 Wang, Q., Tang, A.M., Cui, Y.J., Delage, P., Barnichon, J.D., Ye, W.M., 2013. The effects of
859 technological voids on the hydro-mechanical behaviour of compacted bentonite-sand mixture.
860 *Soils Found.* 53, 232–245. <https://doi.org/10.1016/j.sandf.2013.02.004>
- 861 Wang, Q., Tang, A.M., Cui, Y.J., Delage, P., Gatmiri, B., 2012. Experimental study on the swelling
862 behaviour of bentonite/claystone mixture. *Eng. Geol.* 124, 59–66.
863 <https://doi.org/10.1016/j.enggeo.2011.10.003>
- 864 Westman, A.E.R., Hugill, H.R., 1930. the Packing of Particles. *J. Am. Ceram. Soc.* 13, 767–779.
865 <https://doi.org/10.1111/j.1151-2916.1930.tb16222.x>
- 866 Yerazunis, S., Cornel, S.W., Wintner, B., 1965. Dense random packing of binary mixtures of spheres.
867 *Nature* 207, 835.
- 868 Zhang, Z., Ye, W., Liu, Z., Wang, Q., Cui, Y., 2020. Mechanical behavior of GMZ bentonite pellet
869 mixtures over a wide suction range. *Eng. Geol.* 264, 105383.
870 <https://doi.org/10.1016/j.enggeo.2019.105383>

871

872 **Appendix A**

873 In a granular assembly, the diagonal components of the Cauchy stress tensor can be written as
874 follows:

$$\sigma_{\alpha\alpha} = \frac{1}{\Omega} \left(\sum_{i=1}^{N_c} F_{N\alpha} r_{\alpha} \right) \quad (52)$$

875 where α is the reference coordinate axis ($1 \leq \alpha \leq 3$), Ω is the total volume of the granular assembly, N_c
876 is the number of contacts, r_{α} are the α -coordinates of vector \mathbf{r} joining the centre of particles in contact,

877 $F_{N\alpha}$ are the α -coordinates of the corresponding contact force F_N (Agnolin and Roux, 2007;
 878 Christoffersen and Mehrabadi, 1981). From this expression, the mean stress p in the granular assembly
 879 is written as:

$$p = \frac{1}{3\Omega} \sum_{\alpha=1}^3 \left(\sum_{i=1}^{N_c} F_{N\alpha} r_{\alpha} \right) \quad (53)$$

880 The volume fraction of pellet Φ_l and the coordination number Z of the granular assembly are
 881 written as follows:

$$\Phi_1 = \frac{N_1 \Omega_1}{\Omega} \quad (54)$$

882 And

$$Z = \frac{2N_c}{N_1} \quad (55)$$

883 where N_l is the number of spheres and Ω_l is the sphere volume.

884 From (54) and (55), in conditions where normal deflection at contact is negligible compared to the
 885 sphere diameters (*i.e.* $r_{\alpha} \approx a$), (53) can be written as:

$$p = \frac{Z\Phi_1}{\pi a^2} F_N^* \quad (56)$$

886 where F_N^* is the mean contact normal force in the granular assembly.

887 From equations (5) and (56), the following expression can be written:

$$\left(\frac{p}{\frac{E_1}{1 - \nu_1^2}} \right)^{\frac{2}{3}} = \frac{\delta_N^*}{a} \left(\frac{Z \Phi_1}{3\pi} \right)^{\frac{2}{3}} \quad (57)$$

888 where δ_N^* is the average normal deflection. From equation (57), the dimensionless parameter m_g is
 889 defined as:

$$m_g = \left(\frac{p}{\frac{E_1}{1 - \nu_1^2}} \right)^{\frac{2}{3}} \quad (58)$$

890

891 **Appendix B**

892 In a granular assembly of spherical particles, considering elastic contacts and particle volume
 893 increase at constant particle stiffness and particle Young modulus, the mean stress in the granular
 894 assembly can be written from the Hertz law (equation (5)) and equation (56):

$$p_0 = \frac{1}{3 \Omega_0} N_c \overline{(r_0 F_{N0})} \quad (59)$$

$$p = \frac{1}{3 \Omega_0 (1 + \varepsilon_V)} N_c \overline{(r_0 (1 + \varepsilon_V)^{\frac{1}{3}} F_N)} \quad (60)$$

