

Liquid water transport model in hydrophilic granular media: Preliminary validation with drying rate of hierarchical granular material

Hyuga Yasuda, Hiroaki Katsuragi , and Makoto Katsura ^{*}

Department of Earth and Space Science, [Osaka University](#), 1-1 Machikaneyama, Toyonaka 560-0043, Japan



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The drying rate profile of granular beds can be divided into the constant rate period (CRP), which is characterized by a nearly constant drying rate, and the falling rate period (FRP), in which the drying rate rapidly decays. In order to explain this behavior quantitatively, we proposed a simple one-dimensional power law model in which the product of the water permeability and the pressure gradient are assumed to be proportional to the cube of the saturation. To test this model, we measured the drying rates of glass beads and hierarchical granular materials produced by sintering and breaking glass beads. Our results and those of previous experiments showed consistency with the power law. The obtained proportional constant of the experimental power law also shows a rough agreement with that estimated from previous studies on water permeability and capillary pressure. Drying behavior in FRP also agrees with our model in some points. The remnant deviation of the model from experimental results may be attributed to the inhomogeneity of granular media, which was qualitatively verified.

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I. INTRODUCTION

Many studies have been conducted on the drying rates from porous media. The experimental results generally indicate a duration with a nearly constant drying rate (CRP: constant rate period), followed by a rapid decrease in the drying rate (FRP: falling rate period). In CRP, pore water is transported to the surface of the porous medium by permeable flow, and evaporation from the surface continues. However, there is no unified understanding of the conditions for the transition from CRP to FRP or the drying rate in FRP. Peat [1] and food products [2,3] exhibit exponential drying curves in FRP, corresponding to the dehydration process of binding water. A similar drying rate phenomenon is also possible in powders owing to the Kelvin effect when the pore diameter is less than a few nanometers. However, this is not essential for granular beds with large particle diameters. Exceptionally, this effect has been reported [4] in the case of granular beds with diameters of several hundred microns and very low moisture content ($\lesssim 1\%$).

Shokri *et al.* [5–7] saturated the granular media of submillimeter size in a container with dyed water and measured the drying rate. They used dye deposition to help identify the boundary between fully saturated, partially saturated, and dry regions. We cannot ignore the effect of gravity because the depth of the container exceeds the capillary rise:

$$\frac{2\Gamma \cos \theta}{\rho g r}, \quad (1)$$

^{*}Contact author: mhirai@ess.sci.osaka-u.ac.jp (he/him/his)

where Γ , ρ , g , r , and θ are the surface energy of water, the density of water, the acceleration of gravity, pore radius, and contact angle, respectively. As their container had a closed bottom, there was always a water-saturated region at the bottom of the sample. During CRP, the water-containing region continued to the sample surface because of the water sucked by capillary forces. During FRP (which they mentioned as a “transition period”), a dry region develops and expands under the surface. As for the period following FRP, which they mentioned it as “stage-2,” they found that the depth of the dry region obeys a power law with the system’s Bond number and insisted that the drying rate was controlled by water vapor diffusion through the dry region. However, they mentioned little about the transition mechanism from CRP to FRP and the behavior of the drying rate in FRP.

Although Thierry *et al.* [8] used bottomed containers, the effect of gravity was negligible because of their small pore diameters (mostly submicrometers). With MRI, they confirmed the growth of the dry region in FRP and showed for the first time that the falling rate can be understood quantitatively by water vapor diffusion through the dry region. Thierry *et al.* [8] defined the critical saturation, $\langle S \rangle^*$, as the spatially averaged water saturation when the water saturation gradient appears in the wet region and found that the transition occurs approximately at the same time. They also showed an empirical formula as below:

$$\langle S \rangle^* d^{1/4} = 1.0 \times 10^{-2} \text{m}^{1/4}, \quad (2)$$

where d was originally described as radius; however, we exchanged it with diameter according to P. Coussot (private communication, June 4, 2024). Here, we define “transition point” as the spatially averaged water saturation at the transition from CRP to FRP. Although the transition point is expected to be almost the same value as the critical saturation in granular materials, as discussed above, such a redefinition is more convenient as the spatial distribution of water saturation is technically difficult to observe more than the drying rate. From here on in this paper, we will consider the critical saturation to be equivalent to the transition point. They insisted that the growth of the dry region is controlled by the balance between the water supply by permeation and evaporative loss. They proposed a model assuming that water permeability and capillary pressure are proportional to S^2 and $S^{-0.5}$, respectively. Here, the variable S means local water saturation. However, the quantitative interpretation of the measured transition was not successful.

It remains unclear how the proportionality constant shown in Eq. (2) depends on experimental or sample parameters. In this study, we will construct a more universal physical model through further theoretical exploration and perform experimental verification.

II. POWER LAW MODEL OF LIQUID WATER TRANSPORT IN HYDROPHILIC GRANULAR MEDIA

Tables II and III summarize the variables used in this study, with Greek variables in Table II and Latin variables in Table III (see Appendix C). If water transport in the wet region is regarded as the advection of liquid water driven by capillary pressure gradient and water permeability [8], water saturation must satisfy the following equation:

$$\begin{aligned} \frac{\partial S}{\partial t} &= \nabla(D_L \nabla S), \\ D_L &:= \frac{K}{\eta} \frac{dP_c}{dS}, \end{aligned} \quad (3)$$

where t , K , η , and P_c are time, permeability of wet region, viscosity of water, and capillary pressure, respectively. This equation is in the form of a diffusion equation, even though it represents the advection of liquid water. Let us assume that the apparent diffusion coefficient is proportional to the n -th power of water saturation in order to establish the power dependency of equation Eq. (2):

$$D_L = D_L^0 S^n. \quad (4)$$

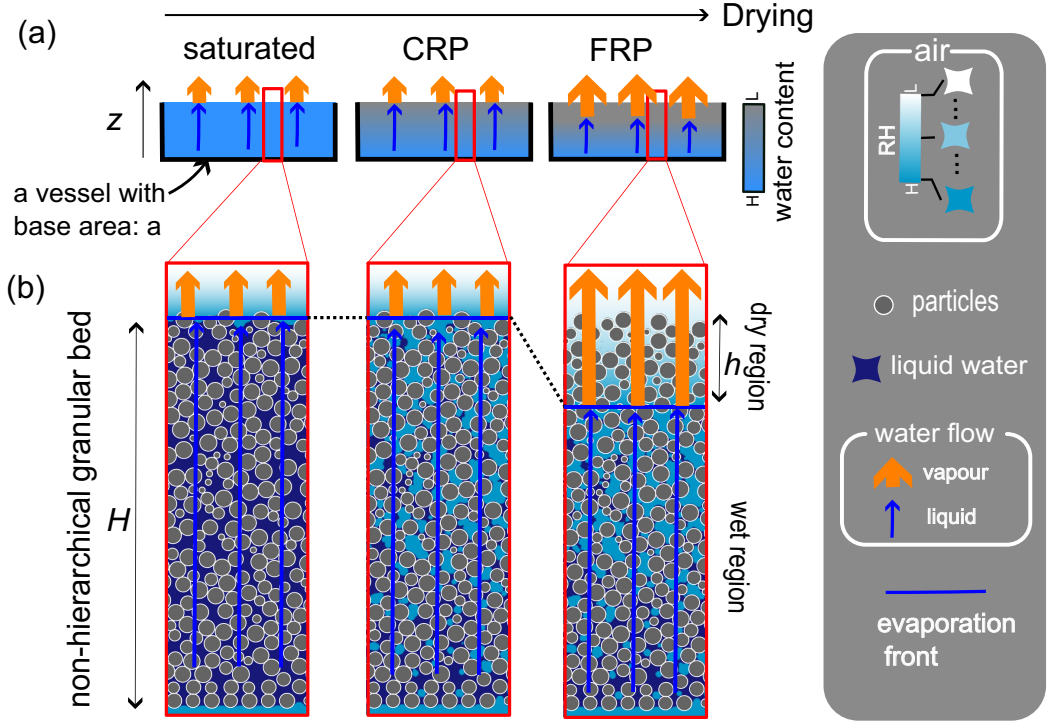


FIG. 1. Conceptual diagram of a granular bed, (a), and its enlarged cross-sectional view, (b). Granular was poured in a vessel with the base area of a . Evaporation creates a gradient in water saturation, resulting in an upward flow of liquid water. During CRP, air in the pore is almost 100% humid, so evaporation only occurs on the top surface of the granular. After the transition to FRP, a dry region generated at the top of the granular material recedes to the bottom. As there is no liquid water here, evaporation only occurs at the boundary with the wet region. This means that the evaporation front is present on the top surface in CRP and on the boundary between the two regions in FRP.

