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Pore-scale modelling and tomographic visualisation of drying in granular media

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Abstract

Spatio-temporal evolution of liquid phase clusters during drying of a granular medium (realised by random packing of cylindrical particles) has been investigated at the length-scale of individual pores. X-ray microtomography has been used to explicitly resolve the three-dimensional spatial distribution of the solid, liquid, and gas phases within the wet particle assemblies. The propagation of liquid menisci through the granular medium during drying was dynamically followed. The effect of contact angle on the degree of dispersion of the drying front has been studied by observing drying in a layer of untreated (hydrophilic) and silanised particles; the drying front was found to be sharper in the case of the silanised (less hydrophilic) particles. This observation was confirmed by direct numerical simulations of drying in a digitally encoded porous medium identical in structure to the experimental one. The simulations also revealed that the average gas–liquid interfacial area in a given porous microstructure strongly depends on the contact angle.

Keywords: Drying front; Porous media; Particle packing; Contact angle; Meniscus; Interfacial area; X-ray microtomography; Volume-of-fluid method

1. Introduction

Drying is a widely used industrial process, employed in applications such as isolation of particulate solids [1] or dewatering of foods and other bio-materials. Drying can be described on several length-scales [2], as shown in Fig. 1. At the process unit length-scale, aggregate quantities such as the total heat transfer rate into a process vessel and the total moisture removal rate are of interest. At the so-called effective medium length-scale, spatial distribution of moisture content and temperature within the material are resolved. Finally, details of the wet material microstructure (packed bed of particles, porous solid, etc.) and the morphology of liquid menisci within the individual pores are revealed at the pore space length-scale.

Mathematical modelling of drying at the effective medium length-scale involves the solution of volume-averaged transport equations—the Fourier's law for heat conduction, the Fick's law for diffusion, and the Darcy's law for fluid flow [3,4]. Knowledge of the dependence of effective transport coefficients (effective thermal conductivity, effective vapour-phase diffusivity, and liquid-phase permeability) on the microstructure and the moisture content of the porous medium is required as input information [5] and can be obtained by simulations at the pore space length-scale [6,7]. Published models of drying at the pore space length-scale have traditionally been based on approximating the porous microstructure by a two- or three-dimensional capillary network and the solution of vapour diffusion and capillary flow problems on that network [8–10]. This approach is methodologically similar to that used for network-based modelling of imbibition phenomena in porous media [11]. Drying can be regarded as an invasion percolation phenomenon, and indeed drying fronts with fractal structure have been shown to evolve [12,13].

An alternative approach to pore-scale modelling of drying is to construct "realistic" three-dimensional models of the porous medium, such as regular or random packings of spheres [14–16], and to determine liquid menisci configuration in the structural model either analytically (if possible) or numerically [17]. The objective of the present work is to take this

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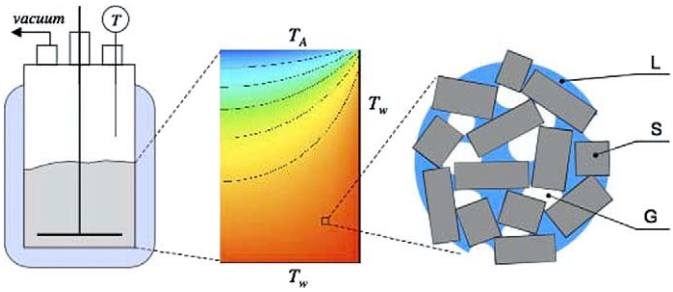

Fig. 1. Hierarchy of length-scales at which drying processes can be described: unit operation length-scale (left), effective medium length-scale (middle), and pore space length-scale (right).

approach one step further and perform numerical simulation of liquid menisci propagation through digitally encoded porous structures [18] obtained by X-ray microtomography and therefore identical to a physical granular medium in which drying experiments have been carried out. The numerical procedure is based on the volume-of-fluid (VOF) method which has been used in the past for the simulation of other processes involving liquid interface propagation in spatially complex structures, such as capillary condensation [19], bubble nucleation in porous media [20], or coating of rough particle surfaces [21].

