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**Title:**

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

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**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 $e_M$
$f_{ma}$	Model parameter related to contact plasticity
$f_{mb}$	Model parameter related to contact plasticity
$f_{Mm}$	Model parameter
$F_N$	Normal contact force

$F_{N0}$	Initial value of $F_N$
$F_{N\alpha}$	$\alpha$ -coordinates of the contact force $F_N$
$F_N^*$	Mean contact normal force in the granular assembly
$\mathbf{F}_T$	Tangential reaction vector
$f_\varepsilon$	Dimensionless stiffness parameter for the granular assembly
$f_{\varepsilon a}$	Value of $f_\varepsilon$ if contacts are considered elastic
$f_{\varepsilon p}$	Value of $f_\varepsilon$ 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
$m_1$	Mass of the pellet
$m_g$	Dimensionless pressure parameter
$m_g^*$	Limit value of $m_g$
$N_I$	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 $r_{vf}$
$r_\alpha$	$\alpha$ -coordinates of vector $\mathbf{r}$
$s$	Total suction
$s^*$	Threshold suction
$s_0$	Initial suction
$s_I$	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
$\alpha_m$	Model parameter related to microstructural stiffness
$\beta_m$	Model parameter related to microstructural stiffness
$\delta_N$	Normal deflection
$\delta_N^*$	Average normal deflection
$\varepsilon_V$	Total volumetric strain
$\varepsilon_{V I}$	Volumetric strain of the pellet

$\varepsilon_{V2}$	Volumetric strains of the powder grains
$\varepsilon_{VM}$	Volumetric strain of the macrostructure
$\kappa$	Dimensionless elastic stiffness parameter of the macrostructure related to changes in mean stress
$\kappa_m$	Dimensionless elastic stiffness parameter of the microstructure
$\kappa_s$	Dimensionless elastic stiffness parameter of the macrostructure related to changes in suction
$\lambda$	Dimensionless elastoplastic stiffness parameter of the macrostructure
$\lambda_0$	Value of $\lambda$ in fully saturated conditions
$\nu_1$	Poisson ratio of the pellet
$\rho$	Ratio $\lambda(s \rightarrow \infty)/\lambda_0$
$\rho_d$	Dry density of the mixture
$\rho_{d10}$	Initial pellet dry density
$\rho_s$	Density of solid particles
$\sigma_{\alpha\alpha}$	Diagonal components of the Cauchy stress tensor
$\sigma_{xx0}$	Initial xx components of the Cauchy stress tensor
$\sigma_{yy0}$	Initial yy components of the Cauchy stress tensor
$\sigma_{zz0}$	Initial zz components of the Cauchy stress tensor
$\Phi_1$	Pellet volume fraction in experiments
$\phi_1$	Pellet volume fraction in the model
$\Phi_{10}$	Initial pellet volume fraction in experiments
$\phi_{10}$	Initial pellet volume fraction in the model
$\Phi_2$	Powder volume fraction in experiments
$\phi_2$	Powder volume fraction in the model
$\Phi_{20}$	Initial powder volume fraction in experiment
$\phi_M$	Volume fraction of macrostructural voids
$\phi_{mat}$	Volume fraction of the powder in inter-pellet voids

$\phi_{mat\ 0}$	Initial volume fraction of the powder in inter-pellet voids
$\omega$	Model parameter related to the increase in $\lambda$ with suction
$\Omega$	Total volume of the granular assembly
$\Omega_0$	Initial value of $\Omega$
$\Omega_I$	Volume of pellet
$\Omega_s$	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
$\Omega_v$	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

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## 1. Introduction

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

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

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

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

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

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

## 2. Swelling pressure tests

### 2.1. Tested materials

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

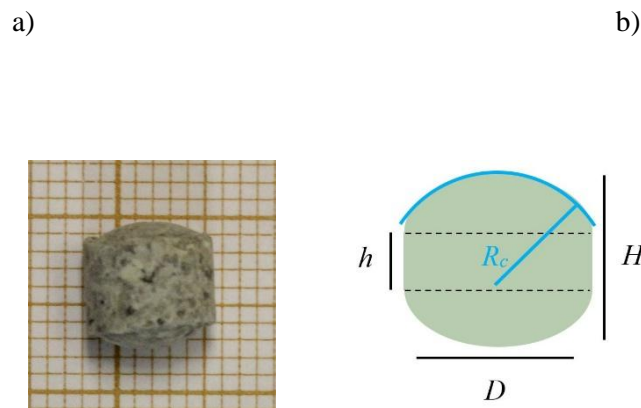


Figure 1: a) Picture of a pellet; b) Schematic view of a pellet.

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, $\rho_s$ (Saba et al., 2014)	2.77 Mg/m <sup>3</sup>
Initial dry density, $\rho_{d10}$	1.91 Mg/m <sup>3</sup>
Initial void ratio, $e_{10}$	0.45
Initial suction, $s_0$	89 MPa

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

Three pellet-powder mixtures are prepared at the same pellet volume fraction and three different powder volume fractions. Pellet initial volume fraction,  $\Phi_{10}$  (volume of pellets / total volume), is 0.553 in all samples. The three powder initial volume fractions,  $\Phi_{20}$ , are 0.236, 0.118, and 0. The highest value of  $\Phi_{20}$  corresponds to pellet/powder proportions of 70/30 in dry mass. To highlight that  $\Phi_{10}$  remains identical in all samples, the three mixtures are referred to as 70/30, 70/15, and 70/0 in the following. The dry densities ( $\rho_d$ ) of the 70/30, 70/15, and 70/0 materials are 1.50 Mg/m<sup>3</sup>, 1.275 Mg/m<sup>3</sup>, and 1.05 Mg/m<sup>3</sup>, respectively.

