Integrated characterisation of bentonite pellet structures for in situ test installation

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ABSTRACT

Bentonite pellet-based mixtures are currently investigated as candidate materials in large-scale in situ sealing experiments such as the Vertical SEALing project by the Institute for Radiological protection and Nuclear Safety (IRSN, France) due to their expected gap-filling capacity in engineered barrier systems for the geological disposal of radioactive waste. In the short term, after installation, the hydro-mechanical behaviour of these pellet-based structures is governed by pellet-pellet interactions, which have been characterised experimentally by oedometer compression tests and numerically by discrete-element modelling of single-layered pellet skeleton samples. In this work, we integrate these experimental and numerical tools to characterise the compressibility of three-layered pellet-based structures with axis-oriented arrangements. We validate the simulated deformation of bentonite pellet samples against experimental data and analyse the evolution of the dry density distribution and axial stiffness along the oedometer compression curve. Our integrated approach has implications for the emplacement and monitoring in situ tests on multi-layered pellet-based barrier systems.

Keywords: engineered barrier systems; in situ tests; bentonite pellets; discrete-element modelling.

1. Introduction

The performance of radioactive waste management solutions by containment in deep geological repositories relies on engineered barrier systems (EBS) design and placement. EBS safety is ensured by filling the technological voids and fissures around the excavation damaged zone (EDZ), thereby limiting radionuclide and fluid transport away from the disposal facilities. Bentonite-based mixtures of blocks, pellets, and granular material are considered in EBS as buffering/sealing materials for their high swelling capacity and low permeability upon saturation (Wilson et al. 2011).

For the past decades, the behaviour of bentonite-based EBS has been studied in various large-scale in situ test concepts. In the Spanish reference concept, the Full-scale Engineered Barriers EXperiment (FEBEX) investigated the disposal of waste canisters in a horizontal drift excavated in crystalline rock at the Grimsel Test Site. After dismantling, the EBS of highly-compacted bentonite blocks presented lower dry densities at the back of the gallery, associated with larger technological voids generated during block installation (Villar, Iglesias, and García-Siñeriz 2020). The Engineered Barrier (EB) emplacement experiment in the Swiss repository concept was set up inside a horizontal niche in a claystone formation at the Mont Terri rock laboratory. After dismantling, the highly compacted bentonite pellet buffer surrounding a dummy canister laid out on bentonite blocks presented some heterogeneity. Lower dry densities were observed at the bottom of the niche, which was related to the looser packing of the auger-fed pellets around the hydration components (Wieczorek et al. 2017). Within the French concept, a series of SEALing EXperiments (SEALEX) were installed in deep argillaceous rock at the Tournemire Underground Research Laboratory. In the ongoing

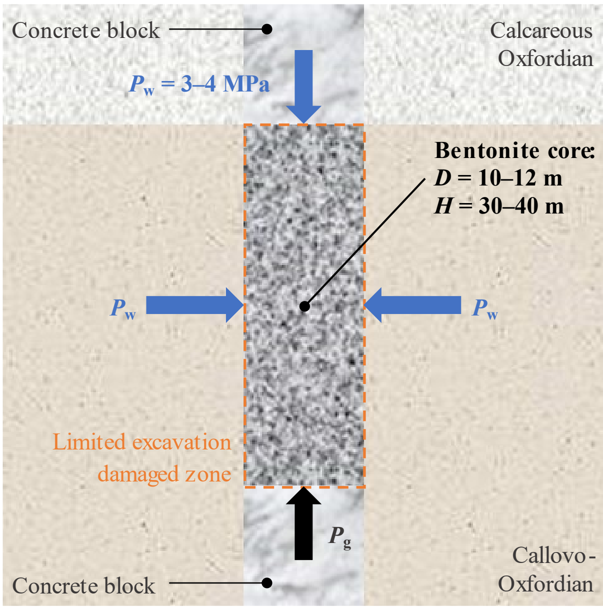

Figure 1. Diagram of the seal core in the vertical sealing system of the French concept, with annotated bentonite core diameter D and height H, and the boundary mid- to long-term water (P_w) and gas (P_g) pressure conditions. Adapted from Mesa-Alcantara (2021).

