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Macroscopic capillarity and hysteresis for flow in porous media

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A macroscopic theory for capillarity in porous media is presented, challenging the established view that capillary pressure and relative permeability are constitutive parameter functions. The capillary pressure function in the present theory is not an input parameter but an outcome. The theoretical approach is based on introducing the residual saturations explicitly as state variables [as in Phys. Rev. E **58**, 2090 (1998)]. Capillary pressure and relative permeability functions are predicted to exist for special cases. They exhibit hysteresis and process dependence as known from experiment.

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I. INTRODUCTION AND FORMULATION OF THE PROBLEM

A predictive macroscopic theory of two phase flow inside a rigid porous medium is of fundamental importance for many applied sciences such as hydrology, catalysis, petrophysics, or filtration technology [1–8]. Despite being used in innumerable physics and engineering applications, the accepted theory (see below) based on capillary pressure $P_{\rm c}$ and relative permeability functions k^r lacks a sound theoretical foundation, and its basic parameter functions $P_{\rm c}$ and k^r are difficult to measure in experiment [4,6,9].

Modern investigations often abandon the traditional theory and resort to microscopic models (e.g., network models) as an alternative to predict macroscopic immiscible displacement in porous media [10–15]. An important motivation for these alternative investigations is the unresolved problems with the traditional macroscopic theory based on capillary pressure and relative permeabilities and the necessity to relate these functions to pore scale parameters. It is therefore adequate to remind the reader of the traditional theory, introduced more than 60 years ago [16–18], and its problems. One formulation of the traditional theory starts from the fundamental balance laws of continuum mechanics for two fluids (called water $\mathbb{W}$ and oil $\mathbb{O}$) inside the pore space (called $\mathbb{P}$) of a porous sample $\mathbb{S}=\mathbb{P}\cup\mathbb{M}$ with a rigid solid matrix (called $\mathbb{M}$). Recall the law of mass balance in differential form

$$\frac{\partial(\phi_i\varrho_i)}{\partial t}+\nabla\cdot(\phi_i\varrho_i\mathbf{v}_i)=M_i, \tag{1}$$

where $\varrho_i(\mathbf{x},t)$, $\phi_i(\mathbf{x},t)$, $\mathbf{v}_i(\mathbf{x},t)$ denote mass density, volume fraction, and velocity of phase $i=\mathbb{W},\mathbb{O}$ as functions of position $\mathbf{x}\in\mathbb{S}\subset\mathbb{R}^3$ and time $t\in\mathbb{R}_+$. Exchange of mass between the two phases is described by mass transfer rates M_i giving the amount of mass by which phase i changes per unit time and volume. Momentum balance for the two fluids requires in addition

$$\phi_i\varrho_i\frac{\mathrm{D}^i}{\mathrm{D}t}\mathbf{v}_i-\phi_i\nabla\cdot\Sigma_i-\phi_i\mathbf{F}_i=\mathbf{m}_i-\mathbf{v}_iM_i, \tag{2}$$

where Σ_i is the stress tensor in the ith phase, $\mathbf{F}_i$ is the body force per unit volume acting on the ith phase, $\mathbf{m}_i$ is the momentum transfer into phase i from all the other phases, and $\mathrm{D}^i/\mathrm{D}t=\partial/\partial t+\mathbf{v}_i\cdot\nabla$ denotes the material derivative for phase $i=\mathbb{W},\mathbb{O}$.

Defining the saturations $S_i(\mathbf{x},t)$ as the volume fraction of pore space $\mathbb{P}$ filled with phase i one has the relation $\phi_i=\phi S_i$, where ϕ is the porosity of the sample. Expressing volume conservation $\phi_{\mathbb{W}}+\phi_{\mathbb{O}}=\phi$ in terms of saturations yields

$$S_{\mathbb{W}}+S_{\mathbb{O}}=1. \tag{3}$$

In order to get the traditional theory, these balance laws for mass, momentum, and volume have to be combined with specific constitutive assumptions for M_i, $\mathbf{m}_i$, $\mathbf{F}_i$, and Σ_i.

Great simplification is afforded by assuming that the porous medium is macroscopically homogeneous

$$\phi(\mathbf{x})=\phi=\mathrm{const}, \tag{4}$$

although this assumption can be relaxed and is rarely valid in practice [19]. Let us further assume that the fluids are incompressible so that

$$\varrho_{\mathbb{W}}(\mathbf{x},t)=\varrho_{\mathbb{W}}, \tag{5a}$$

$$\varrho_{\mathbb{O}}(\mathbf{x},t)=\varrho_{\mathbb{O}}, \tag{5b}$$

where the constants $\varrho_{\mathbb{W}},\varrho_{\mathbb{O}}$ are independent of $\mathbf{x}$ and t. One assumes next that the stress tensor of the fluids is diagonal

$$\Sigma_{\mathbb{W}}=-P_{\mathbb{W}}\mathbf{1}, \tag{6a}$$

$$\Sigma_{\mathbb{O}}=-P_{\mathbb{O}}\mathbf{1}, \tag{6b}$$

where $P_{\mathbb{W}},P_{\mathbb{O}}$ are the fluid pressures. Realistic subsurface flows have low Reynolds numbers so that the inertial term

$$\frac{\mathrm{D}^i}{\mathrm{D}t}\mathbf{v}_i=0 \tag{7}$$

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can be neglected in the momentum balance equation (2). It is further assumed that the body forces

$$\mathbf{F}_{\mathbb{W}} = \varrho_{\mathbb{W}}\mathbf{g} \tag{8a}$$

$$\mathbf{F}_{\mathbb{O}} = \varrho_{\mathbb{O}}\mathbf{g} \tag{8b}$$

are given by gravity. As long as there are no chemical reactions between the fluids, the mass transfer rates vanish so that