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## 2.2. Method

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

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

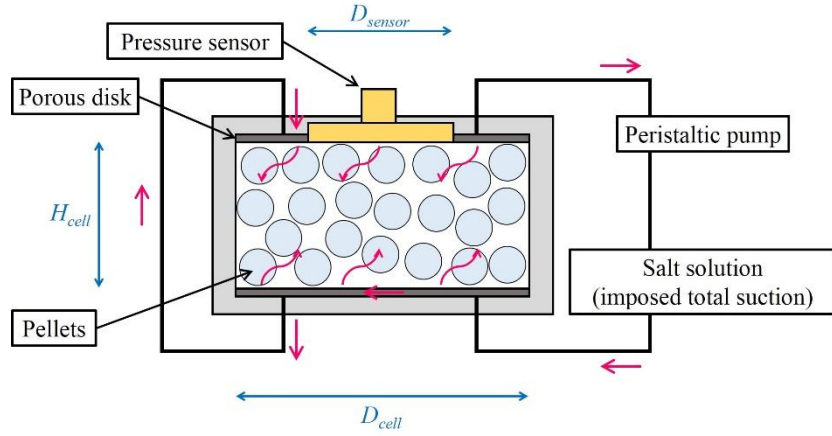


Figure 2: Schematic view of the experimental setup. Red arrows represent humid air circulation.

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

Table 2: Properties of the pellet-powder mixtures.

Sample	Mixture	$\Phi_{10}$	$\Phi_{20}$	$\rho_d$ : Mg/m <sup>3</sup>
SP30	70/30	0.553	0.236	1.50
SP15	70/15		0.118	1.275
SP0a	70/0		0	1.05
SP0b	70/0		0	1.05

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

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

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*

\*: samples wetted using synthetic liquid water

## 2.3. Experimental results and discussion

### 2.3.1. Evolution of swelling pressure upon hydration

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

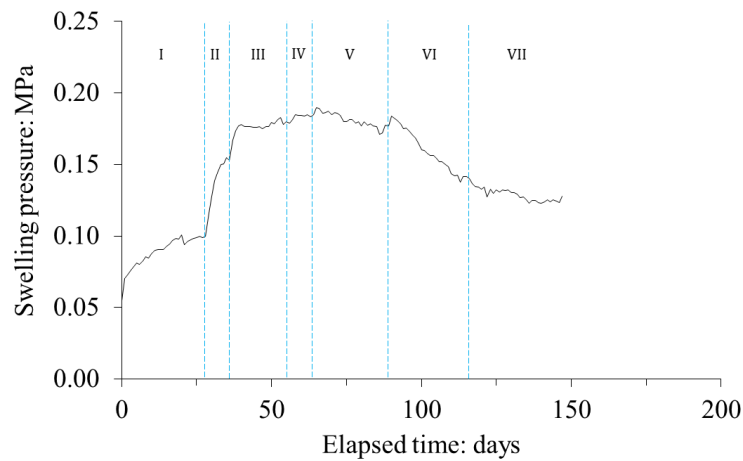
The swelling pressure in SP0a increases from 0.055 MPa (pressure following the closure of the cell) to 0.173 MPa, from initial state to  $s = 38$  MPa. Upon hydration from  $s = 38$  MPa to  $s = 9$  MPa, the swelling pressure decreases to 0.128 MPa.

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

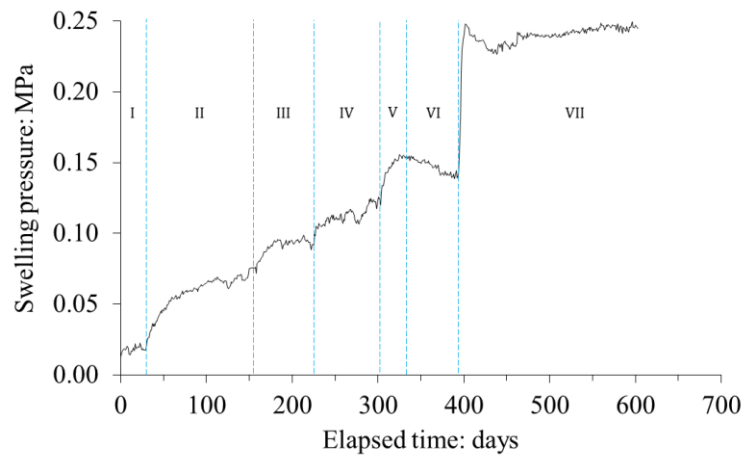
The swelling pressure in SP15 increases from 0.015 MPa to 0.290 MPa from initial state to  $s = 40$  MPa. Upon hydration from  $s = 40$  MPa to  $s = 4$  MPa, the swelling pressure decreases to 0.200 MPa. Following liquid water flooding, the swelling pressure increases and stabilises at 0.910 MPa after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

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

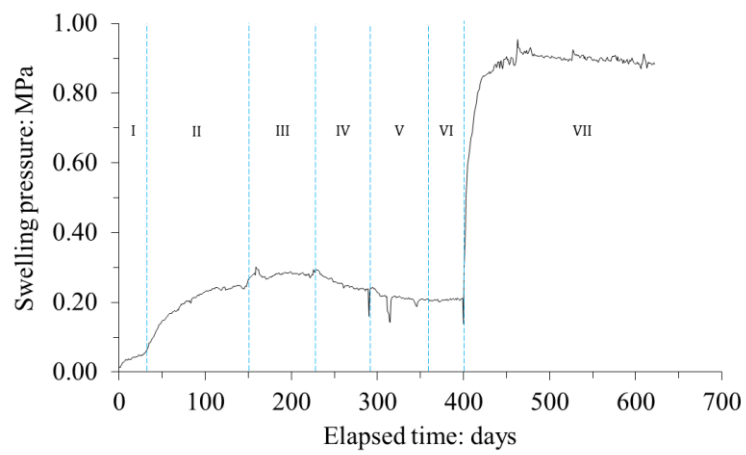
a)



b)



c)



d)