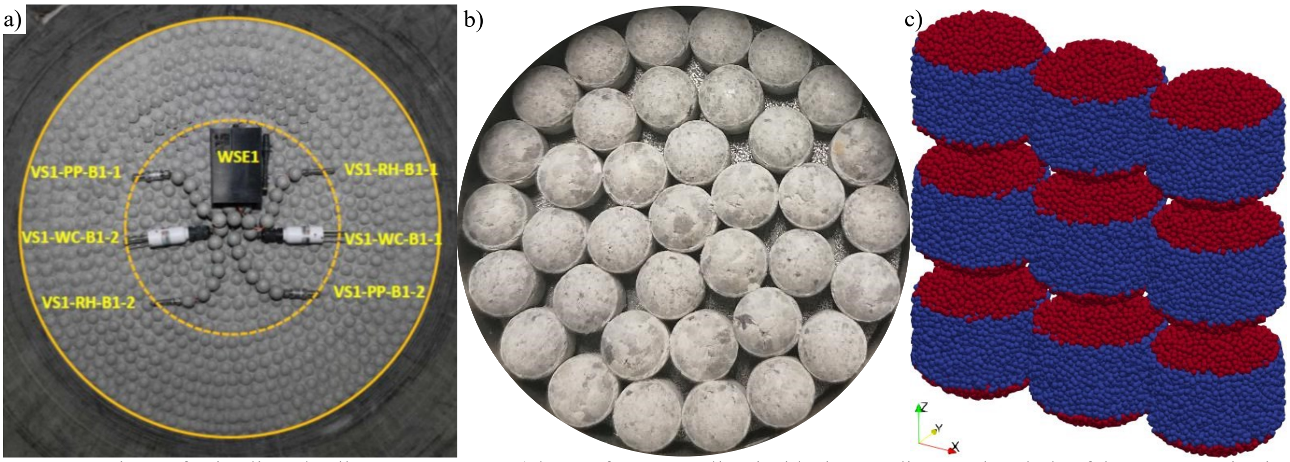

Figure 2. Views of axis-aligned pellet arrangements: a) layer of 32 mm pellets inside the 1 m diameter borehole of the VSEAL 1 in situ test instrumented with wireless sensors (Mokni, Cabrera, and Deleruyelle 2023); b) layer of 7 mm pellets closely packed in a 50 mm oedometer cell (Mesa-Alcantara et al. 2024); c) 21×7×21 mm^3 particle system modelling three layers of 7 mm pellets by DEM.

PT-N1 SEALEX test, a core of disk-shaped blocks of a pre-compacted sand-bentonite mixture (70%-30% in dry mass) was lodged in a horizontal borehole. The technological voids between blocks and the EDZ were filled with granular bentonite to reduce the establishment of preferential flow paths through the gaps. Hydro-mechanical modelling foresaw an effect of dry density variability on the long-term performance of the test (Mokni and Barnichon 2016).

Vertical sealing systems are currently under research as part of the Vertical SEALing (VSEAL) project of the French Institute for Radiological protection and Nuclear Safety (IRSN) at the Tournemire Underground Research Laboratory (Mokni, Cabrera, and Deleruyelle 2023). The VSEAL concept is intended to study gas transport in partially- and fully-saturated conditions inside surface-to-bottom seals (Fig. 1). In this context, an in situ experiment at a 1:10 scale (VSEAL 1) evaluates hydration from the top boundary of a bentonite-based core before gas injection in a vertical borehole excavated in argillite. The core comprised layers of Wyoming-type bentonite (MX-80) highly-compacted pellets and granular bentonite in 80% and 20% dry mass fractions, respectively. The layers were carefully installed, keeping an axis-oriented arrangement of the pellets. Granular bentonite obtained from crushed pellets was used to fill the inter-pellet voids. Initial structural heterogeneities were observed, resulting in a technological gap at the top, associated with an uneven distribution of the granular bentonite throughout the core layers. At five sections of the core-EDZ interface, wired sensors were distributed to monitor pore and total pressures. Wireless pore pressure, relative humidity, and water content sensors were installed at three core sections (Fig. 2a).