Experimental techniques suitable for non-invasive monitoring of fluid-phase profiles in porous media include laser-induced fluorescence [22], magnetic resonance imaging [23–27], and X-ray computed tomography [28,29]. The latter two have been used for the visualisation of drying phenomena at the effective medium length-scale. While information about moisture distribution in the bulk material was obtained, neither the microstructure of the porous medium nor the individual liquid menisci within the pores were explicitly resolved. The objective of the present work is to use X-ray microtomography to visualise individual liquid–vapour interfaces within the pores, and to compare the experimental liquid-phase morphologies with those obtained numerically in identical microstructures. Furthermore, the effect of particle surface wettability on the shape of the drying front will be investigated.

2. Materials and methods

2.1. Drying experiments set-up

For the observation of the drying process, the system of water and cylindrical alumina extrudates (Criterion Catalysts and Technologies, USA) with the diameter 1 mm and the length 3–4 mm has been chosen. In order to change the wetting properties of the extrudate particles from hydrophilic to neutral, they have been impregnated in a 5% solution of dichlorodimethylsilane in trichloroethylene according to a procedure described in [30]. The reaction taking place at the surface of alumina is the condensation of the dichlorodimethylsilane with the surface hydroxyl groups. Both the untreated (hydrophilic) and the silanised (neutral) particles have been then used in the drying experiments.

The sample of extrudates was placed into a cylindrical plastic container with the diameter 12 mm, forming a packed bed of particles of the height approximately 10 mm. The sample was completely saturated by water. During the drying stage the container was placed inside a heated vessel and kept either at 150 mbar or (for certain experiments) at atmospheric pressure for a period of time varying from 15 to 30 min. Heat inflow to the sample occurred by conduction through the bottom of the container, which was kept at a constant temperature of 65 °C.

At the end of each drying step, the sample was weighted determining the amount of water evaporated during this step. The container was then closed and the geometry of the liquid and the solid phase of the sample was analysed by an X-ray microtomography scanner, as will be described in Section 2.2. Even if great care was taken during the manipulation with the container in order to minimise the movement of the solid phase due to small shakes, it was not possible to completely avoid small dislocations of particles between scans. The dislocations became more pronounced near the end of experiment when the liquid content was very low and the cohesive forces of capillary liquid bridges between particles disappeared.

2.2. Tomography scanning and image processing

A portable X-ray microtomography instrument Skyscan 1074HR (SkyScan, Belgium) equipped with 40 kV, 1 mA X-ray tube and a 768 × 576 pixels X-ray camera with pixel size of 22 μm [31] has been used. The volume of the sample which can be analysed is about 1 cm^3. The principle of X-ray microtomography is illustrated in Fig. 2a. The sample is placed between the X-ray source and the detector. The X-rays are attenuated as they pass through the sample, the extent of this attenuation is proportional to the local density, creating a transmission image (cf. Fig. 2b). The sample is placed on a rotation holder and transmission images obtained from different angles are used for the reconstruction of a three-dimensional map of the attenuation coefficient within the sample [32]. An example of a section of the 3D image obtained by the scanner is shown in Fig. 3a.

Frequency distributions (histograms) of the attenuation coefficient for two experiments are shown in Figs. 4a and 4b. Each curve in these graphs represents one scan; between two consecutive scans the sample was subjected to drying stage as described in Section 2.1. The attenuation coefficient is proportional to the density of the material: the first peak from the right in Fig. 4 represents the solid phase, the second peak from the right is the liquid phase (this peak is disappearing as the content of water in the sample decreases during drying), finally, the two remaining peaks at the left are for the wall of the plastic container and for the gas phase, respectively.

To continue the analysis of the geometry of the sample after scanning, it is necessary to segment the image, i.e., to separate the solid, liquid and gas phases. Two threshold values of the attenuation coefficient were found: a_1 at the local minimum of the frequency distribution between peaks for a container wall and the liquid (i.e., $a_1 \approx 0.14$ in Fig. 4a) and a_2 at the minimum between liquid and solid phase peaks (i.e., $a_2 \approx 0.4$ in Fig. 4a). The solid, the liquid and the gas phases are then distinguished as follows: (i) voxels with the attenuation coefficient lower than a_1 are assigned to the gas phase, (ii) voxels with