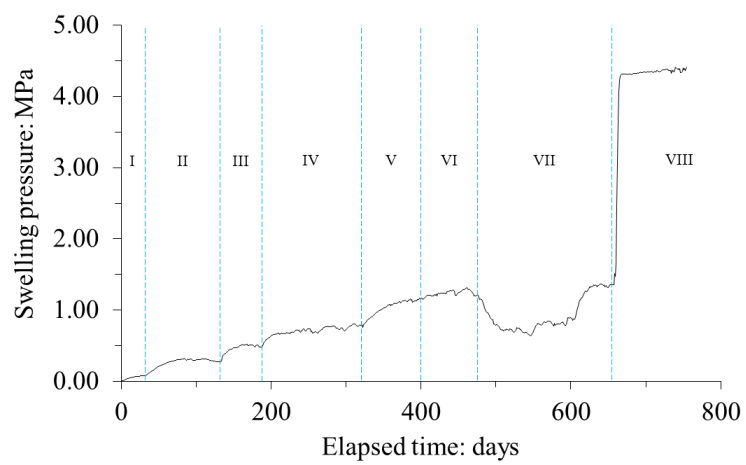
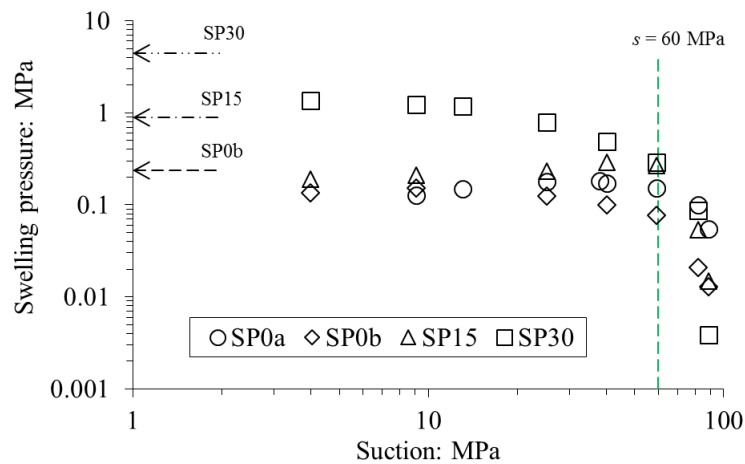


Figure 3: Evolution of swelling pressure as a function of elapsed time. a) SP0a test, b) SP0b test, c) SP15 test, d) SP30 test.

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

a)



b)



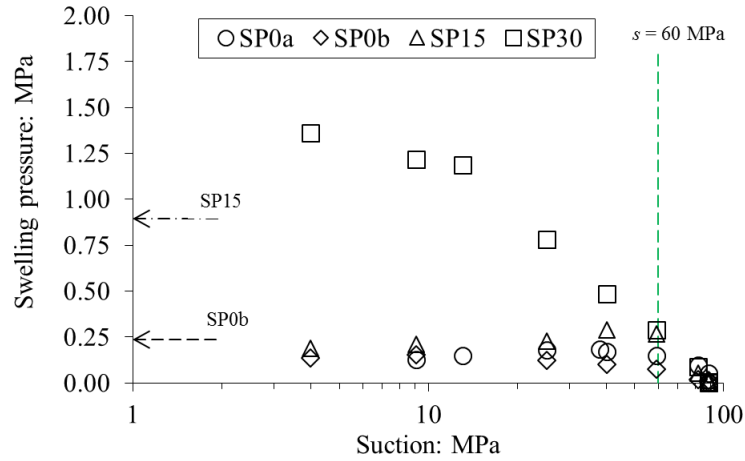


Figure 4: Evolution of swelling pressure as a function of suction upon hydration. a) Swelling pressure on a logarithmic scale, b) Swelling pressure on a linear scale, limited to swelling pressures lower than 2 MPa. Horizontal arrows represent the final swelling pressure of the tested materials.

