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SUB-DARCY-SCALE MODELING OF NON-UNIFORM FLOW THROUGH POROUS MEDIA WITH MIXED WETTABILITIES

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ABSTRACT: A physically-based conceptual model is developed using spatially distributed sub-Darcy-scale clusters in a regular grid to reproduce both the hydraulic properties and the non-uniform wetting and drainage fronts through porous media mixed with hydrophilic and hydrophobic grains. In the model, cellular automaton-like algorithm is employed to route water- and air-intrusion paths through the mixed porous media. Water retention characteristics, which are ones of the macroscopic properties of porous media, are estimated by accumulating a certain numbers of clusters after reaching equilibrium states in a wetting or drainage process. In the experimental part of this study, normal (hydrophilic) and artificially hydrophobized glass spheres, whose diameter is about 0.2 mm, are used as materials, and hydraulic properties of homogeneously mixed glass spheres with various rates are measured. The measured data are compared to model estimation, and the availability of the model is shown.

Keywords: Water Retention Properties, Hydrophilicity, Hydrophobicity, Cellular Automaton, Glass Spheres

1. INTRODUCTION

From the early 1900's, it has been reported that there exist soils that have resistance to water infiltration in various countries such as New Zealand, Australia, Netherlands, and so on, and major concerns have been raised because such soils, which are referred to as water-repellent soils, cause soil erosion and productivity reduction in farmlands [1]. In some cases of a natural environment, soil gains water repellency by being coated with organic matters decomposed by fungi, and it is observed that interfusion of hydrophobic grains into hydrophilic grains makes dramatic differences in flow behavior and hydraulic properties [2].

Various works assessing and quantifying water repellency and hydraulic properties of artificial water-repellent soils as well as natural ones have been conducted, and it is shown that the hydraulic properties of soils change, depending on the increasing mix rate of hydrophobic grains [3][4]. While measured water retention properties could be represented with relative ease in some functional form such as the van Genuchten model, predicting those properties effected by grains' wettabilities needs microscopic approaches. For instance, a functional model is proposed by Ustohal *et al*. (1998) [3], in which the Brooks-Corey model is extended using probability distribution of equivalent contact angles on grain clusters with mixed wettabilities. And pore-scale network modeling also proficiently predicts multiphase flow properties through mixed porous media [5]. But this approach requires large computational load even for several cubic centimeters of porous media.

In this study, a physically-based conceptual model, which requires relatively lower computational load, is developed by adopting the concept on the cluster of grains. The model realizes a spatial distribution of clusters and makes it possible to reproduce non-uniform wetting and drainage fronts through porous media mixed with hydrophilic and hydrophobic grains. And water retention properties of the mixed porous media are estimated from the computed results in a bottom-up manner.

2. MODEL

2.1 Model Structure

The physically-based conceptual model proposed here employs a cellular-automaton-like algorithm to track water- and air-intrusion paths through porous media. Generally, a cellular automaton is referred to as a collection of cells that have several variable states in a regular grid, and the cells evolve according to a set of rules based on the states of neighboring cells during iteration of discrete time steps. The rules are applied to initial cell states, and new cell states are obtained according to the rules. This process is repeated for as many time steps as desired [6].

In this study, cells correspond to clusters of multiple grains as illustrated in Fig. 1. This figure depicts a schematic of the model two-dimensionally, but grains, cells and capillary tubes are defined as three-dimensional objects in the