$$M_{\mathbb{W}} = -M_{\mathbb{O}} = 0 \tag{9}$$

holds. Momentum transfer between the fluids and the rigid matrix is governed by viscous drag in the form

$$\mathbf{m}_{\mathbb{W}} = -\frac{\mu_{\mathbb{W}}\phi_{\mathbb{W}}^2}{kk_{\mathbb{W}}^r(S_{\mathbb{W}})}\mathbf{v}_{\mathbb{W}}, \tag{10a}$$

$$\mathbf{m}_{\mathbb{O}} = -\frac{\mu_{\mathbb{O}}\phi_{\mathbb{O}}^2}{kk_{\mathbb{O}}^r(S_{\mathbb{W}})}\mathbf{v}_{\mathbb{O}}, \tag{10b}$$

where $\mu_{\mathbb{W}},\mu_{\mathbb{O}}$ are the constant fluid viscosities, k is the absolute permeability, and $k_{\mathbb{W}}^r(S_{\mathbb{W}}),k_{\mathbb{O}}^r(S_{\mathbb{W}})$ are the so-called relative permeabilities of water and oil.

Inserting the constitutive assumptions (4)–(10) into the mass balance Eq. (1) yields

$$\frac{\partial S_{\mathbb{W}}}{\partial t} + \nabla(S_{\mathbb{W}}\mathbf{v}_{\mathbb{W}}) = 0, \tag{11a}$$

$$\frac{\partial S_{\mathbb{O}}}{\partial t} + \nabla(S_{\mathbb{O}}\mathbf{v}_{\mathbb{O}}) = 0, \tag{11b}$$

while the momentum balance Eqs. (2)

$$\phi_{\mathbb{W}}\mathbf{v}_{\mathbb{W}} = -\frac{k}{\mu_{\mathbb{W}}}k_{\mathbb{W}}^r(S_{\mathbb{W}})(\nabla P_{\mathbb{W}} - \varrho_{\mathbb{W}}\mathbf{g}) \tag{12a}$$

$$\phi_{\mathbb{O}}\mathbf{v}_{\mathbb{O}} = -\frac{k}{\mu_{\mathbb{O}}}k_{\mathbb{O}}^r(S_{\mathbb{W}})(\nabla P_{\mathbb{O}} - \varrho_{\mathbb{O}}\mathbf{g}) \tag{12b}$$

give the generalized Darcy laws for the Darcy velocities $\phi_i\mathbf{v}_i$ [2, p. 155]. The relative permeabilities $k_{\mathbb{W}}^r(S_{\mathbb{W}}),k_{\mathbb{O}}^r(S_{\mathbb{W}})$ are assumed to be nonlinear constitutive parameter functions. They account for the fact, that the permeability of the two immiscible fluids deviates from their partial permeabilities $kS_{\mathbb{W}},kS_{\mathbb{O}}$ obtained from volume averaging of the absolute permeability.

Equations (11) and (12) together with Eq. (3) provide nine equations for ten unknowns $S_{\mathbb{W}},S_{\mathbb{O}},P_{\mathbb{W}}P_{\mathbb{O}},\mathbf{v}_{\mathbb{W}},\mathbf{v}_{\mathbb{O}}$. An additional equation is needed. Based on the work of Smith on capillary rise in regular packings [20] it was argued in Ref. [18] that the pressure difference between oil and water should in general depend only on saturation

$$P_{\mathbb{O}} - P_{\mathbb{W}} = \sigma_{\mathbb{W}\mathbb{O}}\kappa(S_{\mathbb{W}}) = P_{\mathrm{c}}(S_{\mathbb{W}}), \tag{13}$$

where $\sigma_{\mathbb{W}\mathbb{O}}$ is the oil-water interfacial tension and $\kappa(S_{\mathbb{W}})$ is the mean curvature of the oil-water interface. This assumption has remained the cornerstone of the theory of macroscopic capillarity for 60 years, and it is being challenged here. The nonlinear constitutive parameter function $P_{\mathrm{c}}(S_{\mathbb{W}})$ is called the capillary pressure-saturation relation and it is supposed to describe the macroscopic effect of capillarity in hydrostatic equilibrium (without flow). In practice the P_{c} curve is often parametrized in the so-called van Genuchten parametrization by five parameters consisting of two limiting saturations, two exponents, and one pressure prefactor [8]. The traditional theory requires $5+4+3=12$ parameters to parametrize a bounding hysteresis loop consisting of two drainage and one imbibition curves.

This concludes my presentation of the traditional theory. Equations (3) and (11)–(13) appropriately supplemented with initial and boundary conditions constitute the traditional theory of macroscopic capillarity in porous media. In numerous engineering applications, Eqs. (3) and (11)–(13) are solved by computer programs [8].

Serious problems limit the predictive power of Eqs. (3) and (11)–(13). The biggest problem arises from Eq. (13), because it is not unique and does not account for residual saturations. It is tacitly assumed that fluids trapped in pendular rings, ganglia, or blobs behave in the same way as fluids that percolate to the sample surface. Moreover, combining Eq. (13) with Eq. (12) implies that in static equilibrium, when $\mathbf{v}_{\mathbb{W}}=0=\mathbf{v}_{\mathbb{O}}$, the pressure is everywhere hydrostatic, even in the trapped fluids. This is clearly not the case.

Other problems with $P_{\mathrm{c}}(S_{\mathbb{W}})$ and $k_{\mathbb{W}}^r(S_{\mathbb{W}}),k_{\mathbb{O}}^r(S_{\mathbb{O}})$ are multivaluedness, hysteresis, and dynamic effects. Equation (13) is also problematic because it seems to identify a pressure defined on the pore scale with a macroscopically averaged pressure.