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

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

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

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

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

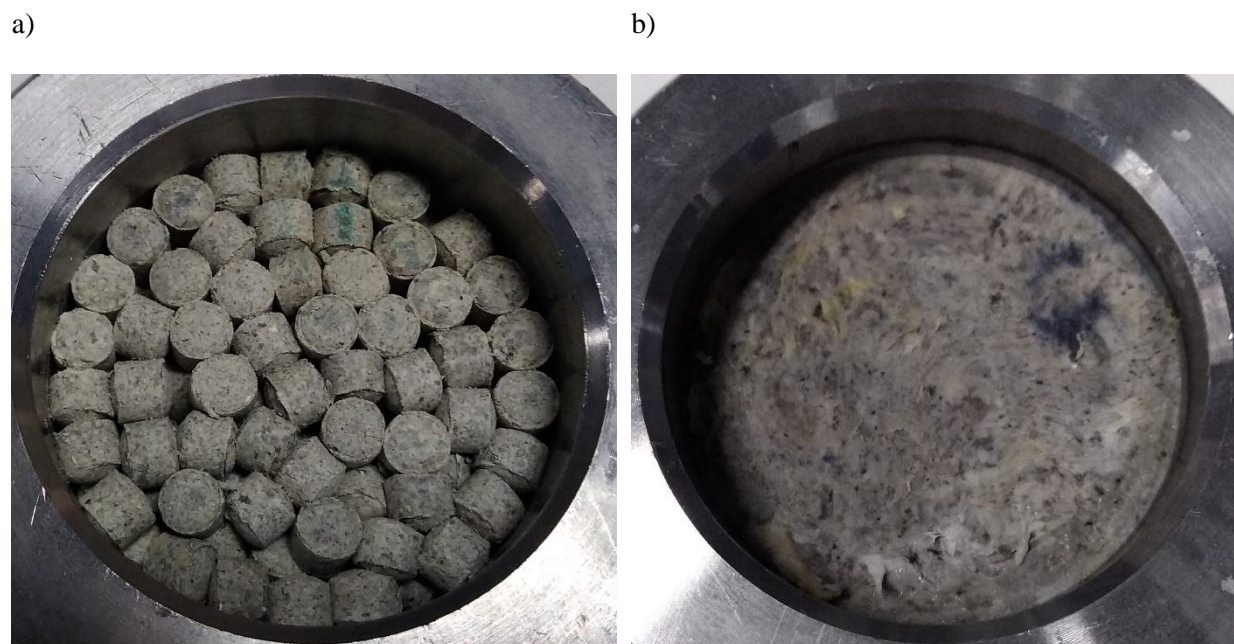


Figure 5: Pictures of SP0 samples. a) SP0a at 9 MPa of suction, b) SP0b following liquid water saturation. The cell diameter is 60 mm.

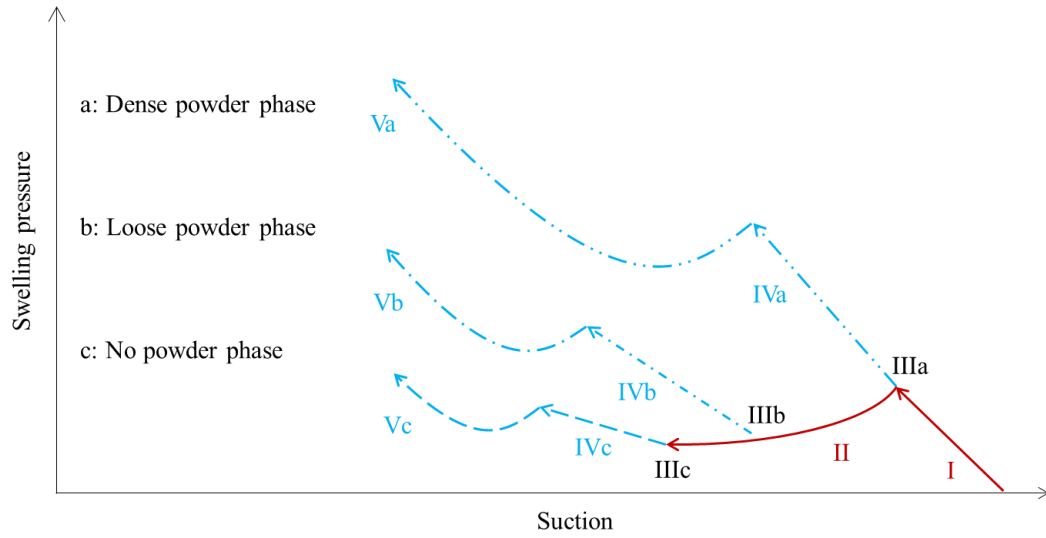
### 2.3.3. Conceptual interpretation of the material behaviour

Experimental results highlight that the material behaviour is controlled by the initial granular structure until powder volume fraction  $\Phi_2$  reaches a maximum value. For the 70/30 material (with  $\Phi_{I0} = 0.553$ ), it occurs at  $s \approx 60$  MPa. Besides, all materials, including the 70/0 material, appear to have reached a high degree of homogenisation in the fully saturated state. In pellet assemblies, the influence of initial granular structure is lost between  $s = 4$  MPa and  $s = 0$  MPa. In this suction range, it has been evidenced that a significant reorganisation of the material fabric occurs (Saiyouri et al., 2004, 2000). Aggregates exfoliation in the macroporosity occurs, the microstructure and hydration mechanisms change, and it is suggested that the initial granular structure no longer controls the macroscopic behaviour. The key phenomena controlling the behaviour of pellet-powder materials are thus: (i) the pellet stiffness; (ii) the pellet strength; (iii) a threshold value of  $\Phi_2$ ; (iv) a threshold value of  $s$ ; and (v) the behaviour of the material when the influence of the initial granular structure is lost.

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

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

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



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

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

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

#### 3.1. Objectives

Existing modelling framework used to describe the behaviour of compacted bentonite materials do not account for the features described in the previous section. Since the pellet assembly can control the mechanical response of the material if  $\Phi_2$  is below a threshold value, it is proposed in the present section to study the mechanical behaviour of pellet assemblies with no powder.

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

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## 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,  $E_I$ , volumetric strain,  $\varepsilon_{V1}$ , and compressive  
 219 strength,  $R_I$ :

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

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

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

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

220 where  $\nu_1$  is the pellet Poisson ratio,  $p'$  is the effective mean stress,  $p'_0$  is the initial effective mean  
 221 stress,  $C_A$ ,  $\alpha_m$  and  $\beta_m$  are model parameters,  $p$  is the total mean stress and  $s$  the total suction.

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

Table 4: Parameters of the model for a pellet.

Parameter	Value
$\alpha_m$	$0.024 \text{ MPa}^{-1}$
$\beta_m$	$0.016 \text{ MPa}^{-1}$
$\nu_1$	0.3
$C_A$	$1.206 \times 10^{-7} \text{ m}^{-2}$
$s_0$	89 MPa

### 3.2.2. Contact laws

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

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

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

Where  $\delta_N$  is the normal deflection.