895 Where p_0 and F_{N0} are the values of p and F_N before particle swelling Ω_0 is the initial value of Ω , r_0
 896 is the initial value of r , the norm of the \mathbf{r} vector, and the horizontal bar denotes the average value over
 897 all contacts. F_N , accounting for the volume variations of the granular assembly resulting from the
 898 volume variations of the spherical particles, is written:

$$F_N = \frac{1}{3} E_1 (a_0)^{\frac{1}{2}} (1 + \varepsilon_{V1})^{\frac{1}{6}} \left[r_0 (1 + \varepsilon_V)^{\frac{1}{3}} - a_0 (1 + \varepsilon_{V1})^{\frac{1}{3}} \right]^{\frac{3}{2}} \quad (61)$$

899 Where a_0 is the initial value of a . In equations (60) and (61) , $r_0(1 + \varepsilon_V)^{\frac{1}{3}}$ and $r_0(1 + \varepsilon_V)^{\frac{1}{3}} -$
900 $a_0(1 + \varepsilon_{V1})^{\frac{1}{3}}$ respectively denotes the distance between two spherical particles in contacts and the
901 deflection in contact between two spherical particles.

902 Following a small increase in the particle volumes, if no rearrangement of the granular assembly
903 occurs and N_c remains constant, then

$$p = p_0 \leftrightarrow \varepsilon_V = \varepsilon_{V1} \quad (62)$$

904 Thus

$$f_{Mm} = \frac{\partial \varepsilon_V^{(e)}}{\partial \varepsilon_{V1}} = 1 \quad (63)$$

905

906

907 **List of Figures**

908 Figure 1: a) Picture of a pellet; b) Schematic view of a pellet.

909 Figure 2: Schematic view of the experimental setup. Red arrows represent humid air circulation.

910 Figure 3: Evolution of swelling pressure as a function of elapsed time. a) SP0a test, b) SP0b test, c)
911 SP15 test, d) SP30 test.

912 Figure 4: Evolution of swelling pressure as a function of suction upon hydration. a) Swelling
913 pressure on a logarithmic scale, b) Swelling pressure on a linear scale, limited to swelling pressures
914 lower than 2 MPa. Horizontal arrows represent the final swelling pressure of the tested materials.

915 Figure 5: Pictures of SP0 samples. a) SP0a at 9 MPa of suction, b) SP0b following liquid water
916 saturation. The cell diameter is 60 mm.

917 Figure 6: Conceptual interpretation of the behaviour of pellet-powder materials upon hydration in
918 constant-volume conditions. The red solid lines represent the influence of the initial granular structure.
919 The blue dashed lines represent the behaviour after the transition to the continuous domain.

920 Figure 8: Evolution of ε_V as a function of m_g during isotropic compression for all granular
921 assemblies. a) A samples, b) B samples, c) C samples, d) D samples, e) E samples, f) comparison
922 between A1, B1, C1, D1, E1.

923 Figure 9: Comparison of the evolution of x_{plas} as a function of m_g for the five types of granular
924 assemblies.

925 Figure 10: Relationship between m_g^* and Φ_{10} for all samples. The dashed line represents the fit
926 corresponding to $m_g^* = 0.067 \Phi_{10} - 0.031$.

927 Figure 11: Preconsolidation pressure as a function of the void ratio in oedometer tests performed on
928 bentonite materials by Molinero-Guerra et al. (2019) (Molinero-Guerra et al., 2019a), Villar (2005)
929 (Villar, 2005), and Marcial (2003) (Marcial, 2003). The green dashed line is obtained using equation
930 (59) with $p_r = 80$ MPa and $n_p = 3.7$.

931 Figure 12: Sketch of the model elastic domain: a) in the p - s plane, b) partition of the elastic domain
932 in the granular and continuous domains.

933 Figure 13: Geometry of the FEM simulations. Lengths are given in m.

934 Figure 14: Comparison between FEM simulation and experimental results of swelling pressure
935 tests for a) 70/0 material; b) 70/15 material; c) 70/30 material, d) comparison of all FEM simulation
936 and experimental results.

937

938 **List of Tables**

939 Table 1: Initial properties of the pellets.

940 Table 2: Properties of the pellet-powder mixtures.

941 Table 3: Hydration paths followed by the samples. Suctions are given in MPa.

942 Table 4: Parameters of the model for a pellet.

943 Table 5: Initial properties of the numerical samples. All samples are cubic and contain 4000
944 spheres.

945 Table 6: Summary of the model equations related to the volumetric strains.

946 Table 7: Model parameters related to the mixture properties.

947 Table 8: Model parameters related to the elastic behaviour of the material.

948 Table 9: Model parameters related to the granular-continuous transition.

949 Table 10: Model parameters related to the plastic behaviour of the material.

950