Here, if the sample is a flat granular bed of uniform thickness as illustrated in Fig. 1(a), and the spatial inhomogeneity of D_L can be neglected, Eq. (3) reduces to a one-dimensional problem in the wet region:

$$\frac{\partial S}{\partial t} = \frac{\partial}{\partial z} \left(D_L \frac{\partial S}{\partial z} \right). \quad (5)$$

Here, we set the z axis upward from the origin at the bottom as illustrated in Fig. 1(a). With the porosity of the granular, ϵ , the liquid water flux is given as $-\epsilon D_L \frac{\partial S}{\partial z}$. If the bed has a closed bottom as in the case of Thiery *et al.* [8], liquid water flow is zero at the bottom:

$$\epsilon D_L \frac{\partial S}{\partial z} \Big|_{z=0} = 0. \quad (6)$$

Because liquid water flow is limited under the boundary of dry and wet regions as illustrated in Fig. 1(b), integration of Eq. (5) around the boundary leads

$$S|_{z=H-h} \frac{dh}{dt} = D_L \frac{\partial S}{\partial z} \Big|_{z=H-h} + V, \quad (7)$$

where h , H , and V represent the thickness of the dry region, the thickness of the granular thickness, and the drying rate as the equivalent velocity of liquid water, respectively. As illustrated in Fig. 1(b), both h and $\frac{dh}{dt}$ must be zero during CRP. During FRP, $S|_{z=H-h}$ should be negligible due to the decreasing D_L at the receding boundary. As a result, liquid water and evaporative flows are balanced as follows:

$$D_L \frac{\partial S}{\partial z} \Big|_{z=H-h} = -V. \quad (8)$$

According to the definition of V , the relationship with the spatially averaged water saturation across the granular bed, $\langle S \rangle$, is as follows:

$$\epsilon H \frac{d}{dt} \langle S \rangle = -V. \quad (9)$$

Thiery *et al.* found that water saturation in the wet region has a large gradient only in a narrow region near the boundary with the dry region; otherwise, it is almost flat [8]. Based on their observation, we can presume an approximated solution as follows.

Let S_o be the water saturation at the origin ($z = 0$), and $\langle S \rangle_w$ be water saturation spatially averaged in the wet region, $\langle S \rangle H / (H - h)$. Assuming that the variation of the diffusion coefficient within the flat region is negligible and can be regarded as a constant value, $D_L^0 \langle S \rangle_w^n$, the water saturation can be approximated as the function, F , defined as follows:

$$F(z) := S_o - \frac{V}{\epsilon H' D_L^0 \langle S \rangle_w^n} z^2, \quad (10)$$

where H' is the height of the flat region and nearly equal to $H - h$. Hence, the approximation at the boundary, S_b , can be written as

$$S_b := F(H - h) \cong S_o - \frac{V(H - h)}{2\epsilon D_L^0 \langle S \rangle_w^n}. \quad (11)$$

In the CRP far before the transition, saturation in the entire wet region should be flat; therefore, the diffusion coefficients corresponding to S_o , S_b , and $\langle S \rangle_w$ are D_{Lo} , D_{Lb} , and $\langle D_L \rangle_w$ and will satisfy the following equation and fulfill the above condition:

$$\frac{D_{Lo} - D_{Lb}}{\langle D_L \rangle_w} \ll 1. \quad (12)$$

On the other hand, near the transition and in the FRP, a dry region is considered to be generated and grow because Eq. (12) is not satisfied. Then, we can set the critical condition as follows:

$$\frac{D_{Lo} - D_{Lb}}{\langle D_L \rangle_w} = m, \quad (13)$$

where m is a constant not sufficiently smaller than 1. By writing down the critical conditions using Eq. (4), the approximate solution can be written as follows:

$$\begin{aligned} \langle S \rangle &= \frac{H - h}{H} \left[\frac{nP}{2m} \right]^{\frac{1}{n+1}}, \\ P &:= \frac{V(H - h)}{\epsilon D_L^0}, \end{aligned} \quad (14)$$

where the definition of the dimensionless quantity P has a similar form of the Péclet number; however, the apparent diffusion coefficient in the denominator represents the advection of liquid water rather than the diffusion of vapor molecules, and the numerator V doesn't represent the advection rate but the drying rate that must be proportional to the vapor molecule diffusion constant. Based on the analysis so far, by normalizing water saturation with the values of P in CRP, and by

nondimensionalizing time and space, we can obtain the following differential equation with respect to σ and $\tilde{h}$:

$$\begin{aligned}\frac{\partial \sigma}{\partial \tilde{t}} &= \frac{\partial}{\partial \tilde{z}} \left(\sigma^n \frac{\partial}{\partial \tilde{z}} \sigma \right), \quad (0 \leq \tilde{z} \leq 1 - \tilde{h}) \\ \sigma(\tilde{t}, \tilde{z}) &:= P_0^{\frac{-1}{n+1}} S, \\ P_0 &:= \frac{V_0 H}{\epsilon D_L^0}, \\ \tilde{z} &:= z/H, \\ \tilde{t} &:= t/\tau_0, \\ \tau_0 &:= \frac{\epsilon H}{V_0} P_0^{\frac{1}{n+1}}, \\ \tilde{h} &:= h/H,\end{aligned}\tag{15}$$

where V_0 is the value of V in CRP. The boundary conditions are as follows:

$$\begin{aligned}\frac{\partial \sigma}{\partial \tilde{z}} &= 0 \quad (\tilde{z} = 0), \\ \frac{\partial \sigma}{\partial \tilde{z}} &= -\frac{V}{V_0} \sigma^{-n} \quad (\tilde{z} = 1 - \tilde{h}).\end{aligned}\tag{16}$$

Consider the solution $(\sigma, \tilde{h})$ of Eqs. (15) and (16) with the uniform distribution of σ as initial condition. If the initial value of σ is sufficiently large, σ should immediately transition to a quadratic function corresponding to Eq. (10) and maintain this distribution shape until near the transition. If $\tilde{h}$ turns positive at the time $\tilde{t} = \tilde{t}^*$, this corresponds to the transition to the FRP, so the transition point is given by the following formula:

$$\begin{aligned}\langle S \rangle^* &= \sigma^* P_0^{\frac{1}{n+1}}, \\ \sigma^* &:= \int_0^1 \sigma(\tilde{t}^*, \tilde{z}) d\tilde{z}.\end{aligned}\tag{17}$$

Relative change in drying rates in FRP is given as

$$\begin{aligned}\frac{V}{V_0} &= \frac{\langle \sigma \rangle'(\tilde{t})}{\langle \sigma \rangle'(\tilde{t}^*)} \\ \langle \sigma \rangle'(\tilde{t}) &:= \frac{d\langle \sigma \rangle(\tilde{t})}{d\tilde{t}} \frac{1}{\tau_0} \\ \langle \sigma \rangle(\tilde{t}) &:= \int_0^1 \sigma(\tilde{t} \geq \tilde{t}^*, \tilde{z}) d\tilde{z}.\end{aligned}\tag{18}$$

We can also rewrite the approximate solution Eq. (14) in the nondimensionalized variable $\sigma, \tilde{h}$ as follows:

$$\frac{\langle S \rangle}{\langle S \rangle^*} = \frac{\langle \sigma \rangle}{\sigma^*} = (1 - \tilde{h})^{\frac{n+2}{n+1}} \left(\frac{V}{V_0} \right)^{\frac{1}{n+1}}.\tag{19}$$

As discussed for nonhierarchical granular beds, the Kelvin effect of the microscopic pores is not significant because of their considerable grain size [8] and the observed water saturation [4]. Therefore, it is reasonable to assume that in the FRP, a dry region expands on the granular bed surfaces; the falling rate is due to the diffusion of vapor through the growing thickness of the dry

region (h). Then, we can express the drying rate in the same way as in the case of a nonhierarchical granular bed [8]:

$$\begin{aligned}\frac{V}{V_0} &= \frac{1}{1 + N\tilde{h}} \\ N &:= \frac{H\tau_g}{\delta\epsilon},\end{aligned}\tag{20}$$

where ϵ is the porosity of the granular bed, τ_g is the diffusion tortuosity in the dry region, and δ is the thickness of the boundary layer in air neighboring the water surface exposed at the sample surface.

To compare the three equations obtained above, Eqs. (8), (19), and (20), with experimental results in FRP, let us eliminate $\tilde{h}$. First, Eqs. (19) and (20) can be jointly constructed as follows:

$$\frac{\langle S \rangle}{\langle S \rangle^*} = \frac{\langle \sigma \rangle}{\sigma^*} = (1 - \tilde{h})^{\frac{n+2}{n+1}} (1 + N\tilde{h})^{-\frac{1}{n+1}}.\tag{21}$$

This formula can be regarded as a function that gives a numerical solution to $\tilde{h}$ given the ratio of saturation to the transition point ($\langle S \rangle / \langle S \rangle^*$) in the FRP. In addition, by jointly establishing Eqs. (8) and (20), we can obtain the following equation giving the time derivative of $\langle \sigma \rangle$:

$$\frac{\epsilon H}{V_0} \frac{d}{dt} \langle S \rangle = \frac{d}{d\tilde{t}} \langle \sigma \rangle = -\frac{1}{1 + N\tilde{h}}.\tag{22}$$

Generally, it is easy to obtain numerical solutions from a function that gives such a time derivative, and we used the `solve_ivp` function of SciPy [9] in this study. In addition, a plot showing the relationship between $\langle S \rangle$ and V can be easily obtained by giving common numerical values to $\tilde{h}$ in both Eqs. (20) and (22).

Based on the Young-Laplace equation, capillary pressure, P_c , must be proportional to the surface energy of water, Γ , and inversely proportional to the microscopic pore radius, which is proportional to d , where $\tilde{P}_c$ is a nondimensional function of S and is independent of d .

Permeability, K , must be proportional to the square of the microscopic pore radius.