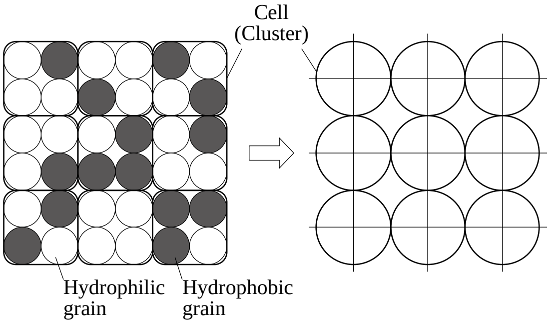


Fig. 1 Relation between grains and cells in a case of a square grid

model. The concept about the cluster is originally used by Ustohal *et al*. (1998) [3] for modeling hydraulic properties of mixed porous media by using a functional form, and they presumed that the contact angle in each cluster is homogeneous, based on the Cassie's law. The contact angle, which is equivalent one synthesized from mosaic of various contact-angle surfaces, is determined, depending on the ratio of hydrophilic and hydrophobic surface areas included in each cluster. While Ustohal *et al*. (1998) [3] treated a cluster itself as a meso-scale wall surface, we assume in this model that each cluster conceptually possesses a bundle of circular tubes with various diameters. And it is also assumed that each tube has constant diameter through a single cell, and that the equivalent contact angle of the tubes in a cell has identical value. The equivalent contact angle is estimated from the Cassie-Baxter equation, depending on the ratio of surface areas of the grains with different contact angles. When grains with different types of contact angle are included, the equivalent contact angle of the i th cell ($i = 1,...,n^{c}$; n^{c} is the number of cells) is represented as follows.

$$\cos\varphi_i^{c} = \sum_{j=1}^{n^{a}} f_{ij} \cos_{j} \quad (1)$$

with

$$f_{ij} = A_{ij} / \sum_{k=1}^{n^{a}} A_{ik} \quad (2)$$

where φ_i^{c} is the equivalent contact angle of the i th cell, φ_j is the j th contact angle among n^{a} types, f_{ij} is the ratio of the surface area with j th contact angle to the total area, and A_{ij} is the surface area. If the size of all grains is equal and two types of wettability, *i.e.*, hydrophilic and hydrophobic grains, are included, Eqs. (1) and (2) could be represented as follows.

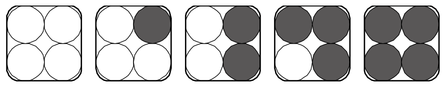
$$\cos\varphi_i^{c} = f_i^{D}\cos\varphi^{D} + (1 - f_i^{D})\cos\varphi^{P} \quad (3)$$

$$f_i^{D} = n_i^{D} / n^{g} \quad (4)$$

where n^{g} is the number of grains included in a cell, and n_i^{D} is the number of the hydrophilic grains in the i th cell. Equations (3) and (4) imply that equivalent contact angle of a cell that has n^{g} grains can have $(n^{g}+1)$ types of realization, depending on the number of n_i^{D} ($= 0,1,...,n^{g}$). When the fraction of the hydrophobic grains included in a whole porous medium is p^{P} , the probability of the realization that has n_i^{D} ($= 0,1,...,n^{g}$) hydrophilic grains is described by the following binominal distribution.

$$P(n_i^{D}, n^{g}) = \frac{n^{g}!}{n_i^{D}!(n^{g} - n_i^{D})!}(1 - p^{P})^{n_i^{D}}(p^{P})^{n^{g} - n_i^{D}} \quad (5)$$

In a case where a cell has four grains, the contact angles of hydrophilic and hydrophobic grains are 45.0 degrees and 120.0 degrees, respectively, and the mix rate of hydrophobic grains is 0.75, a cell has five realizations. The equivalent contact angles and probabilities of the realizations are shown in Fig. 2.



equivalent contact angle (deg)	45.0	66.1	84.1	101.4	120.0
probability (binominal distribution)	0.004	0.05	0.21	0.42	0.32

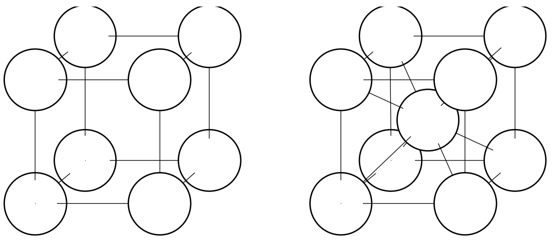
Fig. 2 Realizations of a cell, equivalent contact angles, and probabilities in a case where the mix rate of hydrophobic grains is 0.75

Capillary tubes in each cell have an ability of capillary rise or fall, depending on the equivalent contact angle and the radius of tube, if the lower end of tube is connected to a water pool. The height of the capillary rise or fall from the surface of the water pool is termed 'potential' capillary height here, and the height is represented based on the Young-Laplace equation for circular tubes.

$$h_{ij}^{c} = \frac{2\sigma\cos\varphi_i^{c}}{\rho g r_{ij}} \quad (6)$$

where h_{ij}^{c} and r_{ij} are the potential capillary height and the radius of the j th tube in the i th cell, respectively, σ and ρ are the surface tension and the density of water, respectively, and g is the gravitational acceleration. Equation (6) means that capillary rise could occur when h_{ij}^{c} is positive ($\varphi_i^{c} < 90$ degrees), and that capillary fall when h_{ij}^{c} negative ($\varphi_i^{c} > 90$ degrees).

