Overview and conceptual constitutive framework for pellet-based buffer materials

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Abstract. Buffer materials for nuclear waste disposal applications generally consist of blocks made of highly expansive compacted clay. However, high-density pellets of bentonite are being evaluated as an alternative buffer material for waste isolation. The material response of pellet-based buffers may be quite different from that of compacted buffers, because of the peculiar discontinuous porosity presented. An overview of the literature available on pellet-based buffers is presented and, in particular, two main topics are discussed: firstly, the characteristics of the fabric of the pellets that can be observed through techniques of micro-structural investigation, secondly, the most important behavioural features that can be seen during material testing. Additionally, the constitutive frameworks that have already been developed specifically for pellets are also reviewed. The overall objective of the paper is to highlight the differences between compacted and pellet-based bentonite buffers, in order to propose suitable assumptions to start developing a constitutive model for the latter.

1 Introduction

The investigation into buffer materials, which are employed as seals in deep geological repositories for the long-term storage of nuclear waste, is an important research topic. Currently, the most widely accepted solution is that buffers are made of compacted blocks of bentonite. Nevertheless, in order to facilitate the installation of this layer of the engineered barrier that isolates the radioactive material, pellets are being considered as an alternative to blocks. A pellet can be of different shapes (cylindrical, pillow-shaped, or irregularly-shaped), with dimensions that are less than 10 mm, and it is obtained through compaction at high pressure. Consequently, it has a very high density, a high swelling potential and a low permeability. Pellet-based barriers can have different fabrics. They can be made of only pellets or they can be pellet-powder mixtures or simply compacted powder obtained by crushing pellets. The objective of this paper is to review the major findings on these materials in order to highlight their most important hydro-mechanical features. A comparison between pellets and blocks can then be established. Since the knowledge on the latter is relatively more advanced than that on pellets, this parallel can constitute a solid theoretical base on which ideas about a numerical framework for pellets can be developed. The materials discussed in this paper are all different types of bentonites that have a large swelling potential due to their mineralogical composition, which comprises a large percentage of montmorillonite.

2 Observed behaviour of pellets and pellet-based materials

2.1 Fabric

Pellet samples present a multi-modal porous structure ([1], [2]), which can be observed using techniques such as Mercury Intrusion Porosimetry (MIP) and X-ray computed microtomography. A single pellet exhibits a double porosity structure ([1], [2], [3]), which is the result of the compaction on the dry side of optimum moisture content that is employed in the production process. Figure 1(b) from [1] presents the bimodal Pore Size Distribution (PSD) curve of a single, highly compacted pellet (ρ_d = 1.95 Mg/m^3) with characteristic pore sizes of 13 nm and 3 μm. The smallest pores are deemed intra-aggregate pores, while the larger pores are deemed inter-aggregate-pores. A qualitatively similar result was obtained by [3], who analysed a single pellet (ρ_d = 2.06 Mg/m^3, suction 135 MPa) of MX-80 bentonite. Interestingly, the inter-aggregate porosity (with an average diameter around 4-5 μm) was found to represent barely 7% of the total porosity. Figure 1(a) shows the PSD of a sample made of pellets prepared using a gravity fall compaction procedure (ρ_d = 1.15 Mg/m^3), which is a three-mode curve with characteristic pore sizes of 13 nm, 3 μm and 250 μm ([1]). The largest pore group represents the inter-pellet spaces. Even larger voids are not uncommon in pellet samples, due to irregular packing, but they cannot be detected with MIP,

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as the largest detectable size is 500 μm ([2]). Furthermore, the maximum void ratio intruded in MIP investigations is less than the total void ratio, because heavily compacted pellets present a so-called infra-porosity ([4]), which is made of small pores with an entrance diameter smaller than 5.4 nm.
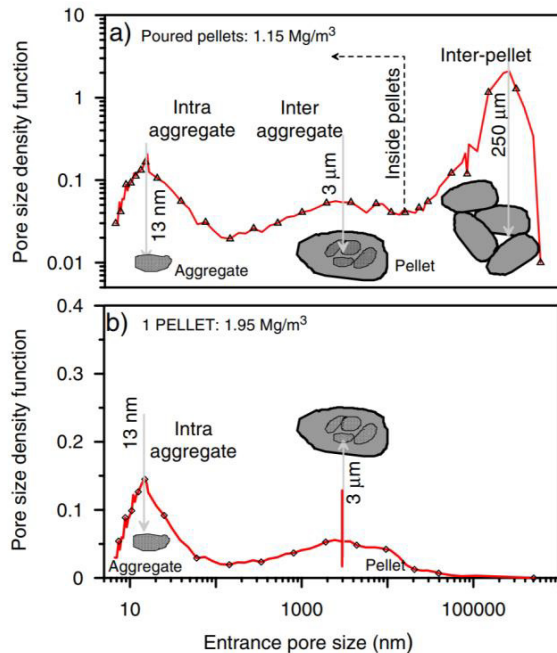