Therefore, D_L can be rewritten as follows:

$$D_L = \tilde{D}_L^0 \frac{\Gamma}{\eta} S^n d,\tag{23}$$

where $\tilde{D}_L^0$ is a nondimensional constant that does not depend on d . Substituting this into the solution Eq. (17) of the power-law model gives the following:

$$\begin{aligned}\langle S \rangle^* &= \tilde{P}_0^{\frac{1}{n+1}} \tilde{D}_L^0^{-\frac{1}{n+1}} \sigma^* \\ \tilde{P}_0 &:= \frac{\eta V_0 H}{\epsilon \Gamma d}.\end{aligned}\tag{24}$$

Using Eq. (15), we can describe the spatially averaged water saturation in FRP as

$$\langle S \rangle = P_0^{-\frac{1}{n+1}} \langle \sigma \rangle(\tilde{t}).\tag{25}$$

We set the approximation function of $\langle \sigma \rangle$ in an exponential form as

$$\langle \sigma \rangle \sim A' \exp(-\tilde{t}/\tilde{\tau}) + C'.\tag{26}$$

The measured drying time constant, τ , is expressed as follows:

$$\tau = \tilde{\tau} \frac{\epsilon H}{V_0} P_0^{\frac{1}{n+1}}.\tag{27}$$

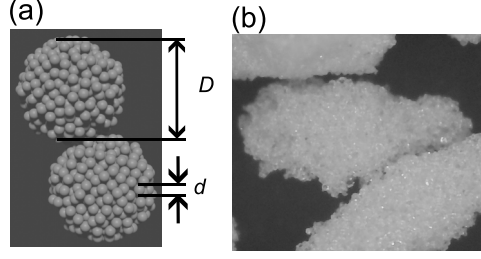


FIG. 2. (a) Conceptual model of spherical agglomerates with the size of D . The primary particle size is denoted as d . (b) Photograph of some agglomerates of hi-d18(S) sample. The width of this picture corresponds to $420\ \mu\text{m}$.

From Eqs. (17) and (27), we can obtain a proportionality as

$$\tau \frac{V_0}{\epsilon H} : \langle S \rangle^* = \tilde{\tau} : \sigma^* = \text{const.} \quad (28)$$

In the following sections, we will evaluate this power-law model by adding new experimental data, comparing them with this model, and considering appropriate values for n .

III. EXPERIMENT

A. Sample preparation

The hierarchical and nonhierarchical granular media and the measurement system used in this study were the same as those used in previous studies [10]. There are two essential length scales. One is the diameter of primary particles, d , and the other is the diameter of secondary particles, D as illustrated in Fig. 2(a). Glass beads were used as primary particles. The nonhierarchical samples, denoted as “non-hi,” are commercially available glass beads with typical diameters of 5, 18, and $400\ \mu\text{m}$ (Potters-Ballotini Co., Ltd.: EMB-10, P-001; As One Corp.: BZ-04). The last sample, non-hi(400), is used only for porosimetry because its pore size is so large that we can not neglect the effect of gravity on liquid water transport.

We obtained sintered glass by heating the dried non-hi $18\ \mu\text{m}$ particles in an alumina ceramic container for 50 to 90 minutes in a muffle furnace set at 650°C . A mortar was used to obtain agglomerates as the secondary particles by lightly breaking the sintered glass. We prepared different types of hierarchical granular samples, denoted as “hi-d18,” by sieving and named them based on the sieve used: XS: $74 - 250\ \mu\text{m}$, S: $250 - 840\ \mu\text{m}$, M: $840 - 2000\ \mu\text{m}$ and L: $2000 - 4760\ \mu\text{m}$. A photograph of some agglomerates of the hi-d18(S) sample is shown in Fig. 2(b) as an example.

Table I shows the increase of the porosity, ϵ , in hi-d18 from non-hi samples due to the macroscopic pore. Because we can neglect neck formation between the connecting beads, the pore

TABLE I. Primary and secondary particle sizes, porosity, and microporosity of the samples.

Sample name	$d(\mu\text{m})$	$D(\mu\text{m})$	ϵ	ϵ_m
non-hi(5)	5		0.51	
non-hi(18)	18		0.46	
non-hi(400)	400		0.40	
hi-d18(XS)	18	74–250	0.71	0.25
hi-d18(S)	18	250–840	0.72	0.24
hi-d18(M)	18	840–2000	0.69	0.27
hi-d18(L)	18	2000–4760	0.64	0.31

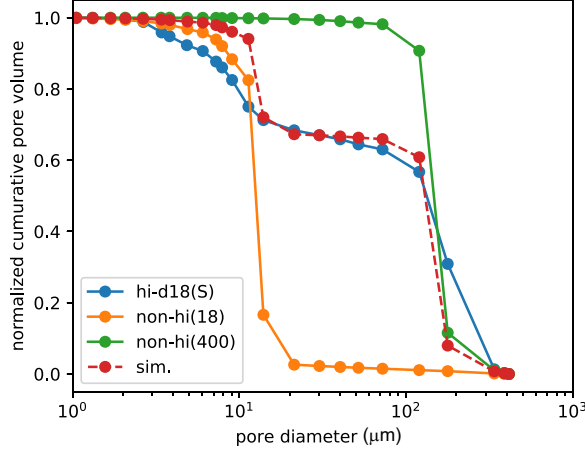


FIG. 3. The normalized cumulative pore volume of hi-d18(S) and non-hi samples are plotted together with non-hi samples with diameters of 18 and 400 μm . The red dashed curve denoted as “sim.” shows their linear combination.

structure in the agglomerates is equivalent to that of the nonhierarchical raw material before heating. Then, we can calculate microscopic porosity, ϵ_m , as

$$\epsilon_m := \epsilon_{\text{non}}(1 - \epsilon)/(1 - \epsilon_{\text{non}}), \quad (29)$$

where ϵ_{non} is the porosity of the corresponding nonhierarchical granular bed, “non-hi(18).” Details of the sample preparation process are available in previous reports [10,11].

Figure 3 shows the normalized cumulative pore volume of hi-d18(S) together with non-hi samples with diameters of 18 and 400 μm measured by mercury intrusion porosimetry (MIP). The non-hi samples for porosimetry were exceptionally sintered in the same way as the hierarchical samples without being broken so that they could be measured with MIP. Specifically, the two representative pore diameters were approximately two to four times smaller than the constituent particle sizes, d and D [10,12]. The larger pores corresponding to D are called macroscopic pores and constitute mainly interagglomerate pores. In contrast, the smaller pores corresponding to d are called microscopic pores and constitute mainly the pore space within the agglomerates [10]. The red dashed curve shows a simulation of the nonhierarchical samples’ data, which is a linear combination of macroscopic and microscopic porosity. The macroscopic pore introduced by breaking the sintered glass is found to be almost equivalent to spherical granular bodies with a diameter of 400 μm . The distribution of microscopic pore size after breaking appears to have changed slightly compared to before destruction, but in this study, it is considered to be equivalent.

B. Experimental setup

We poured 100 g of hi-d18 and non-hi samples into a stainless-steel sieve mounted in a cylindrical frame with a sheet of filter paper at the bottom to construct a granular bed. The inner diameter of the container was 75 mm. One hundred g of water was sprayed on the granular bed for 4 minutes. The weight of the hydrated granular bed, including the container, and the weight of the drained water were logged using two electronic balances (A&D Co., Ltd.: EK-300i) and a PC with a sampling interval of 5 seconds. These systems, excluding the PC, were maintained in an incubator (Isuzu Seisakusho Co. Ltd.: VTR-115) at 35 $^{\circ}\text{C}$. This measurement system was the same as that used in previous studies [10].

Because the drainage stopped a few seconds after water spraying was completed, we changed the sampling interval to 1 minute to measure the weight changes of the granular beds over an extended

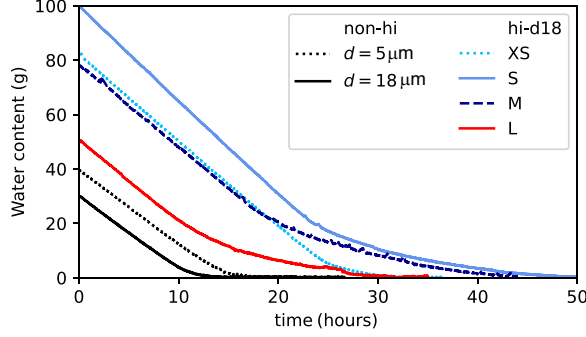


FIG. 4. Time variation of water content in non-hi and hi-d18 samples during drying.

period due to drying. The relative humidity in the incubator during drying fluctuated in the 16 to 32% range.

The drying rate can be determined from the time variation of the weight of the granular beds. Because the circulating air in the incubator hits the electronic balance, the output value of the electronic balance exhibits a more significant fluctuation than its specification. We performed the same runs three times to check the reproducibility of the data and to reduce the effect of this fluctuation under all experimental conditions.

IV. RESULTS AND ANALYSES

A. Drying curves

Figure 4 shows an example of the temporal variation in water content in granular beds due to drying. Initially, the hierarchical granular beds retained more water than the nonhierarchical granular beds because of the water retention of the macroscopic pores [10]. Because most of the water content decreases at a constant rate that is almost independent of grain size and structure, it is clear that the most significant factor in the difference in the time required to dry the granular beds (of the order of hours) among the samples is the initial amount of water retained.

