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Wetting front dynamics in an isotropic porous medium

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A new approach to the modelling of wetting fronts in porous media on the Darcy scale is developed, based on considering the types (modes) of motion the menisci go through on the pore scale. This approach is illustrated using a simple model case of imbibition of a viscous incompressible liquid into an isotropic porous matrix with two modes of motion for the menisci, the wetting mode and the threshold mode. The latter makes it necessary to introduce an essentially new technique of conjugate problems that allows one to link threshold phenomena on the pore scale with the motion on the Darcy scale. The developed approach (a) makes room for incorporating the actual physics of wetting on the pore scale, (b) brings in the physics associated with pore-scale thresholds, which determine when sections of the wetting front will be brought to a halt (pinned), and, importantly, (c) provides a regular framework for constructing models of increasing complexity.

1. Introduction

The dynamics of wetting fronts in porous media remains the subject of intensive research (Adler & Brenner 1988; Olbricht 1996; Alava et al. 2004). Its main motivation comes, first of all, from a host of important applications, notably in oil recovery, hydrogeology and more recently also in carbon dioxide sequestration, microfluidics and fuel cells. This topic also poses some fundamental questions about the modelling of evolutionary processes in systems with complex topology. In a practically relevant case where the scales of the pore-level and the global flow are well separated, one can use an intermediate scale to introduce averaged macroscopic quantities and apply the modelling approach of continuum mechanics. In this case, for an incompressible viscous liquid invading an isotropic porous medium the averaged flow velocity \mathbf{u} and pressure p, both functions of the position vector \mathbf{r} and time t, satisfy the equation of motion in the form of Darcy's law

$$\mathbf{u} = -(\kappa/\mu)\nabla p, \qquad (\mathbf{r} \in \Omega),$$
 (1.1)

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where κ and μ are the permeability of the porous matrix and the fluid's viscosity, respectively, and Ω is part of the porous medium occupied by the fluid. Darcy's law together with the continuity equation $\nabla \cdot \mathbf{u} = 0$ form a closed system adequately describing, on the macroscopic level ('Darcy scale'), the bulk distributions of p and \mathbf{u} . Combining these two equations, one has that the pressure in the flow domain Ω is harmonic

$$\nabla^2 p = 0, \qquad (\mathbf{r} \in \Omega), \tag{1.2}$$

so that, to determine p, one has to specify two boundary conditions on the part of the boundary of Ω whose location is unknown (i.e. the wetting front; hereafter $\partial\Omega_1$) and one boundary condition on the part whose position is known (hereafter $\partial\Omega_2$, so that $\partial\Omega_1\cup\partial\Omega_2=\partial\Omega$ is the boundary of Ω). Hence on the wetting front, in addition to the kinematic condition

$$\frac{\partial f}{\partial t} + \mathbf{u} \cdot \nabla f = 0, \quad (\mathbf{r} \in \partial \Omega_1),$$
 (1.3)

which specifies the evolution of $\partial\Omega_1$ in terms of its a priori unknown location $f(\mathbf{r},t)=0$, we need to formulate an appropriate dynamic boundary condition for p. If the dynamics of the displaced fluid also needs to be considered, as, for example, in the case of one viscous liquid displacing another, one will still need a dynamic boundary condition relating the two fluids' pressures at the interface.

The main issue in determining the dynamic boundary condition is to what extent the actual physics of wetting on the pore scale is accounted for, and how it is represented, on the Darcy-scale level. In particular, as has been known for a long time (Huh & Scriven 1971; Dussan V. & Davis 1974; Dussan V. 1979), the classical model of fluid mechanics does not allow viscous fluids to spread over a solid surface with a contact angle less than 180°, whereas numerous experiments show that they do (e.g. see Ch. 3 of Shikhmurzaev 2007). The way one chooses to overcome this ('moving contact-line') problem for the menisci that collectively form the wetting front in a porous medium is one of the factors determining how realistic the resulting model will be. At present, this aspect of the wetting front dynamics modelling received almost no attention, even in the approach which considers the porous medium as a network of capillaries and hence is potentially capable of capturing exactly the details of the pore-scale physics (Lenormand et al. 1988; Aker & Måløy 2000; Joekar-Niasar et al. 2010).