Most practitioners ignore these problems and continue to use the traditional set of equations. Many physicists, on the other hand, try to overcome these problems by resorting to microscopic model calculations in an attempt to predict macroscopic behavior starting from the pore scale or below [10,21]. My objective in this paper is to present a purely macroscopic approach in the same spirit as the traditional theory, but without requiring capillary pressure P_{c} or relative permeabilities as input parameters. The approach followed here is to modify the constitutive assumptions (4)–(10) based on the insight that the main effect of capillarity is the distinction between percolating and nonpercolating (trapped) fluid phases [22–24]. The fluids flow hydrodynamically in the percolating regions while the trapped fluids are kept in place by capillary forces. Trapped fluid can only move by viscous drag or through coalescence with percolating fluid regions.

Before defining percolating and nonpercolating fluid phases in Sec. II, it is appropriate to comment on the relation to other approaches. First, several authors (including the present one) have emphasized the importance of introducing the fluid-fluid surface area as a state variable (see [22–24] and references therein). The present approach differs from such theories. The present theory is based only on volume fractions. It avoids surface area and its concomitant proliferation of unknowns and constitutive relations. Second, some models generalize Eq. (13) into a so-called dynamic capillary pressure by including a dependence on rates of saturation change (see, e.g., [25] and references therein). The present approach includes dynamic (or viscous) effects on capillarity in a more fundamental way by avoiding the concept of capillary pressure. Third, there exist several *ad hoc* models for hysteresis loops based on rescaling the main loop formed by the bounding drainage and imbibition curves (see, e.g., [26] and references therein). Again, such approaches differ fundamentally from the present one, because they are based on the traditional concepts of capillary pressure and relative permeability, while the present approach challenges the basis of these concepts. It will be seen below that the present theory requires fewer parameters than previous theories.

II. PERCOLATING VERSUS NONPERCOLATING FLUID REGIONS

The necessity to distinguish between percolating and nonpercolating fluid regions arises from the fact that in static equilibrium, the pressure can become hydrostatic only in those fluid regions that are connected (or percolating) to the sample boundary [22–24]. Each of the two fluids $\mathbb{W},\mathbb{O}$ consists of disjoint and path-connected subsets (regions) $\mathbb{W}_i,\mathbb{O}_i$. More precisely one has

$$\mathbb{W} = \bigcup_{i=1}^{N_{\mathbb{W}}} \mathbb{W}_i, \tag{14a}$$

$$\mathbb{O} = \bigcup_{i=1}^{N_{\mathbb{O}}} \mathbb{O}_i, \tag{14b}$$

where the subsets $\mathbb{W}_i,\mathbb{O}_i$ are mutually disjoint, and each of them is path connected. A set is called path connected if any two of its points can be connected by a path contained inside the set. The sets are called mutually disjoint if $\mathbb{O}_i \cap \mathbb{O}_j = \emptyset$ and $\mathbb{W}_i \cap \mathbb{W}_j = \emptyset$ holds for all $i \neq j$. The integers $N_{\mathbb{W}},N_{\mathbb{O}}$ give the total number of path-connected subsets for water, respectively, oil. Of course, these numbers change with time as do the regions $\mathbb{W}_i,\mathbb{O}_i$.

Now define percolating $(\mathbb{F}_1,\mathbb{F}_3)$ and nonpercolating $(\mathbb{F}_2,\mathbb{F}_4)$ fluid regions by classifying the subsets $\mathbb{W}_i,\mathbb{O}_i$ as to whether they have an empty or nonempty intersection with the sample boundary $\partial\mathbb{S}$. More formally, define

$$\mathbb{F}_1 = \bigcup_{\substack{i=1\\ \partial\mathbb{W}_i \cap \partial\mathbb{S} \neq \emptyset}}^{N_{\mathbb{W}}} \mathbb{W}_i, \tag{15a}$$

$$\mathbb{F}_2 = \bigcup_{\substack{i=1\\ \partial\mathbb{W}_i \cap \partial\mathbb{S} = \emptyset}}^{N_{\mathbb{W}}} \mathbb{W}_i, \tag{15b}$$

$$\mathbb{F}_3 = \bigcup_{\substack{i=1\\ \partial\mathbb{O}_i \cap \partial\mathbb{S} \neq \emptyset}}^{N_{\mathbb{O}}} \mathbb{O}_i, \tag{15c}$$

$$\mathbb{F}_4 = \bigcup_{\substack{i=1\\ \partial\mathbb{O}_i \cap \partial\mathbb{S} = \emptyset}}^{N_{\mathbb{O}}} \mathbb{O}_i \tag{15d}$$

so that $\mathbb{F}_1$ is the union of all regions $\mathbb{W}_i$, and is $\mathbb{F}_3$ the union of all regions $\mathbb{O}_i$, that have nonempty intersection with the sample boundary $\partial\mathbb{S}$. Similarly, $\mathbb{F}_2$ is the union of all regions $\mathbb{W}_i$ that have empty intersection with $\partial\mathbb{S}$, and similarly for $\mathbb{F}_4$. In this way, each point in $\mathbb{P}$ belongs to one of four regions $\mathbb{F}_i$, $i=1,2,3,4$. This results in a total of four fluid phases called percolating, respectively nonpercolating water, and percolating, respectively nonpercolating oil. The index $i=5$ will be used for the rigid matrix (=rock).