B. Drying rate curves

We performed least-squares linear fitting to the 30-minute data ($t - 15$ to $t + 14$) to obtain the slope of Fig. 4 with reduced noise. Because the top and bottom surfaces of the granular bed were open, we calculated the drying rate defined as the equivalent liquid velocity, V_{raw} , as a function of time, t by dividing the slope by $2\rho a$, where a is the bed area. The effects of humidity fluctuations were then corrected. The humidity measured at each time, $RH(t)$, was used to calculate the drying rate at a typical humidity of 25%. A preliminary experiment confirmed that the drying rate from bulk water is proportional to $100 - RH$ in our incubator. Therefore, we can calculate the drying rate from our samples, $V(t)$, at a typical humidity as follows:

$$V(t) = V_{\text{raw}}(t) \frac{100 - 25}{100 - RH(t)}. \quad (30)$$

We can also calculate the spatially averaged water saturation of the entire granular bed, $\langle S \rangle$, from the granular bed packing fraction. Nonhierarchical granular beds with large values of d (2000 and 3000 μm) did not exhibit a clear CRP. This trend is expected because the capillary lengths of these grain sizes roughly correspond to the sample height. Other nonhierarchical and hierarchical granular beds show a constant drying rate, at least in the region of $\langle S \rangle > 0.5$. Therefore, we adopted the average drying rate of CRPs (V_0) as the average drying rate within a range of $0.5 < \langle S \rangle < 0.8$.

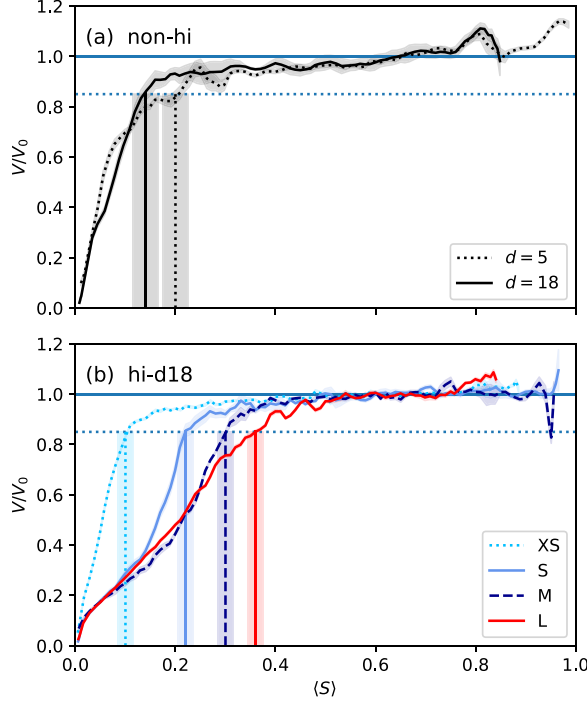


FIG. 5. Drying rate curves normalized by the initial drying rates, V_0 for non-hi (a) and hi-d18 (b) as a function of spatially averaged water saturation, $\langle S \rangle$. We performed bin averaging with a constant bin width of 0.05 for non-hi and 0.03 for all others to obtain these values with less fluctuation. However, the centers of the bins were swept with a minor step of 0.01 to smooth the plotted curve. In addition, the vertical dashed lines indicate the transition points at which the drying rate reaches 0.85 times the average drying rate of CRP. Thinly painted areas on both sides of each dashed line indicate the standard deviation of bin averaging.

For non-hi ($d = 3000 \mu\text{m}$), there is no data in such a region, and we assume V_0 to be that of non-hi ($d = 2000 \mu\text{m}$). The obtained values of V_0 were within a narrow range of 0.29 to 0.35 mm/hour.

The curves in Fig. 5 show the normalized drying rate V/V_0 as a function of $\langle S \rangle$. To reduce the remnant fluctuation in the drying rate curves, we performed bin averaging with a constant bin width on the abscissa ($\langle S \rangle$). We set the bin width to 0.05 for non-hi and 0.03 for hi-d18. The solid and dashed curves indicate the average values. The widths above and below the curves represent the standard deviations. The drying rate fluctuated over the standard deviation. This fluctuation is not likely due to the noise of the electronic balance but to some random characteristics of the sample's evaporation or fluctuations of the air velocity in the incubator.

In addition, FRP was defined as a period when the drying rate is less than 0.85 times V_0 . The vertical dashed lines in Fig. 5 indicate transition points. The thinly painted area on both sides of each dashed line indicates half of the bin width as the transition point error.

In non-hi, the determined transition points, $\langle S \rangle^*$, decrease monotonically with increasing d . This trend is similar to that reported in [8] for granular beds from micrometer to nanometer scale. It is plausible that the capillary pressure is inversely proportional to the particle size, whereas water permeability is proportional to the square of particle size. In coarser granular materials, the significant permeability continues to lower the water saturation, facilitates the water supply to the surface, and maintains CRP.

The transition points of hi-d18 showed a monotonous increase with D .

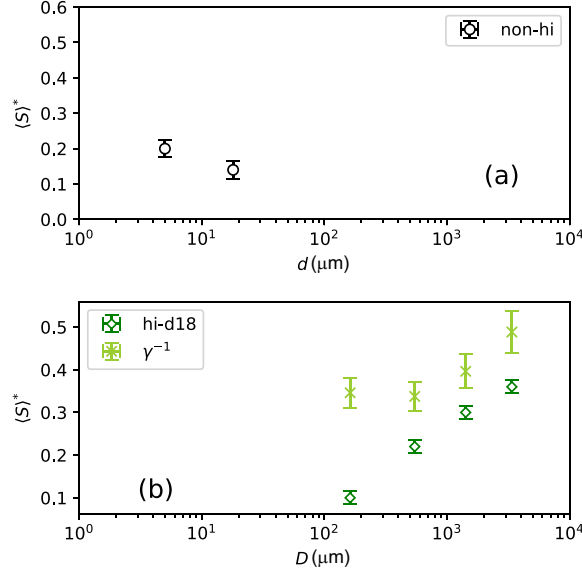


FIG. 6. Transition points ($\langle S \rangle^*$) and particle sizes (d and D) for hierarchical and non-hierarchical granular beds. Panel (a) shows non-hi with the x axis of the primary particle size, d . Panel (b) shows hi-d18 with the x axis of the secondary particle size, D . For comparison, γ^{-1} are shown.

C. Transition point and drying timescale

We now proceed with further analysis by introducing the volume fraction of microporosity to the total porosity, $\gamma^{-1} := \epsilon_m/\epsilon$. When the water saturation is equal to γ^{-1} , if the water flow from the macroscopic pores to the microscopic pores owing to the capillary pressure difference is sufficiently rapid, the macroscopic pores are considered almost empty, and the microscopic pores are fully saturated. In addition, when the water saturation was less than γ^{-1} , $\gamma \langle S \rangle$ means the degree of water saturation in the microscopic pores. For nonhierarchical granular beds, $\gamma^{-1} = 1$.

The calculated γ^{-1} values are shown in Fig. 6. We adopt the mid-range of the agglomerate size as the representative value, D , of hi-d18. For all the hierarchical granular beds, the transition points, $\langle S \rangle^*$, were smaller than γ^{-1} . This means that when $\langle S \rangle$ reaches γ^{-1} , the water flow to the bed surface is sufficient to maintain CRP. That is, the water in the microscopic pores is not completely isolated by the air-filled macroscopic pores.

We can define the saturation time, t^* , when saturation reaches the transition point, $\langle S \rangle^*$. Figure 7(a) shows one of the drying curves during the FRP, $t > t^*$, of M-sized samples with $d = 18$ μm theoretical curves and the fitting curve of an exponential function:

$$\langle S \rangle = A \times \exp(-t/\tau) + C, \quad (31)$$

where A , C , and τ are the fitting parameters. The theoretical curves are calculated with Eqs. (19) and (20). The residue of the fitting curve is not like random noise but shows a systematic change (b). However, it is smaller than the residue of any of the theoretical curves. The change in saturation appears to be approximated by an exponential function better than our model. The obtained values of τ are a measure of the drying time in the FRP, which we call the drying time constant.

Figure 8 shows the correlation of the drying time constant, τ , to the total and the microscopic pore saturation at the transition, $\langle S \rangle^*$ and $\gamma \langle S \rangle^*$. The transition point, $\langle S \rangle^*$, of hierarchical granular bodies is on a different trend from nonhierarchical granular bodies, while the spatially averaged

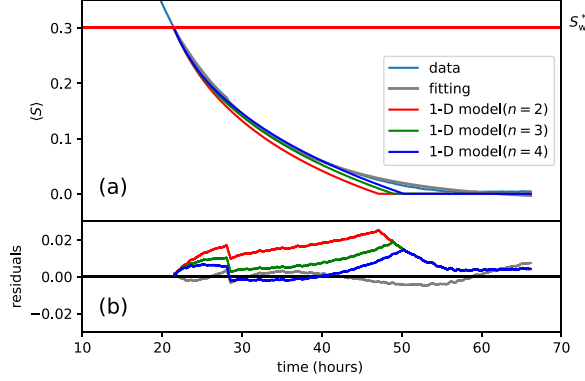


FIG. 7. Time validation curve of the second run of the M-sized sample of hi-d18 as an example of drying time constant, τ , determination. (a) The gray curve is optimized with the function: $\langle S \rangle = A \times \exp(-t/\tau) + C$ to fit the experimental data, a blue solid curve. Dashed curves are theoretically calculated with Eqs. (21) and (22). (b) Residue for the optimization and theoretical curves.

microsaturation at the transition, $\gamma \langle S \rangle^*$, appears to be on the same trend as the transition point of nonhierarchical granular bodies. This result suggests that, for hierarchical granular bodies, microscopic saturation is more essential for the transition to FRP and for the drying rate in FRP than saturation.