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

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

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

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

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

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

$m_g$  is a convenient parameter to study the behaviour of the pellet assembly, since a single term depending on both mean stress and pellet stiffness can account for features such as deflection in 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  $\Phi_{10}$   
 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		$Z$	$\Phi_{10}$
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

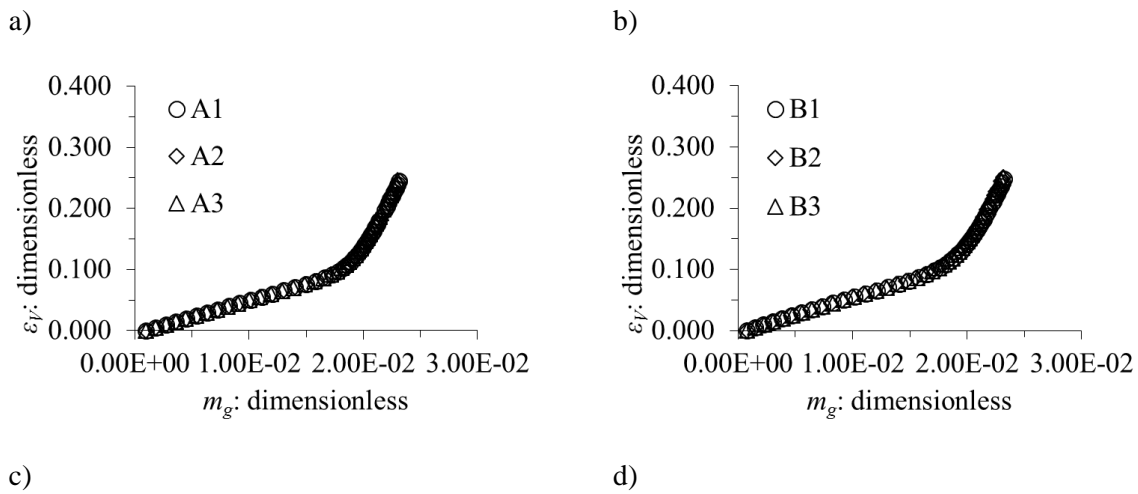


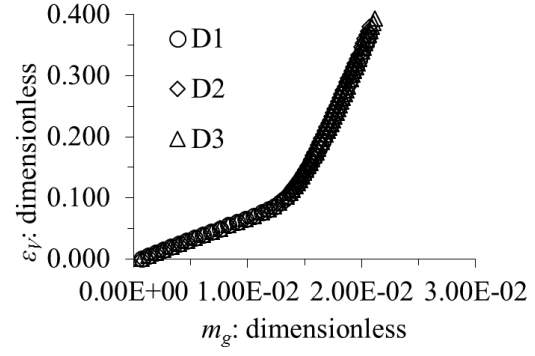
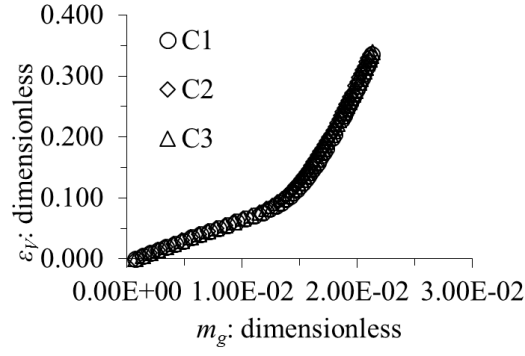
		3	4.3	0.635
	C	1	4.4	0.595
		2	4.3	0.593
		3	4.4	0.594
	D	1	4.5	0.577
		2	4.4	0.575
		3	4.4	0.578
	E	1	5.8	0.628
		2	5.8	0.627
		3	5.8	0.627

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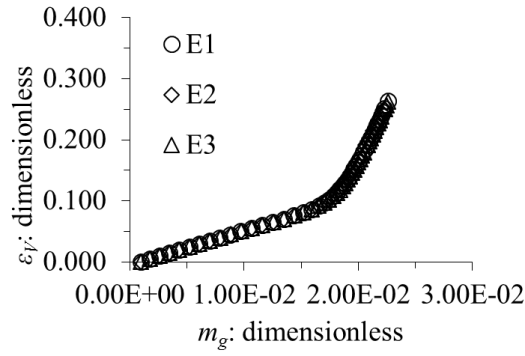
### 277 3.3. DEM simulation results and discussion

278 The evolution of volumetric strain, denoted by  $\varepsilon_V$ , 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.

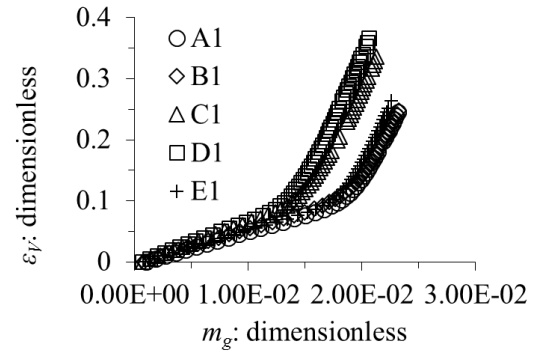




e)



f)



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282

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

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

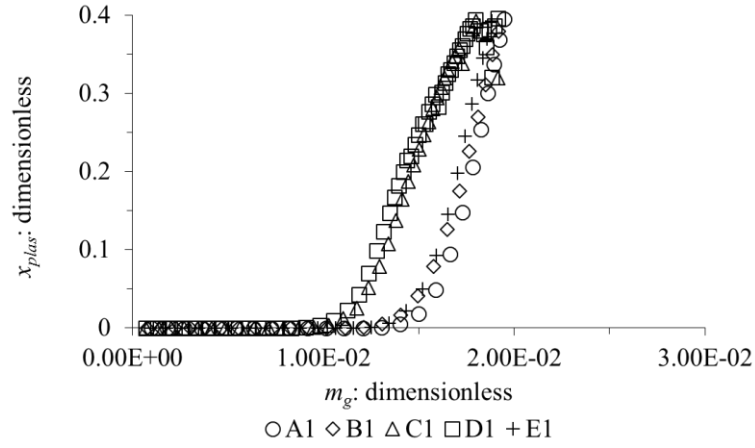


Figure 8: Comparison of the evolution of  $x_{plas}$  as a function of  $m_g$  for the five types of granular assemblies.