There are various options for arrangement of cells and grains. For cell arrangement, the following grids are considered, such as a simple cubic grid and body-centered cubic grid as shown in Fig. 3. Two classes of grain packings are introduced in Ustohal *et al*. (1998) [3]: tetrahedral packings and cubic packings. One of the essential indices for selection of grid is connectivity of cells, which is the number of neighboring cells. It is reported that the connectivity of pores is an essential factor that effects hydraulic properties of porous media by Gharbi and Blunt (2012) [5]. In this model, the cell connectivity has an effect on likelihood of state change of cells.



(a) Simple cubic grid (b) Body-centered cubic grid

Fig. 3 Three-dimensional cell arrangement

2.2 Rules for Wetting and Drainage Processes

In this model, each capillary tube in cells has two states: dry and wet states, and therefore each cell has three states: dry, partially wet, and completely wet states. The dry and completely wet states of a cell imply that all the capillary tubes in the cell are in the dry state and the wet state, respectively, and the partially wet state of a cell implies that some (not all) tubes are wet. Here, it is assumed that dry and completely wet cells have air permeability and water permeability, respectively, and that partially wet cells have both air and water permeabilities. The description such as a just 'wet' cell represents that the cell is 'partially wet' or 'completely wet'.

One state of a capillary tube could change into the other, depending only on the current states of its own and neighboring cells. After all tubes are evaluated based on the current states, all the states of tubes are updated simultaneously and a new set of states is obtained. This process is iterated until any change does not occur or desired time steps are satisfied. To simulate water intrusion into a dry porous medium (wetting process) or air intrusion into a fully wet medium (drainage process), the conditions for state change are given as described below.

In the wetting process, initially each cell in an objective domain is set to be dry state, and cells at the upper end and at the bottom end are supposed to connect to exterior air and water pools, respectively. An objective capillary tube in a dry state changes into wet, if the tube holds the following four conditions:

1) the cell including the objective dry tube connects to the air pool through other dry or partially wet cells, or directly,
2) any of its neighboring cells is wet,
3) any of the neighboring wet cells connects to the water pool through other wet cells or directly, and
4) the potential capillary height of the objective j th tube in the i th cell satisfies the following inequality.

$$h_{ij}^{\mathrm{c}} > h_i^{\mathrm{p}} \tag{7}$$

where h_i^{p} ($= -p_i / \rho g$, p_i is the water pressure placed on the i th cell) is equal to the height from the surface of the water pool.

The first condition assumes that the air pressure in dry tubes that connect to the exterior air pool is equal to atmospheric pressure, which is supposed to be constant here, and that the air pressure of enclosed air in dry tubes that do not connect to the air pool could change and constrict water entry by air pressure increase. The fourth condition assumes that the water pressure in wet cells that connect to the water pool is hydrostatic one.

In the drainage process, initially all tubes are set to be wet. Cells at the top end and at the bottom end connect to exterior air and water pools, respectively, in the same way with the wetting process. An objective wet tube changes into dry, if the wet tube holds the following four conditions.

1) the cell including the objective wet tube connects to the water pool through other wet cells or directly,
2) any of its neighboring cells is dry or partially wet,
3) any of the neighboring dry or partially wet cells connects to the air pool through other dry or partially wet cells, or directly, and
4) the potential capillary height of the objective j th capillary tubes in the i th cell satisfies the following inequality.

$$h_{ij}^{\mathrm{c}} < h_i^{\mathrm{p}} \tag{8}$$

2.3 Estimation of Water Retention Properties

The proposed model treats water and air intrusions in a sub-Darcy scale as mentioned above. The hydraulic properties of porous media such as the water retention property and permeability are macro-scale ones. So, to estimate such macro-scopic properties, some operation such as integration and averaging over some part of a computational domain is needed using a computed result.