V. DISCUSSIONS

A. Drying rate

As shown in Fig. 5, we observed CRP and FRP in almost all hierarchical granular beds. Considering the typical humidity in the chamber of 25%, the initial drying rate during CRP, V_0 ,

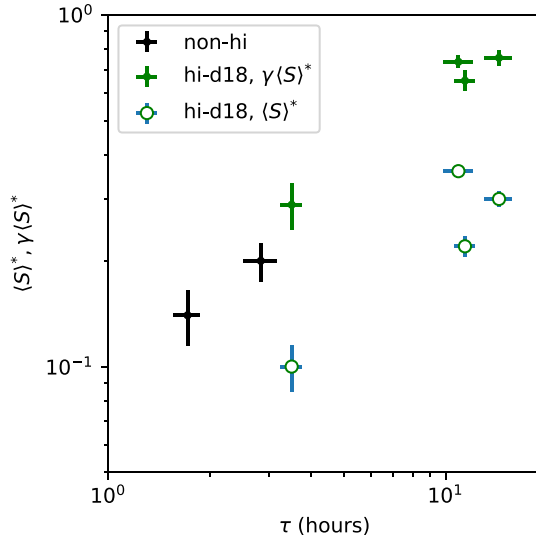


FIG. 8. Total and microscopic pore saturation at the transition, $\langle S \rangle^*$ and $\gamma \langle S \rangle^*$ are plotted versus the drying time constant, τ .

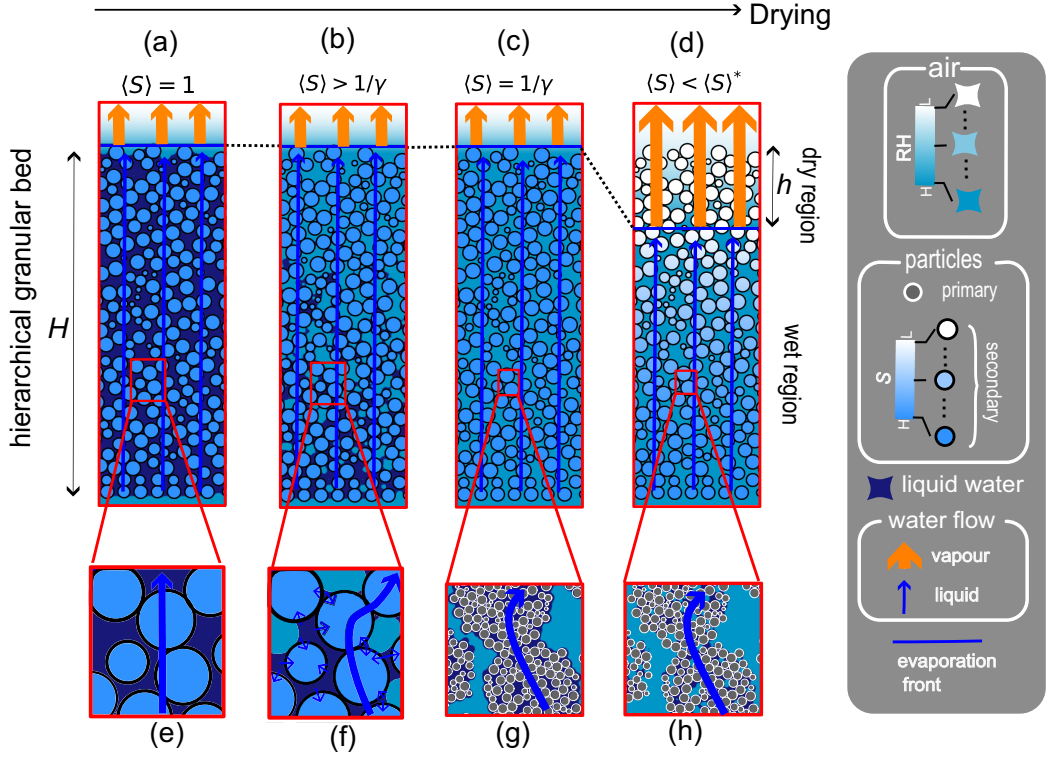


FIG. 9. Conceptual diagram of the drying process of a hierarchical granular bed as cross-section view. (a)–(c), (e)–(g) represent CRP, while (d) and (h) represent FRP. At first, both macroscopic and microscopic pores are filled with water, (a) and (e). At this stage, liquid water flow can path through both pores. At $\langle S \rangle > 1/\gamma$, macroscopic pore water between secondary particles remains, (b), but is being absorbed into the microscopic pore by the capillary pressure gradient created by the upward liquid water flow as shown in (f). At $\langle S \rangle = 1/\gamma$, all liquid water is present in the microscopic pore, which is saturated by water. From this point onward, liquid water flow is limited in the microscopic pore and the microscopic pore becomes unsaturated and less permeable as shown in (g) and (h). In FRP, the microscopic pore in secondary particles in the dry region is dried and does not provide a pathway for liquid water, but does provide a pathway for water vapor diffusing from the evaporation front.

is given as [8]:

$$V_0 = \frac{0.75\rho_0 D_V}{\rho\delta}, \quad (32)$$

where $\rho_0 = 40 \text{ g/m}^3$ is the density of water vapor at the temperature of 35°C , and $D_V = 2.7 \times 10^{-5} \text{ m}^2/\text{s}$ is the diffusion coefficient of water vapor at 35°C . Based on the experimental values of V_0 , the values of δ were calculated within a range of 8.3 to 9.9 mm. The narrow range indicates slight variations in air velocity in the incubator during the measurement. In addition, we observed no significant decrease in the drying rate in the saturation range of $\langle S \rangle > \gamma^{-1}$. This implies that the drying rate in CRP can be maintained only by water vapor diffusion from microscopic pores exposed to the granular bed surfaces. This idea is consistent with the fact that the estimated macroscopic pore size, a measure of the size of blank space of the vapor supplying microscopic pore, is about $D/4 - D/2$, which is smaller than the boundary layer thickness, δ . In Fig. 9, we illustrated the distribution and flow of water at each step based on our experimental results.

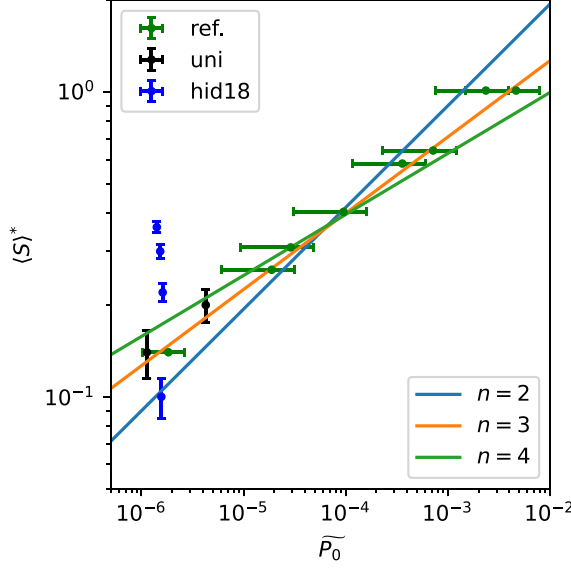


FIG. 10. Log-log plot with $\tilde{P}_0$ on the horizontal axis and $\langle S \rangle^*$ on the vertical axis. The green dots (ref.) are data read from a figure in Ref. [8]. However, $\langle S \rangle^*$ was read directly from the drawing. Since our sample (uni and hi-d18) evaporated from both the top and the bottom sides, half of the sample height was assigned to H . Also, as the values of H and V_0 are only given as ranges in Ref. [8], we used error bars to represent their effects. The three straight lines (blue, orange, and green) correspond to assumed n values (2,3,4); the three lines (blue, orange, and green) correspond to the assumed n values (2,3,4). However, the proportionality constants were determined to pass near the data points other than hi-d18.

B. Power law of the transition point

The relationship between $\tilde{P}_0$ and $\langle S \rangle^*$ is shown in Fig. 10 based on our data and those of Thiery *et al.* Since evaporation occurred from both the top and bottom sides of our sample, half of the sample height was substituted for H . As the values of H and V_0 are not given for each sample in the report [8], the effect of the distribution of the given values is expressed as an error bar.