Fig. 1. PSD of FEBEX bentonite in: a) a compacted sample, and b) a single pellet (modified after [1]).

Following compaction, pellets can contain cracks at the scale of the inter-aggregate porosity. These can be shown through X-ray computed microtomography ([3], [5]), as depicted in Figure 2. Crushed pellets can also be partly reduced to powder ([1]). This results in a decreased inter-pellet void space and an increased inter-aggregate void space. While examining a pellet-powder bentonite mixture, [6] showed a downward movement of the powder, which was induced by gravity, towards large air-filled inter-pellet voids. It can be concluded that the larger peak of a PSD measured on a single pellet is influenced by the phenomena of cracking and crushing. Meanwhile, the smaller peak depends on the inter-layer distance between clay platelets present in every aggregate. X-ray micro-tomography allows the quantification of the evolution of this distance with suction ([5]). Upon wetting, new layers of water are formed in-between platelets and particles tend to be formed by less and less platelets. For this reason, at very low suctions the fabric of pellets presents a high level of disorder.

Pellet samples are heterogenous at the as-compacted state. This heterogeneity is reflected both in the spatial distribution of pellets within the specimen and in each pellet, which contains inclusions, usually made of quartz or pyrite ([6]), that may be of a comparable size with respect to the dimensions of the pellet itself. Figure 3 shows the appearance of a pellet-powder mixture

obtained through X-ray micro-tomography ([6]). When employed as backfill material in the boreholes of deep geological repositories for the long-term storage of nuclear waste, pellets allow for a simpler installation, but care must be taken to avoid segregation ([1]).

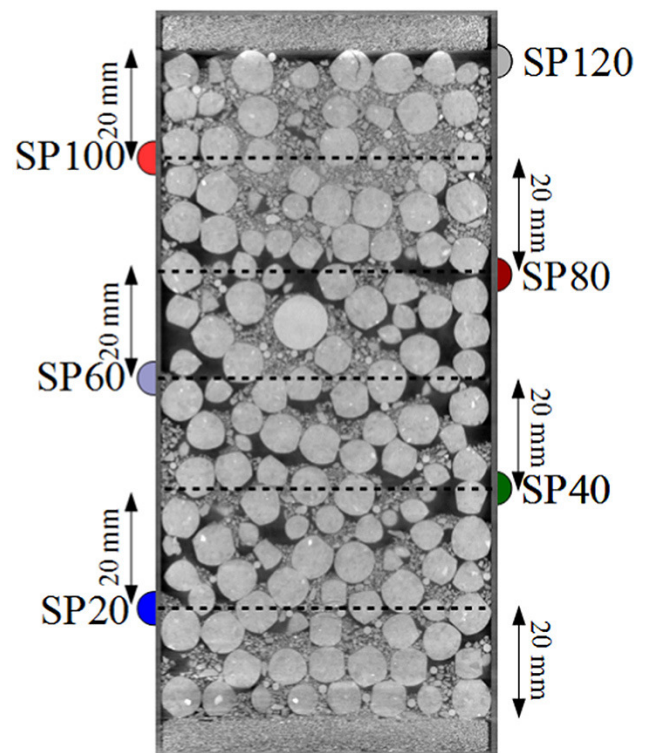**Fig. 2.** Cracks in a compacted pellet, captured with X-ray computed microtomography (after [3]).

Fig. 3. Vertical section of a pellet-powder mixture (after [6]).