III. FORMULATION OF THE MODEL

A. General balance laws

The approach presented here is based on the traditional theory presented in Sec. I combined with the distinction between percolating and nonpercolating phases introduced in Refs. [22–24] and discussed in Sec. II. The volume fractions of the subsets $\mathbb{F}_i \subset \mathbb{S}$, $i=1,2,3,4$, and $\mathbb{M} \subset \mathbb{S}$ are denoted as $\phi_i(\mathbf{x},t)$. Let ϕ denote the porosity (volume fraction of $\mathbb{P}$). Volume conservation implies the relations

$$\phi_1 + \phi_2 + \phi_3 + \phi_4 + \phi_5 = 1, \tag{16a}$$

$$S_1 + S_2 + S_3 + S_4 = 1, \tag{16b}$$

$$1 - \phi = \phi_5, \tag{16c}$$

where $\phi_i = \phi S_i$ $(i=1,2,3,4)$ are volume fractions, and S_i are saturations. The water saturation is defined as $S_{\mathbb{W}} = S_1 + S_2$ and the oil saturation as $S_{\mathbb{O}} = S_3 + S_4$.

The general law of mass balance is again given by Eq. (1) with $i=1,2,3,4$. It provides now four equations instead of two. The general law of momentum balance is given by Eq. (2), also now with $i=1,2,3,4$ instead of $i=\mathbb{W},\mathbb{O}$ as before.

B. General constitutive assumptions

As in the traditional theory, the porous medium is again assumed to be macroscopically homogeneous so that Eq. (4) holds. The flows are slow, and hence, also Eq. (7) continues to hold without change. For incompressible fluids, one has now

$$\varrho_1(\mathbf{x},t) = \varrho_{\mathbb{W}}, \tag{17a}$$

$$\varrho_2(\mathbf{x},t) = \varrho_{\mathbb{W}}, \tag{17b}$$

$$\varrho_3(\mathbf{x},t) = \varrho_{\mathbb{O}}, \tag{17c}$$

$$\varrho_4(\mathbf{x},t) = \varrho_{\mathbb{O}} \tag{17d}$$

analogous to Eq. (5a).

C. Viscous drag

The momentum transfer into phase i from all the other phases is assumed to be a simple viscous drag,

$$\mathbf{m}_i = \sum_{j=1}^{5} R_{ij}(\mathbf{v}_j - \mathbf{v}_i), \tag{18}$$

where the resistance coefficient R_{ij} quantifies the viscous coupling between phase i and j. For the rigid rock matrix,

$\mathbf{v}_5=0$. Hence, $-R_{i5}\mathbf{v}_i$ is the momentum transfer from the wall into phase i. Then

$$\mathbf{m}_1 = R_{13}(\mathbf{v}_3 - \mathbf{v}_1) + R_{14}(\mathbf{v}_4 - \mathbf{v}_1) - R_{15}\mathbf{v}_1, \tag{19a}$$

$$\mathbf{m}_2 = R_{23}(\mathbf{v}_3 - \mathbf{v}_2) + R_{24}(\mathbf{v}_4 - \mathbf{v}_2) - R_{25}\mathbf{v}_2, \tag{19b}$$

$$\mathbf{m}_3 = R_{31}(\mathbf{v}_1 - \mathbf{v}_3) + R_{32}(\mathbf{v}_2 - \mathbf{v}_3) - R_{35}\mathbf{v}_3, \tag{19c}$$

$$\mathbf{m}_4 = R_{41}(\mathbf{v}_1 - \mathbf{v}_4) + R_{42}(\mathbf{v}_2 - \mathbf{v}_4) - R_{45}\mathbf{v}_4, \tag{19d}$$

where $R_{12}=0$ and $R_{34}=0$ was used because there is no common interface and, hence, no direct viscous interaction between these phase pairs. Each R_{ij} is a 3×3 matrix.

D. Capillarity

In the present approach, the body forces are given by gravity plus capillary forces

$$\mathbf{F}_1 = \varrho_1\mathbf{g}, \tag{20a}$$

$$\mathbf{F}_2 = \varrho_2\mathbf{g} + \mathbf{F}_{c\mathbb{W}}, \tag{20b}$$

$$\mathbf{F}_3 = \varrho_3\mathbf{g}, \tag{20c}$$

$$\mathbf{F}_4 = \varrho_4\mathbf{g} + \mathbf{F}_{c\mathbb{O}} \tag{20d}$$

contrary to Eqs. (8). The capillary body forces $\mathbf{F}_{c\mathbb{W}},\mathbf{F}_{c\mathbb{O}}$ are responsible for keeping the trapped fluids inside the medium. They are obtained as gradients of capillary potentials

$$\mathbf{F}_{c\mathbb{W}} = -\nabla\Pi_{c\mathbb{W}}, \tag{21a}$$

$$\mathbf{F}_{c\mathbb{O}} = -\nabla\Pi_{c\mathbb{O}}, \tag{21b}$$

where the capillary potentials $\Pi_{c\mathbb{W}},\Pi_{c\mathbb{O}}$ are defined as

$$\Pi_{c\mathbb{W}} = \Pi_{\mathrm{a}}^{*} - \Pi_{\mathrm{a}}S_1^{-\alpha}, \tag{22a}$$

$$\Pi_{c\mathbb{O}} = \Pi_{\mathrm{b}}^{*} - \Pi_{\mathrm{b}}S_3^{-\beta} \tag{22b}$$

with constants $\Pi_{\mathrm{a}}^{*},\Pi_{\mathrm{b}}^{*},\Pi_{\mathrm{a}},\Pi_{\mathrm{b}}$, and exponents $\alpha,\beta>0$.