Although $\tilde{D}_L^0$ still depends on the pore structure, it has a narrow range (0.42 – 0.51) of porosity ϵ for nonhierarchical samples, so we assume $\tilde{D}_L^0$ to be a constant for non-hi and “ref.” Three lines in Fig. 10 show the model equation $\langle S \rangle^* \propto \tilde{P}_0^{\frac{1}{n+1}}$ assuming 2, 3, and 4 as candidate values of n passing near the data points except for data of hi-d18. The data points other than hi-d18 lie approximately on the line corresponding to $n = 3$. Since this value is also consistent with Eq. (2), “ n ” is assumed to be three thereafter in this paper. From the slope, the following empirical values are obtained:

$$\tilde{D}_L^0{}^{-\frac{1}{4}} \sigma^* = 4.0. \quad (33)$$

In order to examine the validity of these values, numerical solutions for Eqs. (15) and (16) were obtained (Appendix A). We can obtain $\sigma^* = 0.69$. In this Appendix, it is also confirmed that Eqs. (19) and (21) are good enough approximations for the calculations. Appendix B also provides estimates of $\tilde{D}_L^0$ calculated from existing data. Substituting these into the left-hand side of Eq. (33) yields a value of 2.4. This value is not fully consistent with the value on the right-hand side of Eq. (33), suggesting that our model is not sufficiently precise to explain the experimental results quantitatively, and further improvements are needed. Substitution of the value into Eqs. (14) and (17), we can see that m , a constant value defined in Eq. (13), is 6.6 and confirm that it is consistent with its definition, “not sufficiently smaller than 1.”

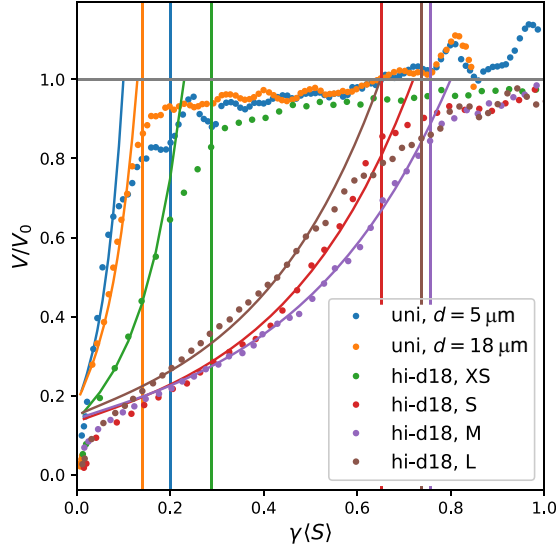


FIG. 11. The dotted line represents the falling drying rate at FRP of our sample (uni, hi-d18). The horizontal axis is the microscopic saturation. The vertical solid lines show the transition ($\gamma(S)^*$) to FRP in the corresponding color data. The curves show the theoretical values with their transition point set to fit the corresponding color data during FRP.

The larger values of the transition point for the hi-d18 samples mean small values of $\widetilde{D}_L^0$, which may be due to a reduction in their permeability due to the dried macroscopic pore.

C. The drying rate in the FRP

As seen in Fig. 7, Eqs. (21) and (22) do not fully reproduce the drying process in FRP. This is presumably due to the fact that the spatial uniformity of D_L , which is assumed as a precondition for these equations, is not fully established. Before adding inhomogeneity to the model, let us see how far Fig. 5 can be reproduced with a uniform model.

In Fig. 11, the experimental data for the saturation dependence of the drying rate were compared with a theoretical curve with $n = 3$. However, when calculating the theoretical curve, the transition points were set to follow the downward convex behavior of the experimental data at low saturation instead of adopting the experimental data. Both the theoretical and experimental curves are characterized by a downward convex curve that asymptotically approaches $1/(1 + N)$ at low saturation. In addition, two main differences can be observed. First, at very low saturations below 0.1, only the experimental values show a reduction in the drying rate beyond the asymptote. This may be due to the fact that at these very low saturations, most of the water is present in isolated bridges between the particles, so the assumption that one-dimensional water vapor diffusion is the rate-limiting factor Eq. (20) does not hold. Second, compared to the theoretical curve, the experimental data do not show a rapid decrease in the drying rate around the transition point. Such a degradation of the curve is common in many types of spectroscopy and is known as inhomogeneous spectral broadening, so our case is analogous to a change originating from inhomogeneity. The bin averaging process used to obtain the experimental values should have resulted in a bin-width-induced degradation, which should also be responsible for both differences.

To qualitatively assess the effects of inhomogeneity, horizontal inhomogeneity of the transition point is introduced. In this case, saturation is no longer a one-dimensional distribution. For simplicity, horizontal advection and diffusion are ignored. In this case, the time variation of the water saturation $\langle S \rangle$ can be written by a linear combination of the theoretical curves presented in Fig. 7 as

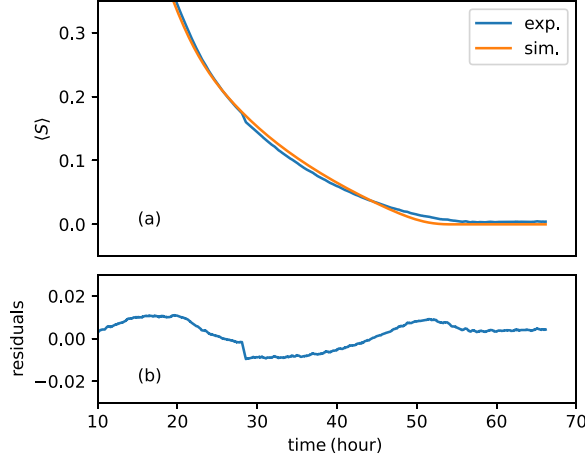


FIG. 12. (a) Time variation of water saturation in the M-sized sample of hi-d18. We can better fit the simulated curve, “sim.”, to the experimental data, “exp.”, by introducing an inhomogeneous transition point. The assumed distribution of transition points is shown in Fig. 13 as a bar chart. (b) The residuals mean the difference between “sim.” and “exp”.

follows:

$$\langle S \rangle(t) = \sum_{i=0}^{i_{\max}} p_i \overline{S_{w,i}}(t), \quad (34)$$

where p_i is the probability that the transition point takes S_w^* and $\overline{S_{w,i}}$ is the time variation of water saturation in that case. Figures 12 and 13 show the results of the calculation in which the normal distribution is introduced as the first trial function for inhomogeneous distribution as an example. The experimental data of interest are the M-sized sample of hi-d18 used in Fig. 7. The assumed distribution of transition points is shown in Fig. 13 as a bar chart. Here, $\gamma \langle S \rangle^* < 1$ was assumed

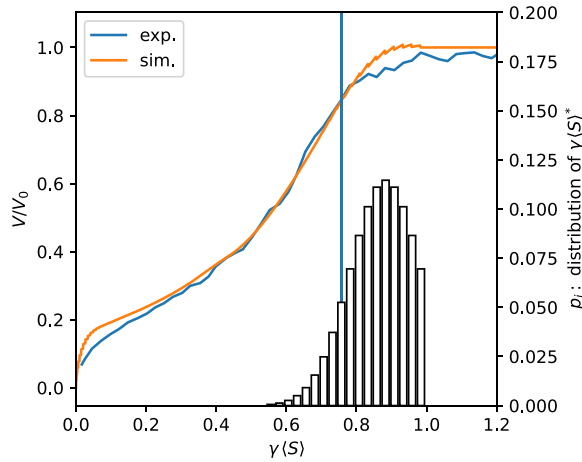


FIG. 13. The dependence of the drying rate on water saturation in the M-sized sample of hi-d18. We can better fit the simulated curve, “sim.”, to the experimental data, “exp.”, by introducing an inhomogeneous transition point. The assumed distribution of transition points is shown as a bar chart.

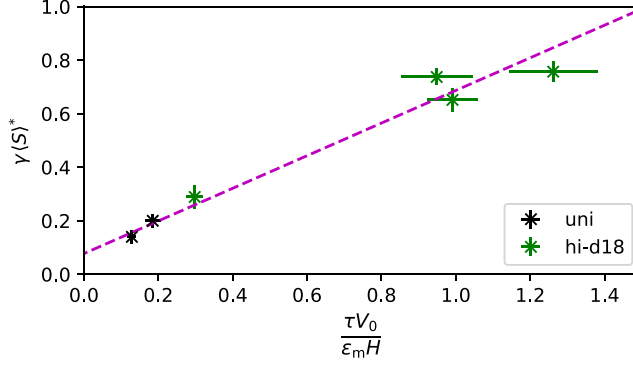


FIG. 14. Nondimensionalized drying time constant $\frac{\tau V_0}{\epsilon_m H}$ and microscopic pore saturation at transition $\gamma \langle S \rangle^*$. The straight line is the fitting line obtained by the weighted least squares method.

in order to maintain the condition that the macroscopic pore is dry at the transition point. It can be confirmed that the time variation of saturation, Fig. 12, and the saturation dependence of the drying rate, Fig. 13, can be brought closer to the experimental results by introducing heterogeneity at the transition point. It was also found that the introduced inhomogeneity can also explain the lower drying rate beyond the asymptote in the low saturation region. Under the assumption of horizontal heterogeneity, areas with small transition points dry out completely earlier than areas with large transition points. In other words, the reduction of the drying rate beyond the asymptote may be attributed to the fact that the evaporating area becomes smaller at low saturation.