2.2 Evolution upon hydration

Upon wetting, inter-pellet porosity is quickly filled by the expansion of the hydrating pellets. [7] showed that once full saturation is reached, a 50/50% FoCa bentonite pellet/powder mixture appears as homogeneous, and its swelling pressure is equal to that of a compacted powder sample of the same dry density. [8] used X-ray computed tomography to show the observed evolution of the distribution of the dry density upon hydration along a section of a 50/50 powder-pellet mixture of FoCa clay sample initially compacted to 1.36 Mg/m^3. During hydration, the initially higher density of pellets decreases due to swelling and the initially lower density of powder increases due to soil compression and water content increase. [6] and [8] both noted that a pellet-powder mixture showed a relatively homogeneous structure even prior to full saturation, after several months of hydration. Figure 4 depicts the evolution of the transverse section of the sample during hydration obtained by X-ray computed microtomography.
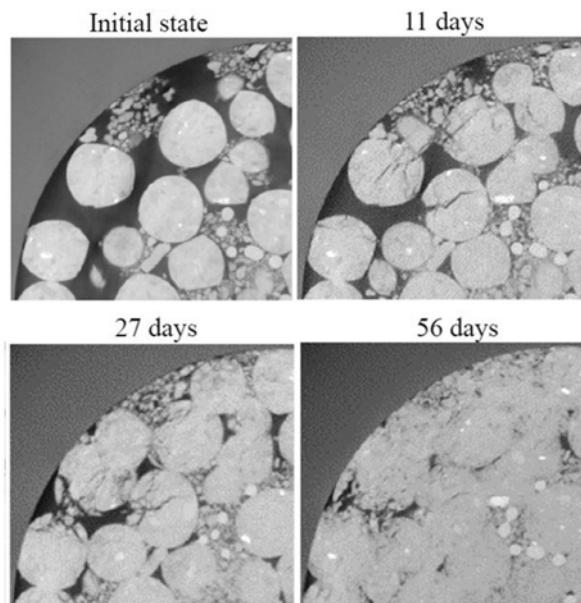

Fig. 4. Evolution upon wetting of a pellet-powder bentonite sample (after [6]).

[5] investigated the change of the micro-structure of an MX-80 pellet compacted to a dry density of $\rho_d = 2.06$ Mg/m^3 upon hydration under free swelling conditions. From an initial value of 135 MPa, suction was decreased until 0.8 MPa and MIP tests were carried out at different stages. The results of these tests are reported in Figure 5, where the intermediate states are identified as P1 (beginning of wetting) to P5 (end of wetting). It has not been possible to impose 0 MPa of suction to the pellet because at this state it was not far from a slurry. The compacted pellet at its initial as-compacted state only presented moderately sized cracks close to its perimeter. At high suctions, i.e. above 82 MPa (Figure 5b), no significant swelling was observed and the tiny changes in the micro-structure are linked to the mechanism of

water adsorption. From 82 to 38 MPa of suction (Figures 5c and 5d), swelling due to the development of pores in the 10-40 µm range is observed. At lower suctions, the diameter of the inter-aggregate pores tends to increase, as it can be seen in Figure 5(f), meanwhile intra-aggregates pores remain mostly unchanged. At this stage, an increasingly interconnected crack network propagates towards the middle of the pellet, as it can be observed through X-ray microtomography featured in [5]. It is thought that this crack network constitutes a preferential path for the propagation of vapour. Based on information gathered by combining MIP and X-ray microtomography investigations, [5] argued that the swelling of a pellet along a wetting path is a combination of crack propagation and grain expansion and that most of the swelling takes place at low suctions. However, it should be noted that, in samples made of multiple pellets, these are subjected to free swelling conditions only as long as the inter-pellet space has not been filled up. Through an MIP investigation on a pellet-powder mixture that was hydrated under constant volume conditions, [9] showed the presence of a dominating family of pores in the range of 200-500 nm in diameter. This was justified considering the reduction in size of the macro-pores to accommodate the increase in micro-pore volume due to hydration. The non-intruded porosity was also shown to be high.
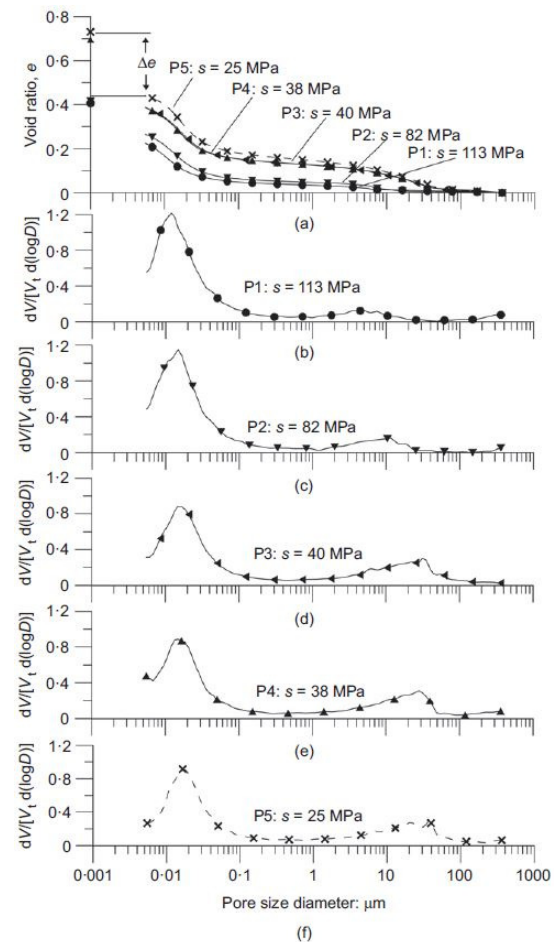