An alternative to the topologically transparent sharp-interface approach outlined above is to consider the wetting front as a transition zone where the volumetric concentration of the displaced and displacing fluid change, with no distinction between continuous and discrete phases, and treat this, essentially multiphase, system as a multicomponent one (Richards 1931; Leverett 1941), using a thermodynamic closure to relate the pressure difference between the two fluid phases with saturations and, possibly, other variables (Hassanizadeh & Gray 1993; Mitkov et al. 1998; Deinert et al. 2008). A difficulty in this approach is that, as experiments aimed at

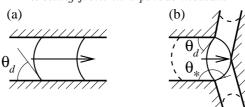


FIGURE 1. Schematic illustration of the meniscus motion in the wetting mode (a) and one of scenarios associated with the threshold mode (b).

determining the closing thermodynamic relationships indicate the need to bring in more and more parameters of state, it is not clear whether or not thermodynamics is an adequate tool to describe this, essentially mechanical, system.

2. The model

In this paper, we introduce a new approach to formulating models describing the propagation of liquidfluid interfaces across porous media based on considering the types (modes) of motion which the menisci that collectively form the free surface go through as they advance across the porous matrix. This approach offers a regular way of building models of increasing complexity accounting for the topological and geometric features of the porous medium. It is worth noting that Darcy's equation (1.1) in the bulk is itself essentially a consequence of the flow profile being approximately parabolic on the pore scale, i.e. it also can be seen as based on a particular type of flow. In this sense, the approach we are developing here considers the boundary conditions conceptually in the same way as the bulk equations.

We will illustrate the new approach using the simple case of a viscous incompressible liquid displacing an inviscid dynamically passive gas from an isotropic homogeneous porous medium. On the pore scale, each meniscus intersects the pore boundary at a 'contact line', forming a certain 'contact angle' with the solid. We can schematically represent the motion of the meniscus on the pore scale as having two principal modes: (i) the wetting mode, where the contact line moves across the pore boundary with negligible variation in the meniscus shape, and (ii) the threshold mode, where the contact line becomes pinned whereas the meniscus deforms until the contact angle it forms with the solid reaches a critical value at which the contact line can move again (Fig. 1). These two modes control the motion as each of them is capable of bringing individual menisci and hence the wetting front as a whole to a halt. In the wetting mode this can happen when the contact angle becomes equal to the equilibrium one, so that the meniscus no longer needs to move to reach an equilibrium state, whereas in the threshold mode, where the contact line is pinned, the flow stops when the pressure building on the meniscus (later referred to as $\bar{p}|_{\partial\Omega_1}$) is insufficient to break through the threshold.

Macroscopically, for the pressure on the wetting front measured with respect to the (presumed constant) pressure in the displaced gas one has

$$p|_{\partial\Omega_1} = A_1 p_1 + A_2 p_2, \tag{2.1}$$

where p_1 , p_2 are the averaged pressures and A_1 , A_2 are the spatio-temporally averaged fractions of the unit area of the free surface corresponding to the two types of motion $(A_1 + A_2 = 1)$. Importantly, A_1 and A_2 do not have to remain constant as the wetting front propagates, and they are yet to be specified.

We will begin by considering the wetting mode. For simplicity, we will represent the pore where the wetting motion takes place as having a circular cross-section. Then, for low capillary and pore-scale Bond numbers, the meniscus will have the shape of a spherical cap and

$$p_1 = -2\sigma\cos\theta_d/a,\tag{2.2}$$

where σ is the liquid-gas surface tension, a is the effective radius of the pore and θ_d is the *dynamic* contact angle that the meniscus forms with the solid wall. As shown by experiments on dynamic wetting, in a general case θ_d depends on, and hence should be regarded as a *functional* of, the flow field in the vicinity of the moving contact line (Blake *et al.* 1999; Clarke & Stattersfield 2006). For flow in a porous medium, the contact angle's dependence on the flow field reduces to its dependence only on the contact-line speed u_1 , which is the leading factor determining the flow field in the vicinity of the contact line. Then, θ_d can be described just as a *function* of the contact-line speed:

$$\theta_d = F(u_1/U_{cl}),\tag{2.3}$$

where U_{cl} is an appropriate scale for the velocity. In principle, this dependence, where, for the wetting mode, u_1 coincides with the speed of the meniscus as a whole, could be determined empirically. The theory of flows with forming interfaces (Shikhmurzaev 2007) specifies the inverse of $F(u_1/U)$ as