Next, the stress tensor for percolating phases can be specified in analogy with Eq. (6) as

$$\Sigma_1 = -P_1\mathbf{1}, \tag{23a}$$

$$\Sigma_3 = -P_3\mathbf{1}, \tag{23b}$$

where P_1 and P_3 are the fluid pressures. The stress tensor Σ_2,Σ_4 for the nonpercolating phases cannot be specified in the same way because the forces cannot propagate in nonpercolating phases. Here it is assumed that these stresses are given by the pressure in the surrounding percolating phase modified by the energy density stored in the common interface with the surrounding percolating phases. This suggests an Ansatz [27]

$$\Sigma_2 = -P_3\mathbf{1} + \frac{\sigma_{\mathbb{W}\mathbb{O}}}{\phi}\frac{\partial A_{32}}{\partial S_2}\mathbf{1}, \tag{24a}$$

$$\Sigma_4 = -P_1 1 + \frac{\sigma_{\mathbb{W}\mathbb{O}}}{\phi}\frac{\partial A_{41}}{\partial S_4}\mathbf{1}, \tag{24b}$$

where $\sigma_{\mathbb{W}\mathbb{O}}$ is the oil-water interfacial tension, and the unknowns $A_{32}(\mathbf{x},t),A_{41}(\mathbf{x},t)$ are the interfacial areas per unit volume of porous medium between phases 3 and 2, respectively 4 and 1. (The factor ϕ arises from the definition of A_{ij} per unit volume of porous medium.) To avoid equations of motion for the unknowns A_{32} and A_{41}, it is assumed that geometrical relations of the form

$$A_{32} = A_2^{*}S_2^{\gamma}, \tag{25a}$$

$$A_{41} = A_4^{*}S_4^{\delta} \tag{25b}$$

hold, where A_2^{*},A_4^{*} are prefactors assumed to be constant. Thus, for the nonpercolating phases,

$$\Sigma_2 = (-P_3 + \gamma P_2^{*}S_2^{\gamma-1})\mathbf{1}, \tag{26a}$$

$$\Sigma_4 = (-P_1 + \delta P_4^{*}S_4^{\delta-1})\mathbf{1} \tag{26b}$$

will be used below. Here

$$P_2^{*} = A_2^{*}\frac{\sigma_{\mathbb{W}\mathbb{O}}}{\phi}, \tag{27a}$$

$$P_4^{*} = A_4^{*}\frac{\sigma_{\mathbb{W}\mathbb{O}}}{\phi} \tag{27b}$$

are constants.

The mass transfer rates must depend on rates of saturation change. They are here assumed to be

$$M_1 = -M_2 = \eta_2\phi\varrho_{\mathbb{W}}\left(\frac{S_2 - S_2{}^{*}}{S_{\mathbb{W}}{}^{*} - S_{\mathbb{W}}}\right)\frac{\partial S_{\mathbb{W}}}{\partial t}, \tag{28a}$$

$$M_3 = -M_4 = \eta_4\phi\varrho_{\mathbb{O}}\left(\frac{S_4 - S_4{}^{*}}{S_{\mathbb{O}}{}^{*} - S_{\mathbb{O}}}\right)\frac{\partial S_{\mathbb{O}}}{\partial t}, \tag{28b}$$

where η_2,η_4 are constants. The parameters $S_{\mathbb{W}}{}^{*},S_{\mathbb{O}}{}^{*},S_2{}^{*},S_4{}^{*}$ are defined by

$$S_{\mathbb{W}}{}^{*} = \frac{1 - S_{\mathbb{O}\mathrm{im}}}{2}\left[1 + \tanh\left(\tau_{\mathbb{W}}\frac{\partial S_{\mathbb{W}}}{\partial t}\right)\right] + \frac{S_{\mathbb{W}\mathrm{dr}}}{2}\left[1 - \tanh\left(\tau_{\mathbb{W}}\frac{\partial S_{\mathbb{W}}}{\partial t}\right)\right], \tag{29a}$$

$$S_{\mathbb{O}}{}^{*} = \frac{1 - S_{\mathbb{W}\mathrm{dr}}}{2}\left[1 + \tanh\left(\tau_{\mathbb{O}}\frac{\partial S_{\mathbb{O}}}{\partial t}\right)\right] + \frac{S_{\mathbb{O}\mathrm{im}}}{2}\left[1 - \tanh\left(\tau_{\mathbb{O}}\frac{\partial S_{\mathbb{O}}}{\partial t}\right)\right], \tag{29b}$$

$$S_2{}^{*} = \frac{S_{\mathbb{W}\mathrm{dr}}}{2}\left[1 - \tanh\left(\tau_2\frac{\partial S_{\mathbb{W}}}{\partial t}\right)\right], \tag{29c}$$

$$S_4{}^* = \frac{S_{\mathbb{O}\mathrm{im}}}{2}\left[1 - \tanh\left(\tau_4 \frac{\partial S_{\mathbb{O}}}{\partial t}\right)\right], \tag{29d}$$

where $S_{\mathbb{W}\,\mathrm{dr}}, S_{\mathbb{O}\,\mathrm{im}}$ are limiting saturations for S_2, S_4 and $\tau_{\mathbb{W}}, \tau_{\mathbb{O}}, \tau_2, \tau_4$ are equilibration time scales for reaching capillary equilibrium. For simplicity

$$\tau = \tau_{\mathbb{W}} = \tau_{\mathbb{O}} = \tau_2 = \tau_4 \tag{30}$$

will be assumed below.

IV. IDENTIFICATION OF CAPILLARY PRESSURE

A. Hydrostatic equilibrium

The constitutive theory proposed above, contrary to the traditional theory, does not postulate a unique capillary pressure as a constitutive parameter function. On the other hand, experimental evidence suggests that capillary pressure is a useful concept to correlate observations. To make contact with the established traditional theory, it is therefore important to check whether the traditional $P_{\mathrm{c}}(S_{\mathbb{W}})$ relation can be viewed as a derived concept within the new theory.

