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Grain-scale modelling of swelling granular materials; application to super absorbent polymers

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ABSTRACT

Swelling is an important process in many natural materials and industrial products, such as swelling clays, paper, and Super Absorbent Polymer (SAP) particles in hygienic products. SAP particles are capable to absorb large amounts of fluid. Each grain of SAP can absorb water 30 to 1000 times its initial mass, depending on the water composition.

To gain insight in the swelling behaviour of a bed of SAP particles, we have developed a grain-scale model and have tested it by comparing it to experiments. The grain-scale model is based on a combination of the Discrete Element Method (DEM) and the Pore Finite Volume (PFV) method, which we have extended to account for the swelling of individual SAP particles. Using this model, we can simulate the behaviour of individual particles inside a water-saturated bed of swelling SAP particles while taking into account the hydro-mechanical effect arising from the presence of pore water. The model input includes physical parameters such as particle stiffness and friction angle, which were found in the literature, as well as particle size distribution and diffusion coefficients, which were measured experimentally. A swelling rate equation was developed to simulate the swelling of individual particles based on water diffusion into a spherical particle. We performed experiments to measure the rise of the surface of a bed of initially dry SAP particles, which were put inside a glass beaker that contained sufficient amount of water for the SAP particles to swell and to remain saturated at all times. We used our model to simulate the swelling of that SAP particle bed as a function of time. Simulations show that the numerical model is in accordance with the experimental data. We have also verified the model with Terzaghi's analytical solution for a small swelling event. Finally, a sensitivity analysis was performed to study the effects of main grain-scale parameters on the larger-scale behaviour of a bed of particles.

1. Introduction

Swelling of porous media occurs in many natural and industrial materials e.g. clays, foods, biological tissues, papers, and absorbent polymers in hygienic products [1–5]. Swelling is a complex process that requires knowledge of the behaviour of both fluid and solid phases and their physical interactions, as well as the chemical interactions inside and around the solid phase [6,7]. The process of swelling is defined as the expansion of a (porous) solid because of absorption of a fluid. A distinction can be made between swelling solids e.g. cartilages, hydrogels, Super Absorbent Polymers (SAP), and swelling granular media, such as a bed of SAP particles. In this work, we focus on irregularly shaped SAP particles, whose size ranges from 45 to 850 μm and are mostly used as absorbent agent in hygienic products.

During the design of SAP, the initial chemical composition determines its stiffness, absorption rate and absorption capacity [8–10]. SAP consists of long hydrophilic polymer chains (e.g. acrylic acid) which expand and bind with water during absorption. In order to prevent these polymers from dissolving into water, other polymers are used to bind them, which are referred to as cross linkers. A larger cross-linker concentration increases the stiffness of a SAP particle, but at the same time reduces the maximum amount of water that it can absorb *e.g.* [10]. In hygienic products, the stiffness is essential as it increases the strength of a particle bed and thus allows the bed to have better flow properties [9]. However, there is an optimum number of cross-linkers that can be added as they decrease the absorption capacity.

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Physical properties not only depend on the polymer chemistry of SAP, but also on the production process which affects the shapes and sizes of particles. Particle size affects the swelling rate; the smaller the particle, the larger the surface area to volume ratio, and thus the higher the initial absorption rate [11]. In lab-controlled experiments, perfect spherical particles may be generated [12,13], which are useful for research purposes. However, in many production processes SAP particles are produced by grinding SAP slabs, resulting in angular particles with a wide range of sizes *e.g* [9,14,15]. Angular particles thus have two main features compared to spherical particles: they have a larger surface area to volume ratio and they are less likely to move to a packing [14]. Particle shapes also significantly affect the porosity and permeability [16] as well as the mechanical strength [17] of a bed of particles.

Numerous processes occur in a bed of swelling SAP particles. These include: flow of water into the particle bed, absorption of water by the particles, and rearrangement of particles due to swelling and deformation. The swelling of a bed of SAP particles is a highly coupled process involving both fluid and solid phases. This is often referred to as hydro-mechanical coupling [18]. The hydro-mechanical coupling in a particle bed is usually studied using macro-scale experiments and simulations *e.g* [1]., where permeability and stiffness of a bed are important parameters, see for example [19]. However, grain-scale effects such as particle-particle friction [20], particle stiffness, and grain shape govern those macro-scale parameters. Experiments that aim to investigate the effect of grain-scale properties on macro-scale parameters are time consuming, difficult, and sometimes not feasible. An alternative is to develop a pore-scale or grain-scale model and use it alongside experimental investigations.