$$\frac{u_1}{U_{cl}} = \left(\frac{(1 + (1 - \rho_{1e}^s)\cos\theta_s)(\cos\theta_s - \cos\theta_d)^2}{4(\cos\theta_s + B)(\cos\theta_d + B)}\right)^{1/2},\tag{2.4}$$

where $B = (1 - \rho_{1e}^s)^{-1}(1 + \rho_{1e}^s u_0(\theta_d)), \theta_s$ is the static contact angle,

$$u_0(\theta_d) = \frac{\sin \theta_d - \theta_d \cos \theta_d}{\sin \theta_d \cos \theta_d - \theta_d}, \qquad U_{cl} = \left(\frac{\gamma \rho_0^s (1 + 4\alpha\beta)}{\tau \beta}\right)^{1/2}$$

is the characteristic speed associated with the parameters that the 'additional' physics of wetting brings in to resolve the moving contact-line problem, ρ_0^s , ρ_{1e}^s , α , β , γ , τ are material constants characterizing the contacting media whose values can be found elsewhere (Blake & Shikhmurzaev 2002; Shikhmurzaev 2007). The comparison of (2.4) with experimental data published in the literature has shown a very good agreement (Shikhmurzaev 2007) so that (2.4) can be regarded as a reliable representation of the dynamic contact angle behaviour.

An implicit assumption we made above, namely that in the wetting mode the contact-line speed u_1 equals to the cross-sectionally averaged flow velocity $u_{1,flow}$ associated with the motion of the meniscus as a whole, is not immediately obvious, especially given that, as the meniscus breaks through the threshold and goes into the wetting mode, initially, one has both the moving contact line and the varying shape of the meniscus. In other words, in general one has

$$u_{1,flow} = u_1 + \frac{a}{(1+\sin\theta_d)^2} \frac{d\theta_d}{dt},$$

where, as before, we used that the meniscus has the shape of a spherical cap, albeit with a time-dependent radius of curvature. Formally, one can add this equation to the model together with an extra variable $u_{1,flow}$, which should replace u_1 in the text below, but this generalization would be beyond the accuracy of the model. Indeed, if L, U and T = L/U are the length, velocity and time scales for the macroscopic (Darcy-level) motion for which we are deriving the boundary conditions, then one has that the difference between $u_{1,flow}$ and u_1 is of O(a/L) and hence negligible in the continuum limit $a/L \to 0$. The possible deviation of the meniscus shape from a spherical cap takes place also on a vanishing scale. Therefore, within the accuracy of O(1) as $a/L \to 0$, in what follows we use that in the wetting mode $u_{1,flow} = u_1$. It is worth pointing out here that the pores (i.e. capillaries) where the meniscus propagates in the wetting mode are assumed to be long compared to a, so that there is room for the meniscus to propagate in the wetting mode as it is described above.

The speed u_1 at which individual menisci propagate in the wetting mode is not equal to the normal component of the velocity of the wetting front as a whole $u_n = \mathbf{n} \cdot \mathbf{u}|_{\partial\Omega_1}$ (\mathbf{n} is an outward normal) since the menisci intermittently go through both modes of motion and hence, on the Darcy scale, u_n must have contributions from both u_1 and the flow speed u_2 associated with the threshold mode. Then, for u_n one has an equation

$$u_n = A_1 u_1 + A_2 u_2, (2.5)$$

which is similar to (2.1), with the contribution of the *i*th mode proportional to its 'weight' A_i . In an isotropic porous medium, the 'weight' A_i of each mode of motion is essentially the relative time the meniscus spends in this mode. If s_i is the fraction of the length on the pore scale corresponding to the *i*th mode of motion $(s_1 + s_2 = 1)$, then the normalized time that the meniscus spends in this mode is s_i/u_i and hence, given that $1/u_n = s_1/u_1 + s_2/u_2$ and $A_1 + A_2 = 1$, one has

$$A_1 = \frac{s_1 u_2}{s_2 u_1 + s_1 u_2}, \quad A_2 = \frac{s_2 u_1}{s_2 u_1 + s_1 u_2}.$$
 (2.6)