To evaluate water retention properties, saturation at a certain water pressure head is calculated from cells within a certain range of water pressure heads as follows.

$$S_{\mathrm{w}}(p) = \sum_{i \in \Omega(p)} \sum_{j=1}^{n^{\mathrm{t}}} \alpha_{ij} r_{ij}^2 \Big/ \sum_{i \in \Omega(p)} \sum_{j=1}^{n^{\mathrm{t}}} r_{ij}^2 \tag{9}$$

with

$$\alpha_{ij} = \begin{cases} 1 & \text{if } j\text{th tube in } i\text{th cell is wet} \\ 0 & \text{if } j\text{th tube in } i\text{th cell is dry} \end{cases} \tag{10}$$

$$\Omega(p) = \left\{ i \in 1, \ldots, n^c \;\middle|\; p - \Delta p < p_i \leq p + \Delta p \right\} \quad (11)$$

where S_w is the saturation, α_{ij} is a coefficient that distinguishes whether the j th tube in the i th cell is wet or dry, $\Omega(p)$ is a set of cells under water pressure from $p - \Delta p$ to $p + \Delta p$, n^c is the number of cells, and p_i is the pressure placed on the i th cell.

3. EXPERIMENT

3.1 Soil Column Method

The soil column method is employed to measure the water retention properties of porous media mixed hydrophilic and hydrophobic grains. The soil column is made by piling up acrylic circular cylinders whose inner diameter is 4.8 cm and height is 4.0 cm, and each cylinder is taped together strictly to avoid air and water leaking. The bottom of the column is covered by a porous filter, and materials, which are dried adequately in a desiccator, are packed in the column at a prescribed density. Finally, the top is covered by a wrap with small holes to prevent evaporation and air entrapment (Fig. 4). In a case of a drainage process, a packed column is saturated by the vacuum method, because materials containing a high percentage of hydrophobic grains will not inundate spontaneously. Then, a certain water pressure is constantly placed on the bottom of the column until the column reaches an equilibrium state (72 hours in this study), and weights of materials in each cylinder is measured before and after oven-dry. All these procedures are conducted in a room in which air temperature is kept at 20 degrees Celsius.

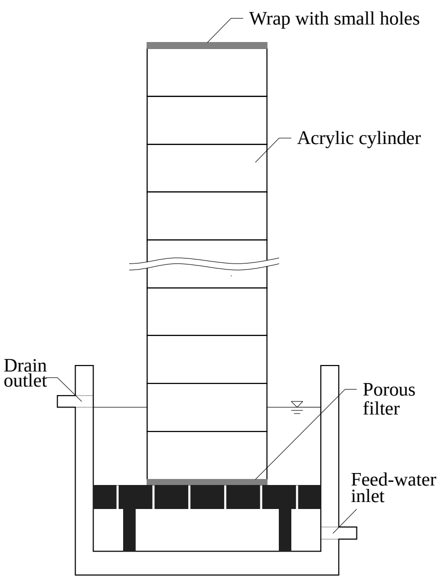


Fig. 4 Soil column method.

3.2 Material

In this model, we assume a uniform grain size for each wettability. So, for the experiments glass spheres whose diameter is 0.2 mm are used as hydrophilic materials. It is checked that the distribution of the diameter is narrow enough from images taken by a microscope. After washing and drying, the glass spheres, which are hydrophilic originally, are hydrophobized by vapor-depositing octyltrichlorosilane ($CH_3(CH_2)_7SiCl_3$, OTS), which is one of popular chemical materials used in other works [3][4]. Specimens are prepared by homo-geneously mixing the hydrophilic and hydrophobic spheres at the following mass-based rates of hydrophobic grains: the mix rates of hydrophobic grains are 0.00, 0.25, 0.50, 0.75, and 1.00. And the bulk density of the specimens is controlled to be 1.58 g/cm^3 when materials are packed in the acrylic cylinders.

4. RESULTS AND DISCUSSION

4.1 Experimental Results

The water retention properties of the glass spheres 0.2 mm in diameter are measured by the soil column method. The measured properties of both wetting and drainage processes are shown in Figs. 5 and 6, respectively. These results show that the mix rate of hydrophobic grains has a great effect on water retention properties, and that the water retentivity decreases as the rate increases in both wetting and drainage processes. And this leads to the feature that the specimens mixed with hydrophobic grains have a larger hysteretic property compared with the specimen with no hydrophobic grains. Furthermore, it is notable that capillary rise is blocked in the specimen mixed with 25 or more percentages of hydrophobic grains.