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.

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  $\varepsilon_V$  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  $\varepsilon_V$ - $m_g$  relationship is interpreted as a consequence of contact forces reaching the pellet strength.

From Figure 7, it is highlighted that  $\Phi_{10}$  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  $\Phi_{10}$ . The relationship between these two values is plotted in Figure 9 for all samples.

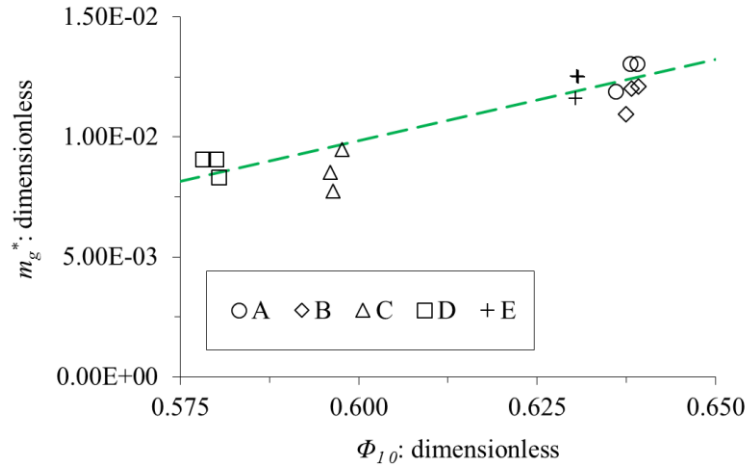


Figure 9: Relationship between  $m_g^*$  and  $\Phi_{I0}$  for all samples. The dashed line represents the fit corresponding to  $m_g^* = 0.067 \Phi_{I0} - 0.031$ .

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

##### 4.1. Conceptual approach

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

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

Three different pore types are considered: the pellet void ratio is denoted by  $e_1$ ; the powder grains 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  $\Omega_{s1}$  and  $\Omega_{s2}$  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.  $\Omega_{v1}$  and  $\Omega_{v2}$  are the volume of void in the pellets and powder  
 327 grains (microstructural pores), and  $\Omega_v$  is the total volume of voids in the mixture. Denoting by  $\Omega_{vM}$  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:

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

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

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

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

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

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

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

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

where  $\phi_1$ ,  $\phi_2$ ,  $\phi_M$  are the volume fractions of pellets, powder, and macrostructural voids. For convenience, an additional volume fraction,  $\phi_{mat}$ , is considered.  $\phi_{mat}$  is the volume fraction of the granular assembly composed of the powder grains within inter-pellet voids:

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

Suction in pellets is denoted by  $s_1$ , suction in powder is denoted by  $s_2$ , suction in macropores is 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  $\phi_i$ ,  $p_i$  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:

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

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

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

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

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

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

It was discussed in (Darde et al., 2020b, 2018) that the model used to study the behaviour of a pellet assembly using DEM was no longer relevant at low suction. Indeed, bentonite materials undergo significant microstructural changes at low suction that have an influence on the macroscopic response under hydromechanical loadings (Molinero-Guerra et al., 2020; Saiyouri et al., 2004; Villar, 2007). For instance, the significant increase of swelling pressure observed at suction lower than 4 MPa in the swelling pressure tests, regardless of the powder content, (Figure 3) cannot be reproduced using the DEM model. These phenomena are not related to the initial granular structure. Thus, in the proposed 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  $\varepsilon_{V1}$ :

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

385 Denoting  $\frac{\partial \varepsilon_V^{(e)}}{\partial m_g}$  by  $f_\varepsilon$  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  $f_\varepsilon$  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  $\phi_{10}$  (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

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

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

with

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

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

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

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

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

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

and

$$d\varepsilon_{V2}^{(e)} = d\varepsilon_{V2} = \frac{\kappa_m}{1 + e_{20}} \frac{dp'_2}{p'_2} \quad (42)$$

where  $\kappa_m$  is a dimensionless stiffness parameter.

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

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

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

$$x_s = \frac{\Omega_{s2}}{\Omega_s} \quad (44)$$

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

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

where  $\kappa$  and  $\kappa_s$  are dimensionless stiffness parameters related to variations of  $p$  and  $s$ , respectively, and  $p_{atm}$  is the atmospheric pressure.

The dependency of the macrostructural swelling potential on the material density is accounted for by assuming a linear relationship between  $\kappa_s$  and  $\kappa_m$ :

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

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  $\lambda$  as a function of suction is  
 422 described as follows:

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

423 where  $\lambda_0$ ,  $\rho$ , and  $\omega$  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):

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

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

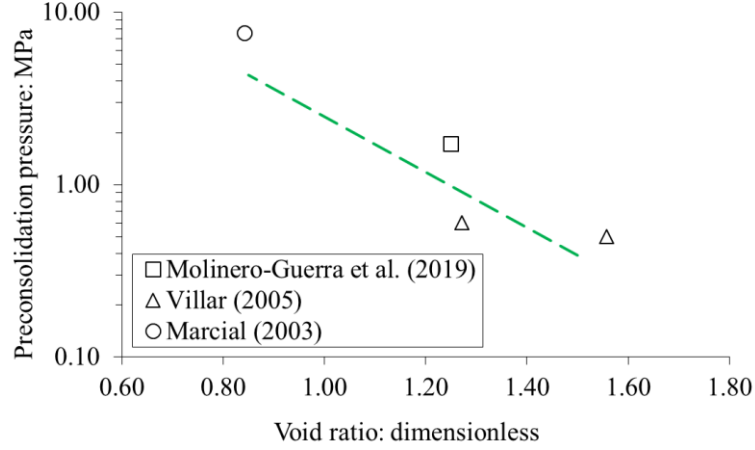


Figure 10: Preconsolidation pressure as a function of the void ratio in oedometer tests performed on bentonite materials by Molinero-Guerra et al. (2019) (Molinero-Guerra et al., 2019a), Villar (2005) (Villar, 2005), and Marcial (2003) (Marcial, 2003). The green dashed line is obtained using equation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