Consider first the case of hydrostatic equilibrium where $\mathbf{v}_i = 0$ for all $i = 1,2,3,4$. In hydrostatic equilibrium, all fluids are at rest. In this case, the traditional theory implies $\partial S_{\mathbb{W}}/\partial t = 0$ and $\partial S_{\mathbb{O}}/\partial t = 0$, by mass balance Eqs. (11). The traditional momentum balance Eqs. (12) can be integrated to give

$$P_{\mathbb{W}}(\mathbf{x}) = P_{\mathbb{W}}(\mathbf{x}_0) + \varrho_{\mathbb{W}}\mathbf{g}(\mathbf{x} - \mathbf{x}_0), \tag{31a}$$

$$P_{\mathbb{O}}(\mathbf{x}) = P_{\mathbb{O}}(\mathbf{x}_0) + \varrho_{\mathbb{O}}\mathbf{g}(\mathbf{x} - \mathbf{x}_0), \tag{31b}$$

where $\mathbf{x}_0$ is a point in the boundary. Combined with the assumption (13), one finds

$$P_{\mathrm{c}}[S_{\mathbb{W}}(\mathbf{x})] = P_{\mathbb{O}}(\mathbf{x}) - P_{\mathbb{W}}(\mathbf{x}) = P_{\mathrm{c}0} + (\varrho_{\mathbb{O}} - \varrho_{\mathbb{W}})\mathbf{g}(\mathbf{x} - \mathbf{x}_0) \tag{32}$$

implying the existence of a unique hydrostatic saturation profile $S_{\mathbb{W}}(\mathbf{x})$. Here $P_{\mathrm{c}0} = P_{\mathrm{c}}(\mathbf{x}_0)$ is the capillary pressure at $\mathbf{x} = \mathbf{x}_0$. Experiments show, however, that hydrostatic saturation profiles are not unique. As a consequence, the traditional theory employs multiple $P_{\mathrm{c}}(S_{\mathbb{W}})$ relations for drainage and imbibition, and this leads to difficult problems when imbibition and drainage occur simultaneously.

The nonlinear theory proposed here can be solved in the special case of hydrostatic equilibrium. Mass balance (1) now implies $\partial S_i/\partial t = 0$ for all $i = 1,2,3,4$. Integrating Eq. (2) yields

$$P_1(\mathbf{x}) = P_1(\mathbf{x}_0) + \varrho_{\mathbb{W}}\mathbf{g}(\mathbf{x} - \mathbf{x}_0), \tag{33a}$$

$$\begin{aligned} P_3(\mathbf{x}) = {} & P_3(\mathbf{x}_0) + \Pi_{\mathrm{a}}(S_1(\mathbf{x})^{-\alpha} - S_1(\mathbf{x}_0)^{-\alpha}) \\ & + \gamma P_2^*(S_2(\mathbf{x})^{\gamma-1} - S_2(\mathbf{x}_0)^{\gamma-1}) + \varrho_{\mathbb{W}}\mathbf{g}(\mathbf{x} - \mathbf{x}_0), \end{aligned} \tag{33b}$$

$$P_3(\mathbf{x}) = P_3(\mathbf{x}_0) + \varrho_{\mathbb{O}}\mathbf{g}(\mathbf{x} - \mathbf{x}_0), \tag{33c}$$

$$\begin{aligned} P_1(\mathbf{x}) = {} & P_1(\mathbf{x}_0) + \Pi_{\mathrm{b}}(S_3(\mathbf{x})^{-\beta} - S_3(\mathbf{x}_0)^{-\beta}) \\ & + \delta P_4^*(S_4(\mathbf{x})^{\delta-1} - S_4(\mathbf{x}_0)^{\delta-1}) + \varrho_{\mathbb{O}}\mathbf{g}(\mathbf{x} - \mathbf{x}_0). \end{aligned} \tag{33d}$$

If one identifies P_1 with $P_{\mathbb{W}}$ and P_3 with $P_{\mathbb{O}}$ then Eqs. (33a) and (33b) suggest to identify P_{c} as $P_3 - P_1$. Then Eqs. (33c) and (33d) combined with $S_1 = S_{\mathbb{W}} - S_2$ and $S_3 = 1 - S_{\mathbb{W}} - S_4$ imply $P_{\mathrm{c}} = P_{\mathrm{c}}(S_{\mathbb{W}}, S_2, S_4)$. The capillary pressure P_{c} depends not only on $S_{\mathbb{W}}$ but also on S_2 and S_4 in hydrostatic equilibrium. In the theory proposed here, it is not possible to identify a unique $P_{\mathrm{c}}(S_{\mathbb{W}})$ relation when all fluids are at rest. This agrees with the experiment.