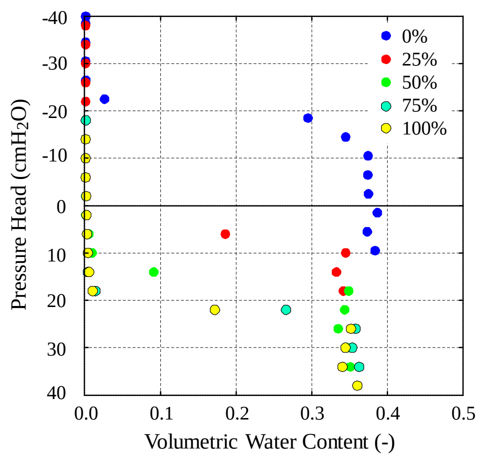


Fig. 5 Measured water retention properties at various mix rates of hydrophobic grains (wetting

process)

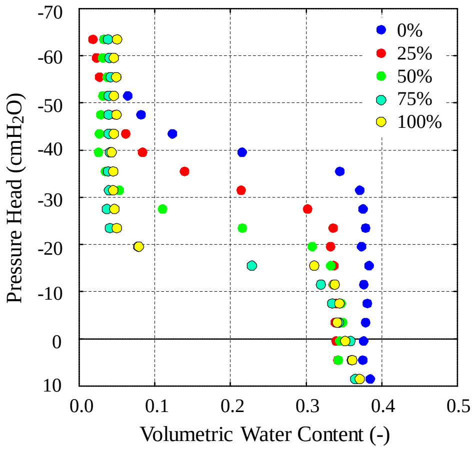


Fig. 6 Measured water retention properties at various mix rates of hydrophobic grains (drainage process)

4.2 Parameters and Computational Conditions

In this study, a simple cubic grid, in which inner cells have six neighboring cells, is employed. The grid interval is estimated from the following equation, based on the diameter of grains and the number of grains in one cell.

$$l = \left(\frac{4\rho_g \pi r_g^3 n^p}{3\rho_b} \right)^{\frac{1}{3}} \tag{12}$$

where l is the grid interval, ρ_g is the density of glass, ρ_b is the bulk density of the specimen, and r_g is the radius of the glass sphere. The number of capillary tubes in a cell is also estimated, assuming grains are arranged in a simple cubic grid.

$$n^t = \mathrm{round}\left(l^2 \left\{ \left(\frac{3\rho_b}{4\rho_g \pi r_g^3} \right)^{\frac{1}{3}} + 1 \right\}^2 \right) \tag{13}$$

where round(·) is the function that rounds input value off to the closest whole number.

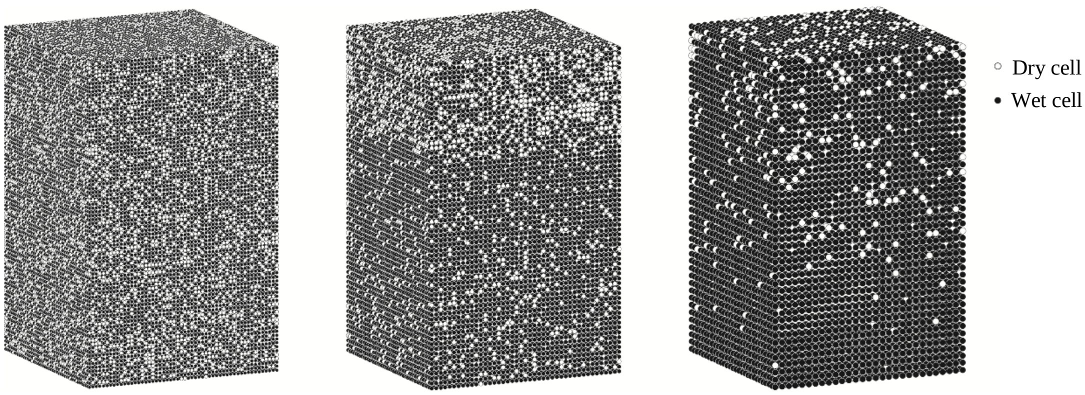
The computational domain is set as a cuboid and the size is $\sqrt{10}\mathrm{cm} \times \sqrt{10}\mathrm{cm} \times 80\mathrm{cm}$ which is based on the experimental column used here. Simulations are conducted with three types of grain number included in one cell: the number is 4 in Type I, 8 in Type II, and 27 in Type III. The parameters included in the model are given or estimated based on the materials used for experiments and those are summarized in Table 1. The radius of the capillary tubes is determined here by the normal distribution whose average μ^t and standard deviation σ^t are 0.04 mm and 0.006 mm, respectively. These values are based on the previous work that estimated hydraulic conductivity of porous media using a pore-scale network model [7].