Laboratory-scale tests have been conducted to understand better the hydro-mechanical behaviour of the MX-80 pellets, pellet packings, and the pellet-granular bentonite mixture (Darde et al. 2018; Molinero Guerra et al. 2018; Mesa Alcantara 2021). In a recent study by Mesa-Alcantara et al. (2024), the compressibility on laterally confined loading of pellet-supported structures was investigated on single-layered samples of axis- and orthogonally-oriented pellets at constant initial coordination number of pellet contacts and dry density conditions (Fig. 2b). The effects of pellet anisotropy and potential heterogeneous granular bentonite distribution—with and without filling—were assessed experimentally.

Innovative numerical modelling strategies have also been applied to the in-depth characterisation of pellet-supported heterogeneous structures after installation and before long-term homogenisation. A novel discrete particle method developed by Navarro et al. (2023) to simulate granular bentonite hydration was successfully adapted to 2D axisymmetric modelling of the progressive transition of pellet-granular bentonite mixture from the pellet-controlled to homogenised behaviour (Navarro et al. 2024). Darde et al. (2020) implemented a mixed discrete-finite element model to reproduce the latter experimental results. Yang et al. (2024) modelled the compacted mixture behaviour with a continuum-based triple porosity model including damage. Torres-Serra, Romero, and Navarro (2023) introduced a discrete element method (DEM) model of pellet compressibility, later applied to capturing the anisotropic deformation of well-oriented pellet-based mixtures in Mesa-Alcantara et al. (2024).

In this paper, we present an integrated experimental and numerical approach to characterising the compressibility of multi-layered MX-80 bentonite pellet-supported structures. We perform oedometer compression testing and DEM modelling of three-layered samples. The results are then compared to single-layer compressibility and discussed in terms of the dry density distribution and mechanical behaviour.

2. Materials and methods

2.1. Oedometer testing

Commercially manufactured, high-density MX-80 pellets are tested, which have an axisymmetric geometry consisting of a cylindrical core with two spherical caps at the top and bottom, and size 7 mm (EXPANGEL®SP7, Laviosa-MPC, France). The pellets are produced by uniaxial compaction from bentonite powder of solid density $\rho_s = 2.77$ Mg/m^3. We prepare a sample comprising three layers of axis-oriented pellets with a total height of 21 mm and measured initial water content $w = 7.6\%$ and total suction $s = 114$ MPa. Oedometer testing is performed in a cylindrical cell of diameter 50 mm where, for each layer, pellets are arranged in three concentric rings around a central pellet (Fig. 2b). The prepared sample's dry density $\rho_d = 1.20$ Mg/m^3 corresponds to a close random packing of initial void ratio $e_0 = 1.31$. The cell allows vertical loading of the pellet packing at constant water content and atmospheric air pressure conditions. We apply successive vertical stress increments, ensuring the vertical strain rate after the previously applied vertical stress is smaller than 0.01%/day at each step.

2.2. DEM numerical model

We model pellets as deformable clumps made of random packings of spheres of radius $r = 0.175$ mm. Pellet anisotropy is included in terms of the density distribution, where the core is observed to be denser than the caps (Fig. 3), related to cap fissuring due to high-stress loading-unloading undergone during the production process. We approximate the dry density field over a domain $\Omega = l_x \times l_y \times l_z$ by

$$\rho_d(\mathbf{x}) = \sum_{i\in\Omega} m_i \Phi_i(\mathbf{x}) \quad (1)$$

where m_i is the i-th particle mass, and Φ_i is the Gaussian function centred on the i-th particle's location $\mathbf{x}_i$ of width $3r$. The modelled pellet presents averaged dry densities $\rho_d = 2.04$ and 1.93 Mg/m^3 of the core and caps, respectively.