Fig. 5. MIP evolution of a compacted pellet upon hydration under free swelling conditions (after [5]).

2.3 Hydraulic properties

[5] determined the water-retention properties of an MX-80 pellet under free swelling conditions, noting that most of the swelling took place at low suctions, when the water content increased considerably. [1] measured the retention properties of several pellet samples compacted to different dry densities, as shown in Figure 6. The dry density only influences the water uptake below a certain value of suction. This threshold corresponds to the state in which free swelling conditions no longer apply to single pellets because the inter-pellet space has been almost entirely closed, which happens when the degree of saturation is in the 62%-75% range ([1]). Closer to saturation, looser pellet samples can retain a larger quantity of water with respect to denser samples, as expected. [1] studied the evolution of permeability upon hydration in pellet samples. Unsurprisingly, the occlusion of inter-pellet pores coincides with the drastic reduction of permeability, as shown in Figure 7. As for compacted clay samples, the value of permeability at saturation is controlled by the dry density.

In the case of pellet samples, the magnitude of the infiltration rate to which the sample is subjected influences the mechanical response ([1]). Fast infiltration, which is usually obtained circulating liquid directly into the specimen, causes an immediate collapse in the fabric because the water fills rapidly the inter-pellet pores, causing the intergranular forces to decrease. Subsequently, the exchange of water between inter-pellet and inter-aggregate pores begins and the swelling of pellets takes place. Eventually, the swelling overcomes the initial collapse and the overall strain of the sample is expansive. This phenomenon can be interpreted considering two water potentials, one of the macro-structure, i.e. of the specimen, and one of the micro-structure, i.e. of the single pellet, that are equalizing. On the other hand, slow infiltration, which is obtained using a vapour transfer technique, triggers swelling from the beginning of hydration because the water goes directly into the inter-aggregate pores and thus the intergranular forces reduce less rapidly than under fast infiltration.
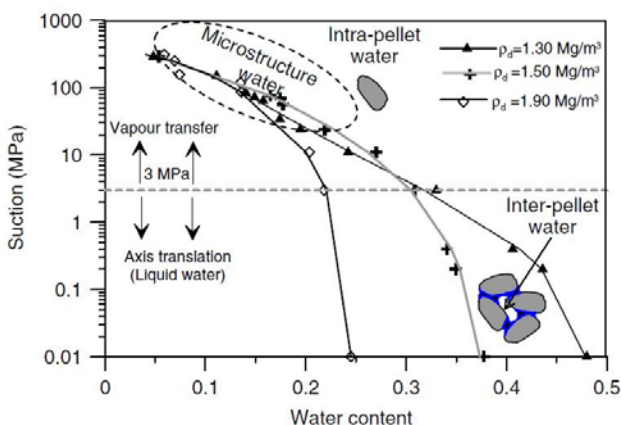
Fig. 6. Water retention curves upon wetting of pellet samples compacted to different dry densities (after [1]).
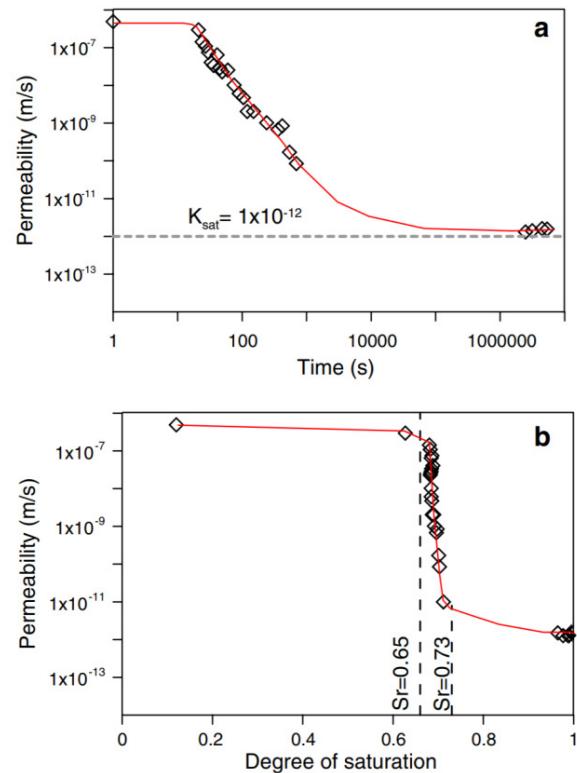
Fig. 7. Evolution of permeability with the degree of saturation (after [1]).