Table 1 Parameters used for each type

	Type I	Type II	Type III
φ^D (deg)		45	
φ^P (deg)		120	
μ^t (mm)		0.04	
σ^t (mm)		0.006	
n^g	4	8	27
n^t	3	4	9
l (mm)	0.298	0.376	0.564

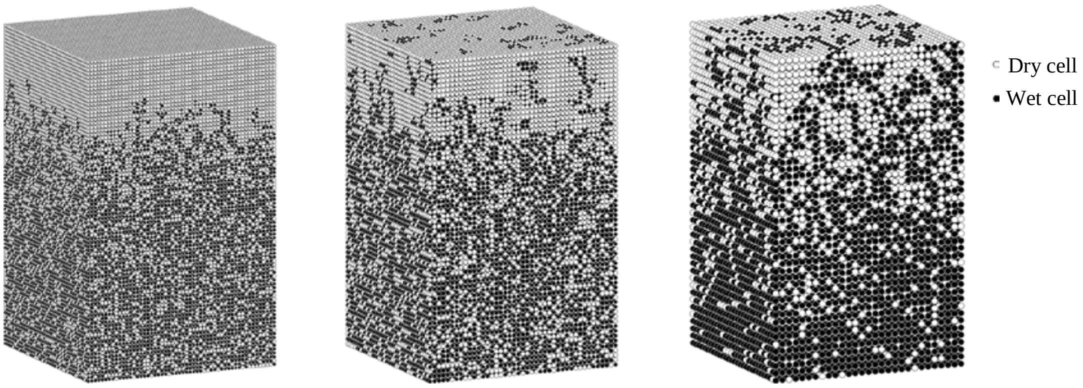
4.3 Computed Results

Some of the computed results of non-uniform water intrusions in wetting processes are shown in Figs. 7 and 8. The mix rates of hydrophobic grains are 50% and 75%, respectively, and the pressures placed on the bottom of the samples are 5 cmH_2O and 10 cmH_2O, respectively. The height of the depicted samples is 5 cm, and those are cut out



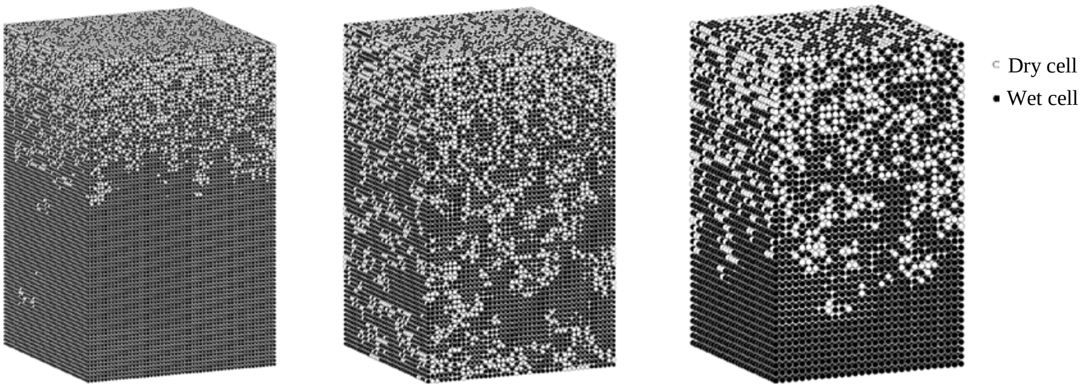
(a) Type I (b) Type II (c) Type III

Fig. 7 Water intrusions in a case where the mix rate of hydrophobic grains is 50% and the pressure placed on the bottom is 5 cmH_2O