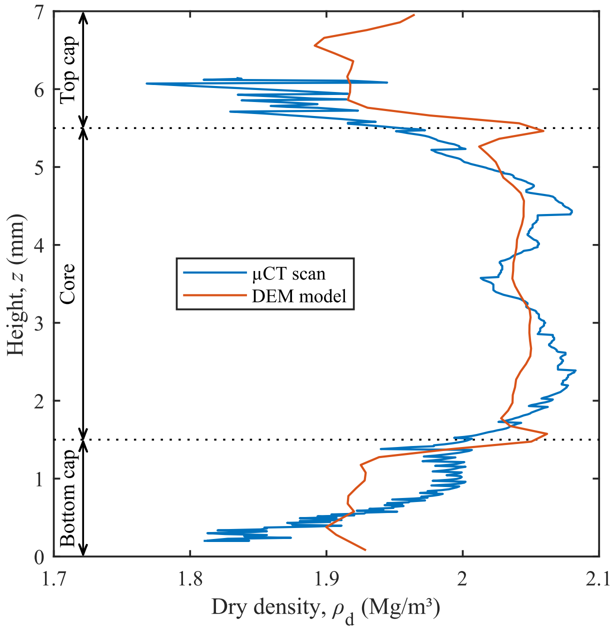

Figure 3. Experimental and numerical dry density profiles of a pellet. X-ray computed microtomography (µCT) data from Mesa-Alcantara et al. (2024).

We use the cohesive particle model available in the open-source code Yade (Šmilauer et al. 2021), providing a spring-slider contact force model controlled by a damage evolution law (Šmilauer 2010). The mechanical parameters in Table 1 are calibrated for the single pellet model introduced by Mesa-Alcantara et al. (2024). Anisotropy is also included with respect to the different Young's moduli E of the pellet core and caps as a function of their void ratio e, estimated with a bilinear model fitting experimental data on oedometer compressibility of binary pellet and granular bentonite mixtures.

Table 1. Cohesive particle model parameters (Mesa-Alcantara et al. 2024).

Parameter	Value
Young's modulus, E (MPa)	110 (core) / 48 (caps)
Poisson's ratio, ν (-)	0.24
Friction coefficient, μ (°)	0.33
Shear cohesion, C_0 (MPa)	1.0
Elastic limit, ε_0 (-)	2.0×10^{-3}
Fracture strain, ε_f (-)	8.0

The simulation domain in Fig. 2c, consisting of a 3×1×3 configuration of axis-oriented pellets, is chosen as a pseudo-axisymmetric representation of the three-layered sample tested in the oedometer cell. The three-pellet width matches the number of concentric pellet rings in the experimental arrangement described above. The particle system is enclosed by flat orthogonal walls with the same mechanical contact parameters as the particles. We vertically compress the sample at a constant speed of 0.35 m/s down to the target vertical strains $\varepsilon_z = 2$, 4, 8, 12, 20, and 25 (%), at which points the compression is halted, and the system is allowed to relax until equilibrium (defined as when the dimensionless ratio of mean resultant force magnitude acting on the particles over the mean contact force magnitude becomes smaller than 1×10^{-5}). Before resuming the compression steps, the vertical stress-strain values attained at equilibrium are incorporated into the compression curve.

3. Results and discussion

The proposed DEM model allows us to track the material packing and dry density during compression. The initial ($\varepsilon_z = 0\%$) vertical section on the axial plane $y = 3.65$ mm shows the well-arranged configuration of axis-oriented pellets with differentiated core and cap regions (Fig. 4a). The initial dry density averaged over a regular grid of resolution 0.1×0.1 mm^2 and along the sample depth ($l_y = 7$ mm) is higher around the pellet cores, as expected (Fig. 4b). At the final loading stage ($\varepsilon_z \approx 25\%$), the loss of the original pellet geometry is mainly attributed to cap deformation. Interestingly, we observe a degradation mechanism by which the cap material can deform the pellet core around the pellet's axis, whereas the initial core height is preserved around the pellet boundary. This effect is predominant in the top layer of the sample (Fig. 4c). The final dry density distribution presents a high densification of the top and bottom layers' caps in contact with the walls (Fig. 4d). We also note high-density values at the pellet-pellet contact regions, more between the middle and top layers. On the contrary, the core material around the pellets' axes is less dense than in the initial configuration, whereas the vertical pellet-pellet contact regions keep the original density distribution, according to the degradation mechanism.