Perhaps the most suitable method for describing the motion of particles during swelling is the Discrete Element Method (DEM). This method allows simulations of the movement of individual grains in a granular porous medium [21] for example in soils [22,23], rocks [24], and industrial applications such as particle flow in silos [25]. The grains are considered as discrete elements that can undergo four relative motions at their contact points: sliding, rolling, spinning, and twisting. DEM has been mainly used for qualitative studies, but recent advancements have shown its capability for quantitative simulations within both the elastic and plastic regimes of deformation [22,26]. Various methods exist to extend DEM and include saturated and/or unsaturated flow. For example, DEM has been coupled to pore network models to simulate 2-dimensional fluid injection into granular materials under saturated conditions [27] and unsaturated conditions [28], but also to simulate 3-dimensional drying of granular materials [29], where the pore structure was assumed to be fixed while the particles in DEM could move. DEM has also been coupled to Lattice Boltzman [30] and to a finite difference scheme for fluid flow on a fixed grid [31]. In those previous works, simplifications were made to enable simulations of flow in DEM, namely: mono-dispersed packings rather than normally distributed packings, 2-dimensional rather than 3-dimensional, small deformations rather than large deformations. Recently, the pore finite volume (PFV) method was introduced by Chareyre et al. [32] and Catalano et al. [33] into DEM for simulating hydro mechanical coupling under saturated conditions. PFV is an efficient scheme for simulating 3-dimensional flow in the pore-space of packings of spheres (having a particle size distribution) accompanied by large deformations [34]. Tong et al. [35] found that the permeability values, which were predicted by the DEM-PFV model for packings of spheres, were in good agreement with experiments.

In those studies, however, absorption of water and swelling of particles were not included. Therefore, the possibility of large swelling of particles has been added to DEM in order to enable simulations of swelling and the corresponding deformation of a packing of particles. The aim is twofold i) to investigate whether DEM-PFV is capable to reproduce experiments of a bed of swelling particles, and ii) to study the effect of mechanical parameters on the swelling behaviour of a bed of SAP particles.

In this paper, we first describe the Discrete Element Method (DEM) and the Pore Finite Volume (PFV) model and how we included swelling of individual particles. Then, we discuss our experimental procedures and the model setup. Finally, we use our numerical model to simulate and reproduce experimental data. The model is then used for performing sensitivity analysis on how changes in various particle-scale properties would affect the macro-scale behaviour of a bed of particles.

2. Numerical model

In this research, we have extended the open-source software Yade-DEM to include swelling. Yade-DEM is a 3-dimensional discrete element code (DEM) [36]. DEM is a particle model that is capable of simulating deformation of granular materials by considering grain-scale interactions. In this research, DEM is employed to simulate the movement of 3-dimensional non-cohesive spheres inside a packing. To account for the presence of pore fluid in between the grains, Chareyre et al. [32] have coupled DEM with the Pore Finite Volume (PFV) method. In the following sections, we explain relevant parameters and equations of an existing DEM code, the extension we apply to include swelling particles, and the PFV method.

2.1. Discrete element method

DEM simulates the motion of individual particles inside particle packings during deformation. Each particle is defined by its properties such as radius (r_i), Young's modulus (E_i), density (ρ_i), Poisson ratio (ν_i), shear modulus (G_i), and friction coefficient (φ). At a contact between two particles the following processes can occur: normal deformation, shear, and sliding.

The contact mechanics are based on the soft sphere approach. Thus, if particles *i* and *j* are pushed towards each other, they may deform locally at their contact. This local deformation is assumed to be linearly elastic and it is measured by the normal displacement δ_{ij}^n [L], defined by:

$$\delta_{ij}^n = \begin{cases} 0 & if \ \ r_i + r_j \le d_{ij} \\ r_i + r_j - d_{ij} & if \ \ r_i + r_j > d_{ij} \end{cases} \tag{1}$$

where d_{ij} [L] is the distance between the centres of particles *i* and *j*. An elastic force arises at the contact area of particles *i* and *j*, which acts towards reversing the overlap of particles. The elastic force is calculated using the Hertz-Mindlin theorem (see *e.g* [37].). In this theorem, small deformations are assumed to occur at the contact points between two particles such that $\delta_{ij}^n \ll \min(r_i, r_j)$. Based on the Hertz-Mindlin contact mechanics, the following effective parameters are defined for two particles *i* and *j* that are in contact with each other: the effective Young's modulus: $E_{ij} = \left(\frac{1-\nu_i^2}{E_i} + \frac{1-\nu_j^2}{E_j}\right)^{-1}$, the harmonic mean of particle radii: $r_{ij} = \frac{r_i r_j}{r_i + r_j}$, the average shear modulus: $G_{ij} = \frac{G_i + G_j}{2}$, and the averaged Poisson ratio: $\nu_{ij} = \frac{\nu_i + \nu_j}{2}$.

Normal displacement causes a normal force at a contact point f_{ij}^n [MLT^{-2}], which is given by [37,38]:

$$f_{ij}^n = -k_{ij}^n \left(\delta_{ij}^n\right)^{3/2} \tag{2}$$

where k_{ij}^n [MT^{-2} L$^{-1/2}$] is the contact stiffness in the normal direction and is given by:

$$k_{ij}^n = \frac{4}{3} E_{ij} \sqrt{r_{ij}} \tag{3}$$