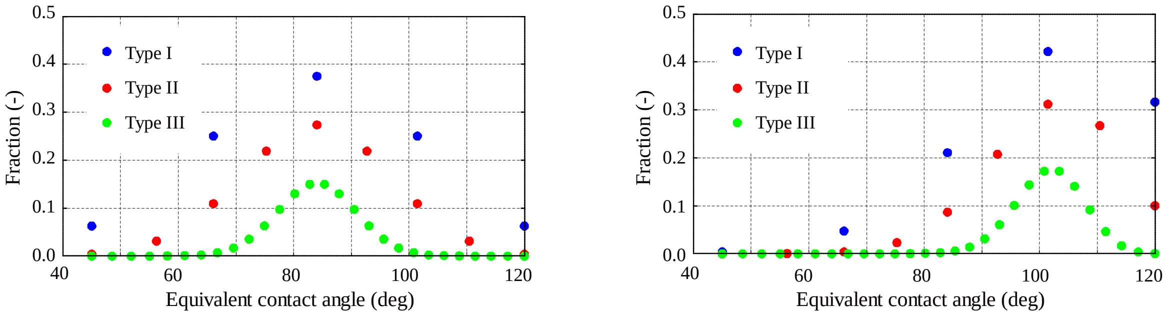
(a) Type I (b) Type II (c) Type III

Fig. 8 Water intrusions in a case where the mix rate of hydrophobic grains is 75% and the pressure placed on the bottom is 10 cmH_2O



(a) Type I (b) Type II (c) Type III

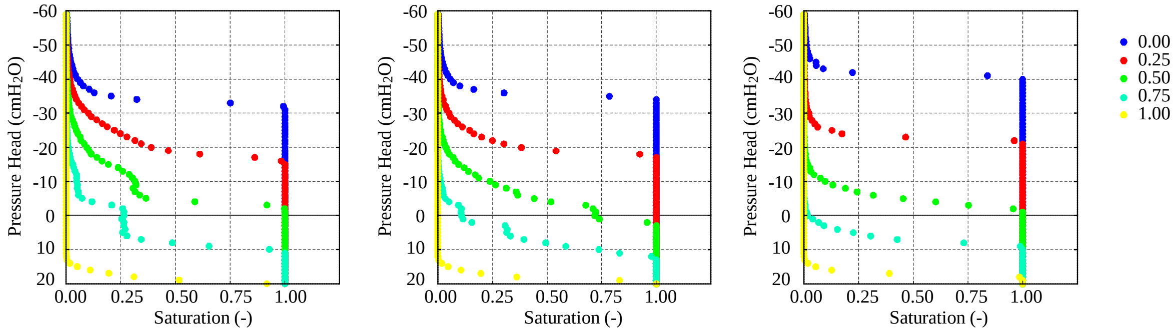
Fig. 9 Air intrusions in a case where the mix rate of hydrophobic grains is 50% and the pressure placed on the bottom is 0 cmH_2O



(a) Mix rate: 50% (b) Mix rate: 75%

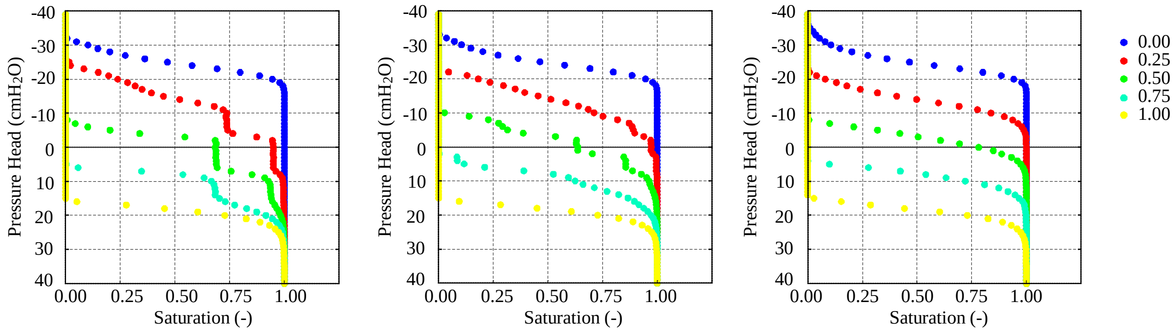
Fig. 10 Fractions of each realization in Type I, Type II, and Type III