D. Drying time constant

With the definition of γ , Eq. (28) can be rewritten as

$$\tau \frac{V_0}{\epsilon_m H} : \gamma \langle S \rangle^* = \tau \frac{V_0}{\epsilon H} : \langle S \rangle^* = \text{const.} \quad (35)$$

Figure 14 shows the linear relationship described in Eq. (35) with a small intercept in the axis of γS at less than 0.1. The saturation degree corresponds to the residual water, S_r , measured for many granular materials in this region, so the slight discrepancy with this model may be due to the fact that the model ignores S_r . We therefore propose the following empirical formula from the approximate straight line in Fig. 14:

$$\gamma \langle S \rangle^* = 0.61 \times \tau \frac{V_0}{\epsilon_m H} + 0.08, \quad (36)$$

where the two constants (0.61 and 0.08) would depend on the structure of the micropore, the former being mainly influenced by the heterogeneity of the pore structure and the latter by S_r .

VI. CONCLUSION

The proposed model, as presented in Eqs. (15) and (16), assuming $n = 3$, is not only consistent with the empirical formula for the transition points of Thiery *et al.* [8] Appendix A examines the difference between approximate and numerical solutions. It is negligible with respect to the error levels observed in the present experiments.

Based on the model, we could introduce a new parameter, $\tilde{P}_0$, to successfully align the transition points of their data and our data for nonhierarchical samples in a single line in log-log plots within a margin of error. Although we can estimate the proportionality constant from existing data on

capillary pressure and water permeability with the same order of magnitude as that of our model in Appendix B, the difference is not negligible.

On the other hand, in the middle range of FRP, experimental data of the drying rate show similar behavior to the theoretical curve approaching the minimum value of the model, $1/(1+N)$. Differences between the model and the experiment are evident near the transition point and in the region of low micropore saturation below 0.1, but it is clear that the discrepancies can be reduced for the case of hierarchical samples by assuming a simple case where the transition point has horizontal inhomogeneity.

The correlation between the experimental drying time constant and micro-pore saturation was obtained by making the drying time constant dimensionless based on our model, and a universal empirical equation [Eq. (36)] was obtained.

The validation of the model carried out in this study is not yet sufficient. Direct proof of the water transport model in hierarchical and nonhierarchical hydrophilic granular media requires the expansion of the dry region to be observed with MRI ([8]; [13]) and dyes ([5–7]), together with quantification of the effect of reduced water permeability in the hierarchical bed. The differences in the transition point may also be caused by sample inhomogeneities. However, there are two issues that remain to be addressed in order to deepen the discussion on the transition point. The first is the method of defining the transition point. In this study, the transition point was defined as the point at which the drying rate was 0.85 times the CRP. This definition was adopted to reduce the effects of measurement errors, but it is arbitrary. In order to redefine the transition based on the sample inhomogeneity, experiments must be conducted to quantitatively clarify the effect of heterogeneity. In this study, the hierarchical granular material that suggested heterogeneity has no method for predicting the rate of decrease in water permeability due to macroscopic pores, so at present, $\widetilde{D}_L^0$ is unknown. In addition, although the value of $\widetilde{D}_L^0$ was estimated for our non-hi samples, the effect of S_r cannot be ignored, and it is thought that the application of our model is close to the limit. Therefore, we propose that the next appropriate step is to measure the drying rate of nonhierarchical samples with large $\widetilde{P}_0$ as in the work of Thierry *et al.*, but with further improved accuracy.

As for the dependence of P_c on S , a linear function was used to approximate the van Genuchten curve in the intermediate region. It is known that similar curves can be reproduced by numerical calculations of meniscus size in partially saturated spherical granular [14]. Therefore, the consistency of our model with data from nanoporous samples [8] suggests that the capillary pressure can be determined from the meniscus size in partially saturated pores without considering water films on hydrophilic surfaces of a few nanometers thickness [15]. On the other hand, it has been reported that in monolithic nanoporous silica glasses, no dry regions are observed during the drying process, and the pore water is present in a water-film-like confined state in the low saturation region [13]. As this difference may be attributed to differences in pore geometry, caution should be exercised when extending our arguments to other nanoporous media.

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The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work.

APPENDIX A: NUMERICAL SOLUTION OF OUR MODEL

To obtain the numerical solution, we apply spatial discretization to the differential Eqs. (15) and (16). The thickness of the granular bed ($0 < z' \leq 1$) is divided into $M = 500$. The saturation values, $\sigma_i(t)$, of the i -th regions and the flux between the i -th and the neighboring regions, u_i , satisfy the

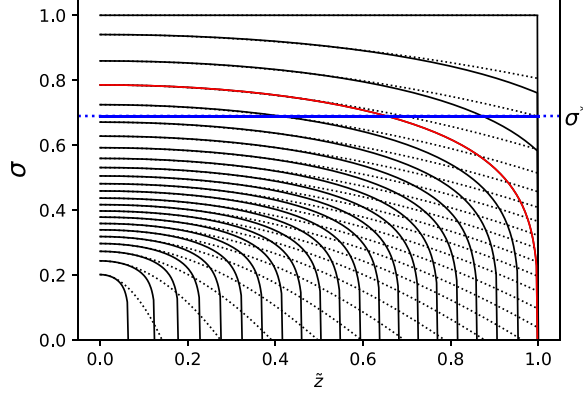


FIG. 15. Black curves show the time variation of the vertical distribution of s . The time intervals between neighboring lines were fixed to be the same value, $0.1\tau_0$. The red curve indicates the distribution at the transition from CRP to FRP. The blue line indicates the transition point, $\sigma^* = 0.689$, defined as the spatial average of σ at the transition. Dotted curves are parabolas fitting to the vertical distributions around the bottom ($\tilde{z} \sim 0$).

following equations:

$$\begin{aligned}
 u_i &= \begin{cases} 0 & (i \geq k+1 \text{ or } i=0), \\ \frac{1}{1+N\tilde{h}} & (i=k), \\ -\left(\frac{\sigma_i+\sigma_{i+1}}{2}\right)^3[(\sigma_{i+1}-\sigma_i)M] & (0 < i < k), \end{cases} \\
 \tilde{h} &= (M-k)/M, \\
 \frac{d\sigma_i}{dt'} &= \begin{cases} -(u_{i+1}-u_i)M & (0 \leq i < k), \\ 0 & (i \geq k), \end{cases} \\
 \sigma_i &= 0 (i \geq k). \tag{A1}
 \end{aligned}$$

We started the numerical simulation with the initial condition of $\sigma_i = 1$ for $i = 0, \dots, M+1$ and $k = M$. The typical value of our samples, 6, was used as N . We used the function “solve_ivp” in an open-source library, “SciPy” [9], which integrates ordinary differential equations with an initial value. We programmed an event to generate when the top cell of the wet region, σ_k , dries out, i.e., reaches the value of 0. If the event occurs, the program updates the boundary conditions by decreasing k .

Figure 15 shows snapshots of σ_i distribution with an interval of $\Delta\tilde{t} = 0.1$. The formation of dry regions is seen beneath the top surface. We averaged σ_i at the transition to get the following value:

$$\sigma^* = 0.69. \tag{A2}$$

Also, we can confirm the spatial distribution of water in the wet region is close to a parabola as predicted, Eq. (10), except for just beneath the evaporating boundary to the dry region.

From Fig. 16, we can confirm that Eqs. (21) and (22) give almost correct solutions of $\langle\sigma\rangle$ without solving Eqs. (15) and (16) directly.

APPENDIX B: EVALUATION OF THE NONDIMENSIONAL DIFFUSION CONSTANT IN NONHIERARCHICAL SAMPLES

There have been many studies on the capillary pressure, P_c [14]. Figure 17 shows the Van Genuchten curves that approximate the experimental results of the dependence of capillary pressure on water saturation for two types of glass beads (GB1 [17] and GB2 [16]). The capillary pressure

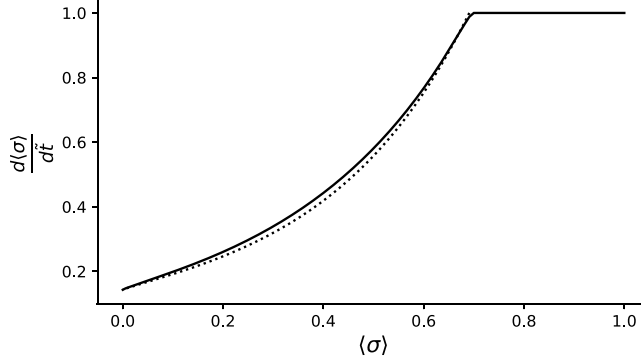


FIG. 16. The solid curve shows the relationship between $\langle \sigma \rangle$ and $d\langle \sigma \rangle / d\tilde{t}$. The dotted curve is a similar plot to the solid curve but is obtained from Eqs. (21) and (22).