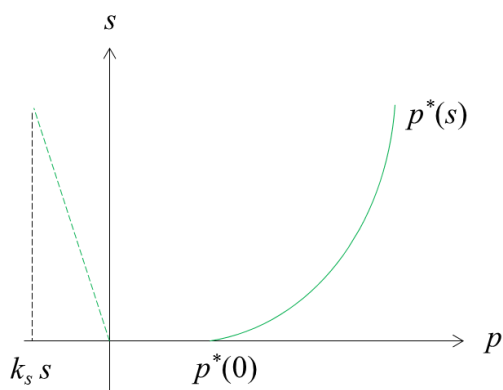
In triaxial stress states, the yield surface is written as in the original BBM:

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

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

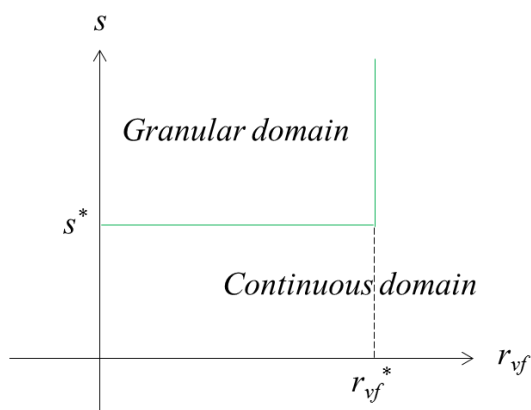
A sketch of the elastic domain in the  $p$ - $s$  plane is presented in Figure 11. A summary of the model equations for volumetric strains is presented in Table 6.

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	$\varepsilon_{V1}$	elastic	Equation (30)	Equation (41)
		plastic	only elastic	only elastic
Powder grains	$\varepsilon_{V2}$	elastic	Equation (31)	Equation (42)
		plastic	only elastic	only elastic
Granular assembly	$\varepsilon_V$	elastic	Equation (33)	Equation (43)
		plastic	Equations (47) to (51)	Equations (47) to (51)

446

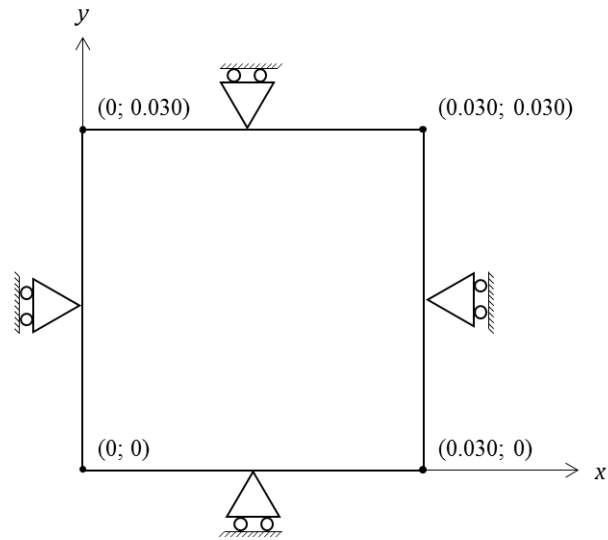
## 447 5. Simulations of swelling pressure tests

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 -  $s_0 = 89 \text{ MPa}$ ;

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



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.  $\kappa$  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.  $\lambda_0$ ,  $\rho$  and  $\omega$  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  $\kappa_m$  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
$\phi_{10}$	0.553	0.553	0.553
$\phi_{mat0}$	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>	
$\alpha_m$ : MPa <sup>-1</sup>	0.024
$\beta_m$ : MPa <sup>-1</sup>	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>	
$\kappa$	0.025
$\kappa_m$	0.015

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

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

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

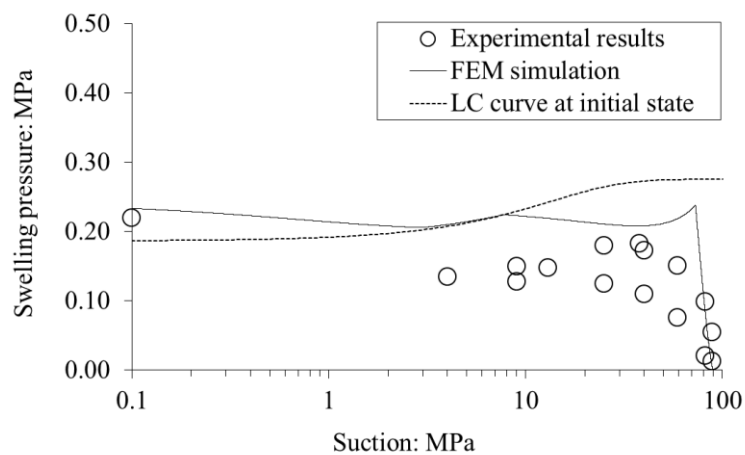
Plasticity parameters	All mixtures
$\lambda_0$	0.20
$p_c$ : MPa	0.050
$\rho$	0.8
$\omega$ : MPa <sup>-1</sup>	0.1
$p_r$ : MPa	80
$n_p$	3.7