Regarding the mechanical behaviour on loading, the compression curves of the following tested samples are depicted in Fig. 5:

- OED1: Oedometer test on one layer of pellets (Mesa-Alcantara et al. 2024);

- DEM1: DEM model of a single pellet (Mesa-Alcantara et al. 2024);
- OED3: Oedometer test on three layers of pellets;
- DEM3: DEM model of three layers of pellets.

The void ratio e is calculated from the vertical strain ε_z, equivalent in our test set-up to the volumetric strain, by

$$e(\varepsilon_z) = e_0 - \varepsilon_z(1 + e_0) \quad (2)$$

where e_0 is the initial void ratio measured experimentally and adopted as a reference value.

We estimate the initial ($\varepsilon_z = 0\%$) and final ($\varepsilon_z \approx 25\%$) slopes of the compression curves from a linear regression through the first and last data points, ($\ln\sigma_z$, e). Across all samples, two loading stages are identified: an initial stage attributed to the compression of the pellets, followed by a final stage of pellet degradation up to the final configuration in Figures 4c and 4d. The yield stress σ_z^* marking the transition from the initial to final loading stages is estimated at the intersection of the respective regression lines (Table 2). Compared to the one-layered case (OED1), the three-layered oedometer test (OED3) is more compressible at lower stress states and stiffens towards the final loading stage. The yield stress is smaller for OED3, suggesting the reduced strength of the multi-layered sample. The DEM model successfully captures the two differentiated loading stages, though overestimating the initial stiffness of the samples, which is attributed to the simplified model features with only six mechanical parameters and also to the idealised pellet geometry and arrangement configurations with respect to the experimental tests.

Table 2. Compressibility parameters from experimental and numerical data.

Test	Compression curve slope, $-\Delta e/\Delta\ln\sigma_z$ (-)		Yield stress, σ_z^* (MPa)
	Initial	Final	
OED1	0.049	0.35	1.5
DEM1	0.047	0.40	1.8
OED3	0.074	0.26	1.0
DEM3	0.053	0.32	1.1