from the original computed domains 80 cm in height. In Fig. 9, non-uniform air intrusions in drainage processes are shown. The mix rate is 50%, and the pressure placed on the bottom of the columns is 0 cmH_2O. These figures indicate that the grain number in a cell has a considerable influence on intrusion regime. As shown in Fig. 10, Type I and Type II have a wide range of equivalent contact angles of cells, compared with Type III. This also implies that cells become more homogeneous as the grain number in a cell increases. It explains the differences in appearance among Type I, II, and III. Furthermore, self-similar water- and air-intrusions are found in the upper quarter of the columns in Fig. 8 (b) and (c), and the lower half of the columns in Fig. 9 (b) and (c). From a viewpoint of the percolation theory, it is known that a self-similar intrusion path is formed when a little greater than 31% of elements (cells in this study) are permeable in the simple cubic lattice [8]. In the case of Type II (Fig. 8 (b)), the fraction of permeable cells, whose equivalent contact angle is below 92.7 degrees, is about 32%, and so a self-similar water-intrusion occurs. In the



(a) Type I (b) Type II (c) Type III

Fig. 11 Water retention properties of wetting processes



(a) Type I (b) Type II (c) Type III

Fig. 12 Water retention properties of drainage processes

case of Type I (Fig. 8 (a)), the fraction of permeable cells, below 84.1 degrees, is about 26%, and so a self-similar intrusion does not occurs.

Next, the computed water retention properties of wetting and drainage processes are shown in Figs. 11 and 12, respectively. These figures show that water retentivity decreases clearly as the mix rate of hydrophobic grains increases. While all the saturations varies smoothly in Type III, there exist several step-like changes in Type I and Type II. Such a step-like change is also observed in Fig. 7 (b), which shows that the ratio of dry cells increases obviously above about three-quarters of the sample height. The cause of the sudden change in saturation is also considered that each realization of the equivalent contact angle in Type I and Type II has a relatively large fraction, compared with Type III as shown in Fig. 10. Therefore, when the water pressure in cells decreases and goes beyond a certain limit corresponding to an equivalent contact angle (Eq. (7)), the relatively large fraction of cells becomes impermeable all together.

Compared with the experimental results (Figs. 5 and 6), the computed results shows some differences. In the wetting processes, water enters capillary tubes under negative water pressure even in the case of 0.50 of the mix rate in the computed results, while spontaneous water-entry is prevented from only 0.25 of the mix rate in the measured ones. This indicates that in actual cases a single hydrophobic grain has an influence on all the hydrophilic neighboring grains, and which is left out of consideration by dividing the grain gathering into many clusters in the model.

In the drainage processes, initially saturated water is drained promptly depending on the mix rate in the computed results, while much water is kept up to -20 cmH_2O even in the high rate cases of the measured results. The reason of this discrepancy is considered that speed of air-water interface movement is not considered in the model. In actual cases, water in large pores is drained in advance of that in small pores, and such prioritized drainage could make water isolation in a pore-network. But in the developed model all waters included in cells that satisfies the drain conditions are drained at once in one discrete time step.

5. CONCLUSION

A physically-based conceptual model was proposed to reproduce non-uniform water- and air-intrusions through porous media mixed with hydrophilic and hydrophobic grains. In the model, the cellular automaton-like algorithm was employed to figure out the intruding paths, and the water retention properties were estimated from the computed intrusion paths in a bottom-up manner.

The computed results that had reproduced wetting and drainage processes were compared with the experimental results using glass spheres hydrophobized by OTS. Although there were discrepancies in the water-entry pressure for the wetting processes and in the air-entry pressure for the drainage processes, the dependence of water retentivity on the mix rate of hydrophobic grains and non-uniform water and air intrusions were illustrated. And it was also shown that the number of grains in a cell was one of the essential factors in this model, because it effects the value and probability of the equivalent contact angle in cells. From these results, it is concluded that the proposed model has the possibility of being provided as a useful tool to estimate the hydraulic properties of porous media with different wettabilities.

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