## 5.2. FEM simulation results

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

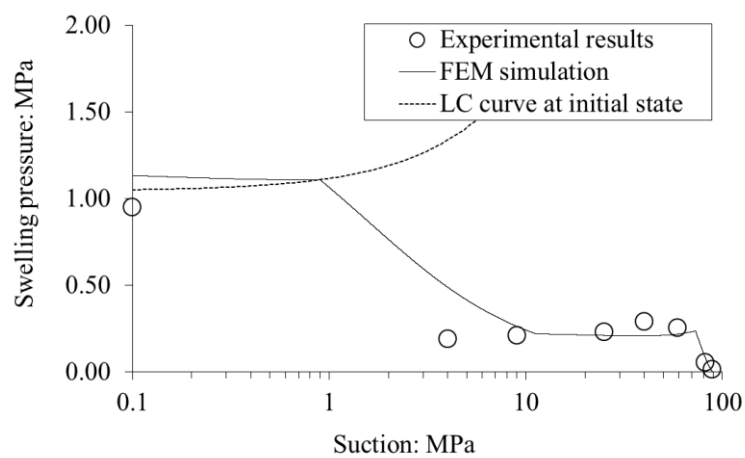
508 experiments. Some differences between experimental and numerical results still can be observed and  
 509 are discussed in the next section.

510 a)



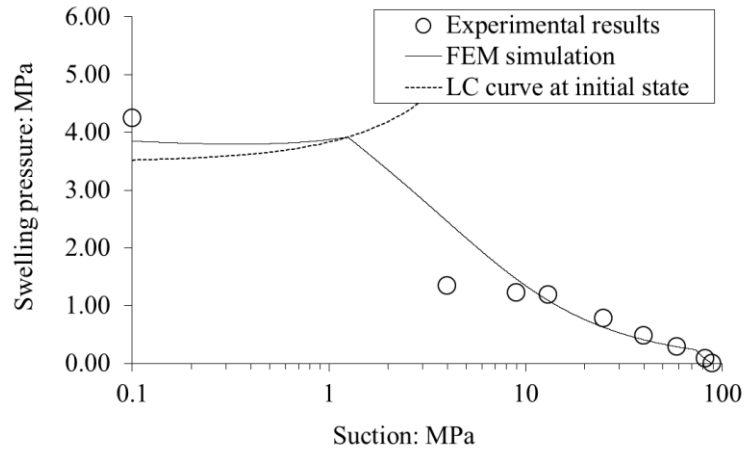
511

512 b)



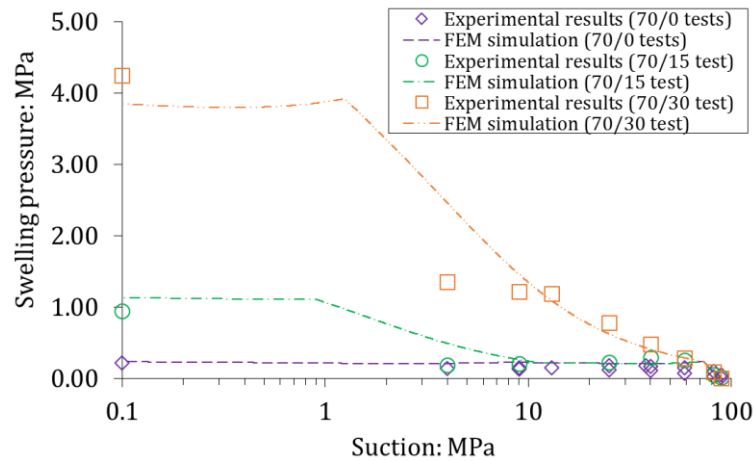
513

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  $\sim 11$  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 ( $\sim 70$  MPa in the simulation) and most of the

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

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

## **6. Discussion**

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

#### **6.1.1. Distinction of two domains and transition criteria**

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

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

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

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

#### 6.1.2. Transition criteria

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

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

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

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

### 6.1.3. Granular material

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



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

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

The post peak behaviour in the granular domain exhibits a moderate decrease followed by a moderate increase of swelling pressure. After the peak, the material macroscopic stiffness is mainly 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  $\varepsilon_v$ - $m_g$  relationship appears sublinear (Figure 7). Thus, even if  $f_{\varepsilon b} = 40$  was suggested (from linear fit), higher values could have also been relevant.

It was demonstrated that  $\frac{\partial \varepsilon_v^{(e)}}{\partial \varepsilon_{v1}} = 1$  in a granular assembly with elastic contacts. This value has been used in all simulations. For plastic contacts,  $f_{Mm}$  progressively becomes smaller than 1. The assumption of a constant “elastic”  $f_{Mm}$  induces larger volumetric strain of the mixture upon pellet volume variations. However, as the macroscopic volumetric strain also depends on  $f_{\varepsilon b}$  through variations of  $m_g$ , a convenient way of handling this problem is to choose a relevant couple  $(f_{Mm}, f_{\varepsilon b})$  to reproduce the behaviour of the mixture when  $m_g > m_g^*$ .

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

#### 6.1.4. Continuous material

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

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

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

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

#### 6.1.5. Hydraulic equilibrium

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

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

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

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

### **6.2.1. Two peaks in no-powder samples**

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

### **6.2.2. Influence of the powder phase in loose powder samples**

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

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

### 6.2.3. Swelling pressure evolution

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

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

### 6.2.4. Applicability to field conditions

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

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

## **7. Conclusion**

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

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

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

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

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

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

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

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

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## Appendix A

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

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

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

877  $F_{N\alpha}$  are the  $\alpha$ -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  $\Phi_I$  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  $N_I$  is the number of spheres and  $\Omega_I$  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  $\delta_N^*$  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):

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

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

895 Where  $p_0$  and  $F_{N0}$  are the values of  $p$  and  $F_N$  before particle swelling  $\Omega_0$  is the initial value of  $\Omega$ ,  $r_0$   
 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

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