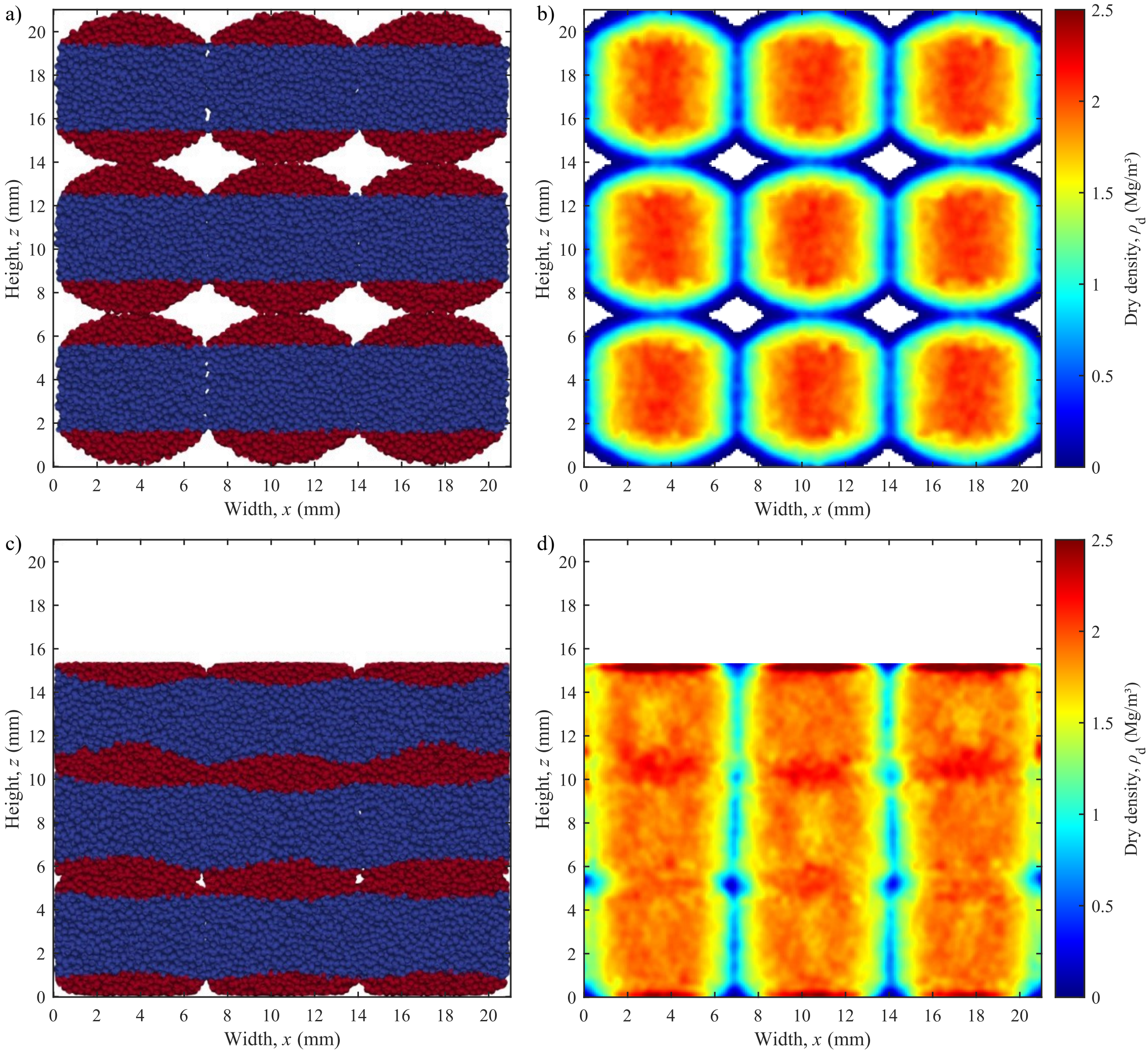

Figure 4. Packing evolution during the DEM simulation: a) initial vertical section; b) initial averaged dry density field; c) final vertical section; d) final averaged dry density field.

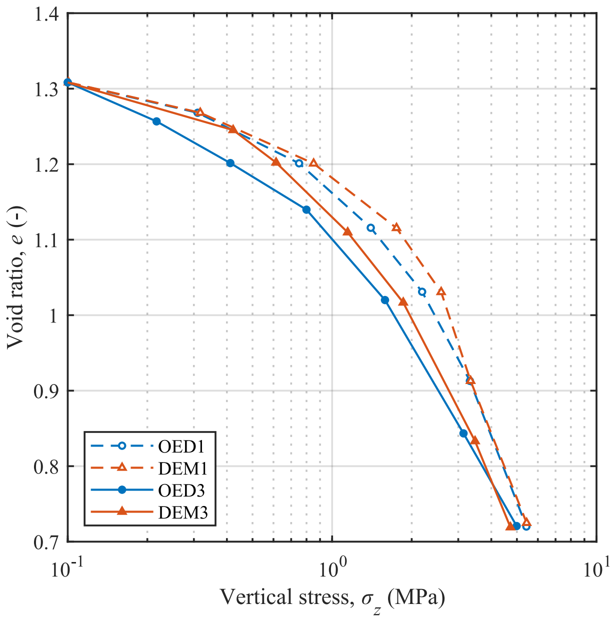

Figure 5. Experimental and numerical compression curves upon loading of one- and three-layered axis-aligned pellet arrangements.

4. Conclusion

We used an integrated approach to characterise the behaviour of MX-80 bentonite pellet-based structures in the short- to mid-term after installation as a buffer material in the large-scale vertical sealing system in situ tests by laboratory-scale experimental and numerical tests.

The laterally confined compression of axis-oriented pellet arrangements produces a heterogeneous density distribution of the bentonite material related to the degradation of the pellet-pellet contact regions between pellet caps. As a result, lower dry densities are observed at the vertical contact regions between pellet cores, where preferential flow paths could form. The use of granular bentonite filling inter-pellet voids is thereby justified. The experimental and numerical three-layered samples showed larger compressibility than previous single-layered tests. The DEM model simulated an overall stiffer behaviour attributed to the tested pellet-based skeletons' idealised geometric and confinement conditions.

Our findings are relevant to installing and monitoring in situ experiments, where technological voids created during construction have been observed to impact the long-term performance of bentonite-based sealing systems.

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