increases rapidly as the water saturation approaches a specific value, S_r , and decreases rapidly as the water saturation approaches 1. Here, S_r corresponds to the water in the isolated bridges, called irreversible water or residual water, and has a hysteresis value of less than 0.2. This figure shows that the capillary pressure can be approximated with a straight line in an intermediate range. GB1 and GB2 had different particle size dispersion widths; the former had a value of 10% of the mean diameter and the latter had a value of approximately 30%. The product of pore pressure and average particle size depends on the dispersion width [14]. Since the glass beads used in this experiment had a wide dispersion of about 50%, we regard the value of GB2 as suitable for our data. Similarly, we regard the value of GB1 as suitable for the narrow dispersion samples used in [8]. Because capillary pressure should be proportional to the surface energy of the water, we can describe the dependence of the capillary pressure on water saturation as follows:

$$\frac{dP_c}{dS} = -\frac{\Gamma}{\alpha d}, \quad (\text{B1})$$

where α is a dimensionless quantity. We can obtain the value for our samples from the slope of the line (GB2) in Fig. 17 as

$$\alpha \sim 0.36. \quad (\text{B2})$$

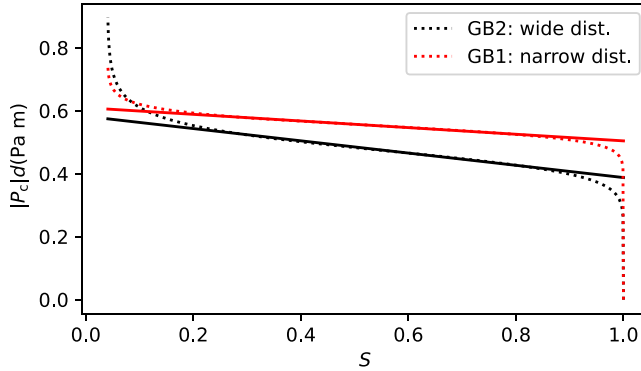


FIG. 17. The two dotted lines are the van Genuchten curves fitted to reported experimental values [16,17] of capillary pressure at various water saturations for nonhierarchical granular materials (GB2, GB1) consisting of two types of spherical glass beads [14]. The solid lines are straight lines that fit the curves at the range of $0.2 < S < 0.8$. The reciprocals of the slopes are -5.1 (GB2) and -9.5 (GB1), respectively.

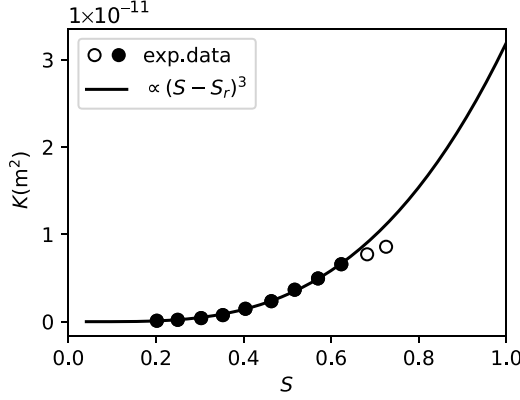


FIG. 18. The two types of circles indicate experimental water permeability from nonhierarchical granular materials consisting of spherical glass beads with an average diameter of $180\ \mu\text{m}$ [18], provided that water viscosity was assumed to be $0.001\ \text{Pa}\cdot\text{s}$. The solid line is the cubic function, $\propto (S - S_r)^3$ fitted to the selected data indicated by the filled circles. We can obtain an optimal value of 0.07 for S_r with the least-squares method.

As shown in Fig. 18, the water permeability, K , of non-hierarchical beds reaches zero when the water saturation approaches S_r . Figure 18 also shows that a cubic function can approximate the permeability. Similarly to the previous report [13], we use the following formula:

$$K = K_0 \left(\frac{S - S_r}{1 - S_r} \right)^3. \quad (\text{B3})$$

Because both samples of Thierry *et al.* and us are granular media of spherical particles, we apply the Kozeny-Carman equation, even though its applicability to nanoporous media is uncertain:

$$K_0 = \frac{\epsilon^3 d^2}{180(1 - \epsilon)^2}. \quad (\text{B4})$$

If we approximate $S \gg S_r$ and $S_r \ll 1$

$$\begin{aligned} K &= K_0 S^3 \\ &= \frac{\epsilon^3}{180(1 - \epsilon)^2} d^2 S^3. \end{aligned} \quad (\text{B5})$$

From Eqs. (B1) and (B5), the following equation is obtained:

$$\frac{K}{\eta} \frac{d}{dS} P_c = - \frac{\epsilon^3 \Gamma}{180(1 - \epsilon)^2 \alpha \eta} S^3 d. \quad (\text{B6})$$

In conclusion, we can successfully confirm Eq. (4) from the experimental data and obtain the following equation:

$$\tilde{D}_L^0 = \frac{\epsilon^3}{180\alpha(1 - \epsilon)^2}. \quad (\text{B7})$$

By substituting $\alpha \sim 0.36$ and $\epsilon \sim 0.49$ into Eq. (B7), we get $\tilde{D}_L^0 \sim 6.9 \times 10^{-3}$.

APPENDIX C: NOMENCLATURE

The notation used in this study is summarized below. Greek and Latin symbols are listed separately in alphabetical order. Each entry includes the symbol's description, unit, and first appearance in the text, which refers to the equation, the table, or the figure where the symbol is introduced. This notation aims to provide clarity and consistency throughout the manuscript.

TABLE II. Greek variables used in this study, arranged in alphabetical order. The table includes their descriptions, corresponding units, and the locations of their first appearances in the text.

Symbol	Description	Unit	First Appearance
α	Parameters related to capillary pressure	ND	Eq. (B1)
γ	$:= \epsilon / \epsilon_m$	ND	Fig. 6
Γ	Surface energy of water	J/m ²	Eq. (1)
δ	Thickness of the boundary layer	m	Eq. (20)
ϵ	Porosity	ND	Eq. (6)
ϵ_m	Microscopic porosity	ND	Table I
ϵ_{non}	porosity of the corresponding “non-hi(18) sample”	ND	Eq. (29)
η	Viscosity of water	Pa s	Eq. (3)
θ	Contact angle of water	rad	Eq. (1)
ρ	Density of water	kg/m ³	Eq. (1)
ρ_0	Density of water vapor	kg/m ³	Eq. (32)
σ	$:= P_0^{\frac{1}{n+1}} S$	ND	Eq. (15)
σ^*	σ at the transition	ND	Eq. (17)
τ	Drying time constant	sec	Eq. (31)
$\tilde{\tau}$	Fitting parameter	ND	Eq. (26)
τ_0	$:= \frac{\epsilon H}{V_0} P_0^{\frac{1}{n+1}}$	sec	Eq. (15)
τ_g	Diffusion tortuosity	ND	Eq. (20)
“ND” means nondimensional			

TABLE III. Latin variables used in this study, arranged in alphabetical order. The table includes their descriptions, corresponding units, and the locations of their first appearances in the text.

Symbol	Description	Unit	First Appearance
a	Bed area	m ²	Fig. 1
A	Fitting parameter	ND	Eq. (31)
A'	Fitting parameter	ND	Eq. (26)
C	Fitting parameter	ND	Eq. (31)
C'	Fitting parameter	ND	Eq. (26)
d	Diameter of primary particles	m	Eq. (2)
D	Diameter of secondary particles	m	Fig. 2
$\langle D_L \rangle_w$	Spatially average of D_L in wet region	m ² /sec	Eq. (12)
D_{Lb}	D_L at the boundary	m ² /sec	Eq. (12)
D_L	$:= \frac{K}{\eta} \frac{dP_c}{dS}$	m ² /sec	Eq. (3)
D_L^0	$:= D_L S^{-n}$	m ² /sec	Eq. (4)
D_{Lo}	D_L at the bottom	m ² /sec	Eq. (12)
D_V	diffusion coefficient of vapor	m ² /sec	Eq. (32)
F(z)	Approximate function of water saturation	ND	Eq. (10)
h	Thickness of the dry region	m	Fig. 1
$\tilde{h}$	$:= h/H$	ND	Eq. (15)
H	Thickness of granular bed	m	Fig. 1

TABLE III. (Continued.)

Symbol	Description	Unit	First Appearance
i	Integer dammy variable	ND	Eq. (34)
K	Permeability of wet region	m ²	Eq. (3)
m	A real constant revealed to be 6.6	ND	Eq. (13)
M	Number of divisions	ND	Eq. (A1)
n	A power index revealed to be three	ND	Eq. (4)
N	$:= \frac{H\tau_g}{\epsilon D_0^0}$	ND	Eq. (20)
P	$:= \frac{V(\beta\epsilon - h)}{\epsilon D_0^0}$	ND	Eq. (14)
P_0	$:= \frac{V_0 H}{\epsilon D_0^0}$	ND	Eq. (15)
$\tilde{P}_0$	$:= \frac{\eta V_0 H}{\epsilon \Gamma d}$	ND	Eq. (24)
P_c	Capillary pressure in wet region	Pa	Eq. (3)
S	Water saturation	ND	Eq. (3)
$\langle S \rangle$	S spatially averaged in entire volume	ND	Eq. (9)
$\langle S \rangle^*$	$\langle S \rangle$ at the transition	ND	Eq. (2)
$\langle S \rangle_w$	Spatially averaged water saturation in the wet region	ND	Eq. (10)
S_b	Approximation of S at the boundary	ND	Eq. (11)
S_o	Approximation of S at $z = 0$	ND	Eq. (10)
S_r	saturation degree corresponds to the residual water	ND	Fig. 18
t	time	sec	Eq. (3)
$\tilde{t}$	$:= t/\tau_0$ (Dimensionless time)	ND	Eq. (15)
V	drying rate as the equivalent velocity of liquid water	m/sec	Eq. (7)
V_0	the value of V in CRP	m/sec	Eq. (15)
z	Upward vertical axis	m	Eq. (5)
$\tilde{z}$	$:= z/H$ (Dimensionless vertical axis)	ND	Eq. (15)
“ND” means nondimensional			

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