B. Residual decoupling approximation

While it is not possible to identify a unique $P_{\mathrm{c}}(S_{\mathbb{W}})$ relation in hydrostatic equilibrium, such a functional relation emerges, nevertheless, from the present theory when the system approaches hydrostatic equilibrium in the *residual decoupling approximation*. The approach to hydrostatic equilibrium in the residual decoupling approximation (RDA) can be formulated mathematically as $\mathbf{v}_4 = 0$, $\mathbf{v}_2 = 0$, and $R_{23} = 0$, $R_{41} = 0$. In addition, it is assumed that the velocities $\mathbf{v}_1, \mathbf{v}_3 \to 0$ are small but nonzero. In the RDA, mass balance becomes

$$\frac{\partial S_1}{\partial t} + \nabla(S_1\mathbf{v}_1) = \eta_2\left(\frac{S_2 - S_2{}^*}{S_{\mathbb{W}}{}^* - S_{\mathbb{W}}}\right)\frac{\partial S_{\mathbb{W}}}{\partial t}, \tag{34a}$$

$$\frac{\partial S_2}{\partial t} = -\,\eta_2\left(\frac{S_2 - S_2{}^*}{S_{\mathbb{W}}{}^* - S_{\mathbb{W}}}\right)\frac{\partial S_{\mathbb{W}}}{\partial t}, \tag{34b}$$

$$\frac{\partial S_3}{\partial t} + \nabla(S_3\mathbf{v}_3) = \eta_4\left(\frac{S_4 - S_4{}^*}{S_{\mathbb{O}}{}^* - S_{\mathbb{O}}}\right)\frac{\partial S_{\mathbb{O}}}{\partial t}, \tag{34c}$$

$$\frac{\partial S_4}{\partial t} = -\,\eta_4\left(\frac{S_4 - S_4{}^*}{S_{\mathbb{O}}{}^* - S_{\mathbb{O}}}\right)\frac{\partial S_{\mathbb{O}}}{\partial t}. \tag{34d}$$

Momentum balance becomes in the RDA

$$\phi_1(\nabla P_1 - \varrho_{\mathbb{W}}\mathbf{g}) = R_{13}\mathbf{v}_3 - (R_1 + M_1)\mathbf{v}_1, \tag{35a}$$

$$0 = \phi_2(\nabla P_3 + \nabla\Pi_{c\mathbb{W}} - \gamma P_2^*\,\nabla\,S_2^{\gamma-1} - \varrho_{\mathbb{W}}\mathbf{g}), \tag{35b}$$

$$\phi_3(\nabla P_3 - \varrho_{\mathbb{O}}\mathbf{g}) = R_{31}\mathbf{v}_1 - (R_3 + M_3)\mathbf{v}_3, \tag{35c}$$

$$0 = \phi_4(\nabla P_1 + \nabla\Pi_{c\mathbb{O}} - \delta P_4^*\,\nabla\,S_4^{\delta-1} - \varrho_{\mathbb{O}}\mathbf{g}), \tag{35d}$$

where the abbreviations

$$R_1 = R_{13} + R_{14} + R_{15}, \tag{36a}$$

$$R_3 = R_{31} + R_{32} + R_{35} \tag{36b}$$

were used. Equations (34) and (35) together with Eq. (16b) provide 17 equations for 12 variables (P_1, P_3, $\mathbf{v}_1$, $\mathbf{v}_3$ and S_i, $i = 1,2,3,4$).

Equations (34) and (35) can now be compared to the traditional equations (11)–(13) with the aim of identifying capillary pressure and relative permeability. Consider first the momentum balance Eqs. (35). As in the traditional theory [2], viscous decoupling is assumed to hold, i.e., $R_{31} = 0$ and $R_{13} = 0$. Next, assuming that $R_1 \gg M_1$, $R_3 \gg M_3$, and $S_i \neq 0$, one finds

$$\phi_1(\nabla P_1-\varrho_{\mathbb{W}}\mathbf{g})=-R_1\mathbf{v}_1=-R_1\frac{\phi_{\mathbb{W}}}{\phi_1}\mathbf{v}_{\mathbb{W}}, \tag{37a}$$

$$\nabla P_3=-\nabla\Pi_{c\mathbb{W}}+\gamma P_2^*\nabla S_2^{\gamma-1}+\varrho_{\mathbb{W}}\mathbf{g}, \tag{37b}$$

$$\phi_3(\nabla P_3-\varrho_{\mathbb{O}}\mathbf{g})=-R_3\mathbf{v}_3=-R_3\frac{\phi_{\mathbb{O}}}{\phi_3}\mathbf{v}_{\mathbb{O}}, \tag{37c}$$

$$\nabla P_1=-\nabla\Pi_{c\mathbb{O}}+\delta P_4^*\nabla S_4^{\delta-1}+\varrho_{\mathbb{O}}\mathbf{g}, \tag{37d}$$

where barycentric velocities $\mathbf{v}_{\mathbb{W}},\mathbf{v}_{\mathbb{O}}$ defined through

$$S_{\mathbb{W}}\mathbf{v}_{\mathbb{W}}=S_1\mathbf{v}_1+S_2\mathbf{v}_2, \tag{38a}$$

$$S_{\mathbb{O}}\mathbf{v}_{\mathbb{O}}=S_3\mathbf{v}_3+S_4\mathbf{v}_4 \tag{38b}$$

have been introduced. Subtracting Eq. (37a) from Eq. (37c), as well as Eq. (37d) from Eq. (37b), and equating the result gives

$$\begin{aligned}2(\varrho_{\mathbb{O}}-\varrho_{\mathbb{W}})\mathbf{g}+\frac{R_1}{\phi_1^2}\phi_{\mathbb{W}}\mathbf{v}_{\mathbb{W}}-\frac{R_3}{\phi_3^2}\phi_{\mathbb{O}}\mathbf{v}_{\mathbb{O}}\\=\nabla(\Pi_{\mathrm{a}}S_1^{-\alpha}-\Pi_{\mathrm{b}}S_3^{-\beta}+\gamma P_2^*S_2^{\gamma-1}-\delta P_4^*S_4^{\delta-1}),\end{aligned} \tag{39}$$

where Eq. (22) has also been employed. This result can be compared to the traditional theory where one finds from Eqs. (12) and (13),

$$(\varrho_{\mathbb{O}}-\varrho_{\mathbb{W}})\mathbf{g}+\frac{\mu_{\mathbb{W}}}{kk_{\mathbb{W}}^r}\phi_{\mathbb{W}}\mathbf{v}_{\mathbb{W}}-\frac{\mu_{\mathbb{O}}}{kk_{\mathbb{O}}^r}\phi_{\mathbb{O}}\mathbf{v}_{\mathbb{O}}=\nabla P_{\mathrm{c}}. \tag{40}$$

Again this seems to imply $P_{\mathrm{c}}=P_{\mathrm{c}}(S_{\mathbb{W}},S_2,S_4)$ as already found above for hydrostatic equilibrium. However, within the RDA additional constraints follow from mass balance (34).