Then, as one would expect, the slowest (controlling) mode of motion tends to make a greater contribution to the pressure at the wetting front and the front's velocity. Now, consider the threshold mode. When the moving meniscus runs into a barrier associated with the threshold mode, such as the edge at the end of a capillary or an asperity, the contact line gets pinned and the meniscus begins to deform until the contact angle reaches a certain value θ_* at which the contact line can move forward again (Fig. 1). This is the essence of the threshold mode, and it comes into play only when θ_d at which the meniscus arrives at a barrier is less than θ_* . In other words,

$$s_1(\theta_d, \theta_*) = \begin{cases} 1, & \theta_d - \theta_* \ge 0 \\ s_{10}, & \theta_d - \theta_* < 0 \end{cases}, \quad s_2 = 1 - s_1, \tag{2.7}$$

where s_{10} (< 1) is a characteristic of the porous matrix. To find the functional dependence of parameters in the threshold mode, consider the dynamics of an individual meniscus. Assume that at a distance l upstream from the meniscus that has just run into a barrier and got its contact line pinned there is a (constant throughout the process) pressure \bar{p} greater than $-2\sigma\cos\theta_d/a$. Then, the meniscus, with the contact line which is now unable to move, will give in to this pressure and deform (Fig. 1). We need to find the flow velocity u_2 as an average over the cross-section and over the time required for the the contact angle to vary from θ_d , at which the meniscus arrived at the barrier, to θ_* , at which the contact line can advance again, and the pressure p_2 as an average over the time of this process.

Neglecting the contribution to the pressure drop due to the deviation of the velocity profile on the pore scale from parabolic in the immediate neighboorhood of the meniscus and calculating the flow rate in the capillary on an assumption that the meniscus retains the shape of a spherical cap (with a varying radius of curvature) throughout the process of its deformation, from the Stokes equation on the pore scale one has that the contact angle θ satisfies an equation

$$\frac{1}{(1+\sin\theta)^2}\frac{d\theta}{dt} = \frac{\sigma}{4\mu l}\left(\frac{\bar{p}a}{2\sigma} + \cos\theta\right),\,$$

which, if multiplied by a, essentially equates the flow velocity averaged over a cross-section to the pressure gradient with a coefficient of proportionality corresponding to the parabolic profile in the pipe flow. Given that there is no discontinuity in the average flow velocity when the contact line becomes instantly pinned and the meniscus starts going from the wetting into the threshold mode, we have an equation

$$\frac{\sigma a}{4\mu l} \left(\frac{\bar{p}a}{2\sigma} + \cos\theta_d \right) = u_1,$$

which can be used to eliminate l. Now, for p_2 , i.e. the pressure $-2\sigma\cos\theta(t)/a$ averaged over a time interval

$$T = au_1^{-1} \left(\frac{\bar{p}a}{2\sigma} + \cos\theta_d \right) I \left(\theta_d, \theta_*; \frac{\bar{p}a}{2\sigma} \right),$$
$$I \left(\theta_d, \theta_*; \frac{\bar{p}a}{2\sigma} \right) = \int_{\theta_d}^{\theta_*} \frac{d\theta}{(1 + \sin\theta)^2 (\bar{p}a/(2\sigma) + \cos\theta)},$$

which is needed for θ to vary from θ_d to θ_* , one has

$$p_2 = \bar{p} - \frac{2\sigma J(\theta_d, \theta_*)}{aI(\theta_d, \theta_*; \bar{p}a/(2\sigma))},$$
(2.8)

where

$$J(\theta_d, \theta_*) = \left[\frac{1}{2} \tan \left(\frac{\theta}{2} - \frac{\pi}{4}\right) + \frac{1}{6} \tan^3 \left(\frac{\theta}{2} - \frac{\pi}{4}\right)\right]_{\theta_d}^{\theta_*},$$

and $[f]_a^b \equiv f(b) - f(a)$. A similar procedure yields the velocity in the threshold mode averaged over T as

$$u_2 = \frac{u_1 J(\theta_d, \theta_*)}{\left(\frac{\bar{p}a}{2\sigma} + \cos\theta_d\right) I\left(\theta_d, \theta_*; \frac{\bar{p}a}{2\sigma}\right)}.$$
 (2.9)