2.4 Testing results

[2] investigated the evolution of the fabric of pellet samples upon compaction. The inter-pellet pores were found to be dependent on the compaction stress, meanwhile the intra-aggregate pores were mostly independent of it ([1]). At very high compaction, inter-pellet and inter-aggregate pores tended to coincide.

[7] carried out several infiltration tests on samples of FoCa bentonite pellet with the scope of measuring the swelling pressure. They found a recurring pattern of stress development with time that seems to be repeated irrespectively of the initial dry density of the material, of the size of the sample and of the fabric, which was either powder or a pellet/powder mixture. Initially, the swelling pressure grows rapidly towards an intermediate peak. Subsequently, a collapse takes place, reducing the stress momentarily. Finally, the swelling pressure increases again and eventually settles on a stable value. Figure 8 reports the evolution of the stress measured in powder samples compacted to different dry densities ([7]). [9] observed that the length of the specimen only influenced the rate of development of the swelling pressure. In light of the considerations outlined in the previous sections of this paper, the interpretation of this pattern is that the collapse, i.e. the diminution of the swelling pressure, coincides with the disappearance of the macro-structural inter-pellet pores. This explains why samples with different dry densities all experience a collapse. However, it was found that high density samples (dry density around $\rho_d = 1.6$ Mg/m3), presented an initial stress peak that was lower than the final stress value.

Meanwhile, low density samples (dry density around ρd = 1.3 Mg/m3) did the opposite, as it can be noted in Figure 8. As in compacted blocks, the relationship between the final swelling pressure and the dry density is exponential and the initial water content does not influence the swelling potential ([1]). Another similarity between blocks and pellets is that the swelling strain depends linearly on the logarithm of the confining vertical stress, as demonstrated by [1] with a series of wetting under constant loading tests.

Pellet samples exhibit a significant anisotropy ([2], [5]). A single pellet that has swollen under unrestrained conditions presents larger axial (εa) than radial strains (εr). [5] have found that the ratio $\varepsilon a / \varepsilon r$ is equal to 6.1 when the pellet has reached 38 MPa of suction. This ratio decreases as the wetting progresses but never attains unity. In some cases, the anisotropy is thought to be induced by the production of pellets that are obtained through one-dimensional compaction in a mould that causes an increase of axial stress without any radial stress. This causes the clay particles to have a preferential sub-horizontal orientation, hence the enhancement of the swelling in the direction that is perpendicular to the clay platelets.
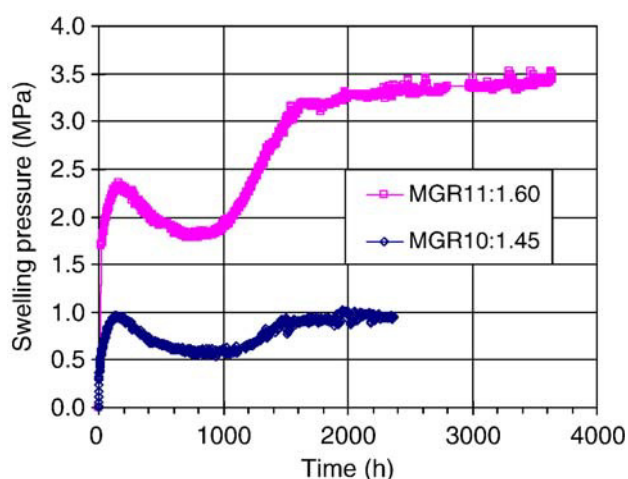

Fig. 8. Evolution of swelling pressure in infiltration tests carried out on powder samples compacted to different dry densities (after [7]).