Now, the quantity \bar{p} in equations (2.8) and (2.9) must be specified in macroscopic terms. On the pore scale, it is the excess of \bar{p} over the threshold capillary pressure $p_* = -2\sigma\cos\theta_*/a$ that allows the meniscus to break through the barrier and go into the wetting regime again, with the contact line moving. If \bar{p} reduces to p_* , then the meniscus comes to a halt, and, given that all menisci are modelled as the same, each of them will meet a similar barrier within a time negligible on the macroscopic scale and the wetting front as a whole will come to a stop. Formally, we have that, if $\bar{p} \searrow p_*$, then $I \to \infty$ and hence, according to (2.8) and (2.9), $p_2 \to \bar{p}$ and $u_2 \to 0$. Then, we have from (2.6) that $A_1 \to 0$, $A_2 \to 1$, so that (2.5) and (2.1) yield that on the Darcy scale $u_n \to 0$ and $p \to \bar{p}$. Thus, macroscopically (i.e. on the Darcy scale) \bar{p} is the stagnation pressure, i.e. the pressure that one would have if the wetting front were at rest in its current position. In other words, macroscopically, we need to solve a conjugate problem

$$\nabla^2 \bar{p} = 0, \quad (\mathbf{r} \in \Omega); \qquad \mathbf{n} \cdot \nabla \bar{p}|_{\partial \Omega_1} = 0, \tag{2.10}$$

with the boundary condition for \bar{p} on $\partial\Omega_2$ being the same as for p. Then, the value $\bar{p}|_{\partial\Omega_1}$ is the one we need to use in (2.8) and (2.9), i.e. it is the pressure that builds on a meniscus whose contact line has been pinned. The conjugate problem must be solved in parallel with the main one as the latter requires the value of $\bar{p}|_{\partial\Omega_1}$ at the wetting front throughout its movement.

Now, we can summarize the model as follows. In order to describe the propagation of the wetting front, one has to consider the bulk equations (1.1) and (1.2) in the domain Ω with some boundary condition on $\partial\Omega_2$ that specifies a particular problem, with the kinematic boundary condition at the wetting front $\partial\Omega_1$ given by (1.3) and the dynamic one by (2.1). The pressures p_1 and p_2 that feature in (2.1) are determined from (2.2), (2.8), with the coefficients A_1 , A_2 specified by (2.6), (2.7). For the three variables θ_d , u_1 and u_2 appearing in (2.2), (2.6), (2.7) and (2.8) one has three equations: (2.3) (in particular (2.4)), (2.5) and (2.9), whereas the pressure $\bar{p}|_{\partial\Omega_1}$ featuring in (2.8) and (2.9) has to be found from the conjugate problem (2.10) with the same boundary condition for \bar{p} on $\partial\Omega_2$ as for p. If gravity is to be taken into account, one has to replace p and \bar{p} in (1.1) and

(2.10) with $(p + \rho gz)$ and $(\bar{p} + \rho gz)$, respectively (ρ is the fluid's density, g is the gravitational acceleration, z is the coordinate directed against gravity). Besides the bulk permeability κ , the geometry of the porous matrix enters the model via three effective parameters, s_{10} , a and θ_* . The above model can be generalized by incorporating threshold modes associated with different values of θ_* , replacing the step-function (2.7) for each of them by a more complex one to account for the types of thresholds and the corresponding generalization of (2.6) to incorporate various possibilities for the pore network topology.

3. An illustrative example

Consider the unsteady one-dimensional imbibition against gravity, with z = h(t) as the position of the wetting front $(\partial\Omega_1)$, p and \bar{p} in (1.1) and (2.10) replaced with $p + \rho gz$ and $\bar{p} + \rho gz$ respectively, and $p = \bar{p} = p_0$ at z = 0 as a boundary condition on $\partial\Omega_2$. Then, Laplace's equations (1.2) and (2.10) for p and \bar{p} in the one-dimensional case yield that both p and \bar{p} are linear functions of z, and from the conjugate problem (2.10) with the condition $\bar{p} = p_0$ at z = 0 we have that $\bar{p}(z,t) = p_0 - \rho gz$. Then, using for p its linear dependence on z and the same condition on $\partial\Omega_2$, i.e. $p = p_0$ at z = 0, we can express the pressure gradient dp/dz at z = h in terms of the current position of the wetting front: $dp(h,t)/dz = (p(h,t) - p_0)/h$. Using this expression in Darcy's equation (1.1), where, as mentioned above, p is replaced with $p + \rho gz$, and substituting the latter into the kinematic condition (1.3), which now can be written down simply as $dh/dt = u_n(h,t)$, we arrive at

$$\frac{dh}{dt} = \frac{\kappa}{\mu} \left(\frac{p_0 - p(h, t)}{h} - \rho g \right).$$

This equation together with algebraic equations (2.1), where $p|_{\partial\Omega_1} \equiv p(h,t)$, (2.2), (2.4), (2.5), where $u_n = dh/dt$, (2.6), (2.7), (2.8), where we now use the solution of the conjugate problem $\bar{p}|_{\partial\Omega_1} = p_0 - \rho gh$, and (2.9) form a closed system for h, p(h,t), p_1 , θ_d , u_1 , A_1 , A_2 , s_1 , s_2 , p_2 and u_2 . Typical curves representing the dependence of h, scaled with $h_0 = 2\sigma/(\rho ga)$ and rising from the initial position h(0) = 0, on time t, scaled with $T_0 = 2\sigma\mu/((\rho g)^2a\kappa)$, are shown in Fig. 2 with the values of parameters given in the figure caption. Comparing these dependencies (curves 2–5) with Washburn's (Washburn 1921) curves for a meniscus propagating with $\theta_d \equiv \theta_s$ in a capillary (curve 0: no gravity; curve 1: gravity included), we can see that the present model, besides realistically giving a finite speed of rise at the onset of imbibition, predicts a slightly lower rate of propagation of the wetting front as it is slowed down by both the velocity-dependence of the contact angle and the presence of the threshold mode. The latter comes into play once $\theta_d < \theta_*$ and dictates that the wetting front will come to a halt before it reaches the maximum possible height of imbibition $h_{max} = 2\sigma \cos\theta_s/(\rho ga)$ and, importantly, contrary to how curves 1 and 2 approach h_{max} asymptotically, this coming to a halt occurs in a finite time.

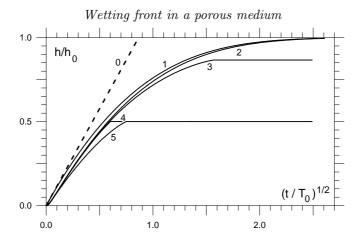


FIGURE 2. Time-dependence of 1D imbibition calculated using the derived model. 0: Washburn, no gravity; 1: Washburn, gravity included; 2: $s_{10}=1$; 3: $s_{10}=0.1$, $\theta_*=30^\circ$; 4: $s_{10}=0.9$, $\theta_*=60^\circ$; 5: $s_{10}=0.1$, $\theta_*=60^\circ$. For all curves $\theta_s=0^\circ$, $p_0=0$, and $\mu U_{cl}/(\kappa \rho g)=10^2$, $\rho_{1e}^s=0.6$ for curves 2–5.

In this position, which is determined only by θ_* (see curves 4 and 5), the wetting front still has the capacity to propagate provided that some other physical mechanism helps it to break through the threshold. In this connection, it is worth pointing out that, as has been observed experimentally, for some systems, there is a change of regime from an essentially Washburn-type to a completely different one halfway between the onset of the process and the maximum imbibition height (Delker *et al.* 1996; Lago & Araujo 2001), so that the present example, considered here as an illustration of how the developed approach works, provides a 'building block' for the modelling of this, yet unexplained, phenomenon.

4. Concluding remarks

The developed approach and, in particular, the technique of conjugate problems it uses to incorporate the threshold mode of motion provide a transparent framework for building models of increased complexity. The threshold mode is the key to describing such effects as formation of trapped pockets of the displaced fluid that can be left behind the wetting front and their subsequent dynamics without resorting to thermodynamic arguments and the necessity to specify, increasingly multi-parametric, thermodynamic dependencies for this, basically mechanical, system. It should be noted, however, that the next step in the developing of the model towards greater complexity, sketched at the end of Section 2, is nontrivial as, for different values of the threshold angle θ_* , it becomes necessary to bring in and specify the topology of the porous matrix with respect to the connectivity of the wetting front.

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