3 Comparison with compacted clays and existing modelling approaches

Using the information gathered in Section 2, it is possible to draw a comparison between pellets and compacted blocks made of bentonite clays. First, pellets are heterogeneous. Single pellets may contain inclusions that are comparable in size to their dimension, and pellet samples are always spatially irregular because their fabric is highly dependent on the preparation process, which is why whenever pellets are employed in nuclear waste disposal it is fundamental to avoid segregation in the installation phase. While [10] have included heterogeneity in their framework, several sources ([7], [3]) have pointed out that a final, quasi-homogeneous

state is obtained following wetting of a pellet sample. However, [2] highlighted that it is particularly important to capture the transient state of the specimen, which involves significant local changes in the fabric. In order to do so, they introduced a physically based, double-porosity 1D model that describes the behaviour of compacted clay pellets using finite differences. The model considers an equivalent porous medium made of 1D pellets, i.e. the micro-structure, that are connected to form the macro-structure. No interactions between microstructural parts of adjacent elements can take place. Consequently, water flows through the inter-connected macro-pores and the pellets are hydrated through a transfer of water from the macro-structure to the micro-structure. Aside from assigning different void ratios to each level of structure, [2] also distinguish two complementary water potential fields. This implies that the model is formulated using two suctions variables. This point differentiates pellet samples from block samples. In fact, the latter are widely represented with double structure models where micro- and macro-structures are hydraulically in equilibrium ([11], [12], [13]). [9] enhanced the framework by [13] in order to account for the exchange of water between different levels of structure and satisfactorily reproduced the hydro-mechanical behaviour of a pellet-powder mixture both at the laboratory scale and at the field scale, in the context of long-term sealing test where the mixture was used as engineered barrier for nuclear waste disposal. Furthermore, the framework by [2] was able to simulate the progressive loss of permeability during wetting, which coincides with the disappearance of inter-pellet spaces, and the effects of different infiltration rates. [14] introduced a discrete "two-permeability" model in order to account for the increase of permeability with the degree of saturation. Characterising the hydraulic permeability of pellet samples is of paramount importance, because its value spans over several orders of magnitude during wetting. Furthermore, the interconnected crack network that forms in pellets when suction goes below 40 MPa constitutes a preferential path for both water and vapour. The latter yields effects that still have to be fully investigated and understood.

4 Discussion and Conclusions

Double porosity structure models for unsaturated compacted blocks cannot capture the full behaviour of pellet-based materials. For example, the Imperial College Double Structure Model (ICDSM, [11], [12]) accounts for the evolution of the fabric upon wetting only after the first full saturation, when MIP investigations have revealed that the PSD becomes mono-modal. Given the information gathered in this literature review, it is possible to conclude that the fabric of pellet-based materials evolves well in advance of full saturation, causing significant changes especially in the hydraulic properties. Furthermore, it is unclear whether the PSD of a fully saturated pellet is mono-modal, as it is difficult to run tests at very low suctions when the pellet resembles a slurry. It might be reasonable to employ

double structure models developed for compacted blocks to capture the final state of pellet-based materials. In terms of swelling pressure, it has been shown that the measured values are similar, and similarly related to the dry density, for pellets and blocks. However, the phenomenology that allows the stress to develop in pellets cannot be captured by double structure models developed for compacted blocks.

Cracking during both hydraulic and mechanical loading seems very relevant in pellets, while it is usually neglected in modelling compacted blocks, where the dimensions of the sample are always several orders of magnitude larger than the cracks. It would be of great interest to investigate how vapour is transferred through the macro-structure of pellet materials and how it affects the micro-structure, i.e. the single pellets. It seems necessary for a numerical framework developed for pellets to take into account the exchange of water from the inter-pellet space to the inter-aggregate space. This can only be accomplished using multiple suction variables, as some existing models have already done.

Finally, heterogeneity is another aspect that distinguishes blocks from pellets. It is unclear to what extent the overall behaviour of the latter is influenced by this factor. On the other hand, studies have shown that the compaction process causes pellets to behave in an anisotropic manner. In particular, the swelling develops preferentially in the direction that is parallel to the applied compaction stress. Similar observations have been made for compacted blocks, however the available data is not sufficient to properly characterise the anisotropy.

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