First, observe that adding (34a) and (34b), respectively (34c) and (34d), with the help of Eq. (38a) yields the traditional mass balance Eqs. (11). Next, verify by insertion that Eqs. (34b) and (34d) admit the solutions

$$S_2(\mathbf{x},t)=S_2{}^*(\mathbf{x})+[S_{20}(\mathbf{x})-S_2{}^*(\mathbf{x})]\left(\frac{S_{\mathbb{W}}{}^*(\mathbf{x})-S_{\mathbb{W}}(\mathbf{x},t)}{S_{\mathbb{W}}{}^*(\mathbf{x})-S_{\mathbb{W}0}(\mathbf{x})}\right)^{\eta_2}, \tag{41a}$$

$$S_4(\mathbf{x},t)=S_4{}^*(\mathbf{x})+[S_{40}(\mathbf{x})-S_4{}^*(\mathbf{x})]\left(\frac{S_{\mathbb{W}}(\mathbf{x},t)-S_{\mathbb{W}}{}^*(\mathbf{x})}{S_{\mathbb{W}0}(\mathbf{x})-S_{\mathbb{W}}{}^*(\mathbf{x})}\right)^{\eta_4}, \tag{41b}$$

where the displacement process is assumed to start from the initial conditions

$$S_{\mathbb{W}}(\mathbf{x},t_0)=S_{\mathbb{W}0}(\mathbf{x}), \tag{42a}$$

$$S_2(\mathbf{x},t_0)=S_{20}(\mathbf{x}), \tag{42b}$$

$$S_4(\mathbf{x},t_0)=S_{40}(\mathbf{x}) \tag{42c}$$

at some initial instant t_0. The limiting saturations $S_{\mathbb{W}}{}^*,S_{\mathbb{O}}{}^*,S_2{}^*,S_4{}^*$ are given by Eqs. (29). They depend only on the sign of $\partial S_{\mathbb{W}}/\partial t$ if $\tau\gg\partial S_{\mathbb{W}}/\partial t$ can be assumed to hold. One finds in this case,

$$S_{\mathbb{W}}{}^*=1-S_{\mathbb{O}\mathrm{im}}, \tag{43a}$$

$$S_{\mathbb{O}}{}^*=S_{\mathbb{O}\mathrm{im}}, \tag{43b}$$

$$S_2{}^*=0, \tag{43c}$$

$$S_4{}^*=S_{\mathbb{O}\mathrm{im}} \tag{43d}$$

for imbibition processes (i.e., $\partial S_{\mathbb{W}}/\partial t>0$), respectively

$$S_{\mathbb{W}}{}^*=S_{\mathbb{W}\mathrm{dr}}, \tag{44a}$$

$$S_{\mathbb{O}}{}^*=1-S_{\mathbb{W}\mathrm{dr}}, \tag{44b}$$

$$S_2{}^*=S_{\mathbb{W}\mathrm{dr}}, \tag{44c}$$

$$S_4{}^*=0 \tag{44d}$$

for drainage processes (i.e., $\partial S_{\mathbb{W}}/\partial t<0$).

With these solutions in hand, the capillary pressure can be identified up to a constant as

$$\begin{aligned}P_{\mathrm{c}}(S_{\mathbb{W}})=\frac{1}{2}[\Pi_{\mathrm{a}}(S_{\mathbb{W}}-S_2)^{-\alpha}-\Pi_{\mathrm{b}}(1-S_{\mathbb{W}}-S_4)^{-\beta}\\+\gamma P_2^*S_2^{\gamma-1}-\delta P_4^*S_4^{\delta-1}],\end{aligned} \tag{45}$$

where $S_2=S_2(S_{\mathbb{W}})$ and $S_4=S_4(S_{\mathbb{W}})$ are given by Eqs. (41). This result holds in the RDA combined with the assumptions above. Furthermore, Eqs. (37a) and (37c) are recognized as generalized Darcy laws with relative permeabilities identified as

$$k_{\mathbb{W}}^r(S_{\mathbb{W}})=2R_1^{-1}\frac{\mu_{\mathbb{W}}}{k}\phi^2(S_{\mathbb{W}}-S_2)^2, \tag{46a}$$

$$k_{\mathbb{O}}^r(S_{\mathbb{W}})=2R_3^{-1}\frac{\mu_{\mathbb{O}}}{k}\phi^2(1-S_{\mathbb{W}}-S_4)^2, \tag{46b}$$

where $S_2=S_2(S_{\mathbb{W}})$ and $S_4=S_4(S_{\mathbb{W}})$ are again given by Eqs. (41).

C. Reproduction of experimental observations

Figure 1 visualizes the results obtained by fitting Eq. (45) to experiment. The experimental results are depicted as triangles (primary drainage) and squares (imbibition). The experiments were performed in a medium grained unconsolidated water wet sand of porosity $\phi=0.34$. Water was used as wetting fluid while air respectively trichloroethylene (TCE) was used as the nonwetting fluid. The experiments were carried out over a period of several weeks at the Versuchseinrichtung zur Grundwasser-und Altlastensanierung (VEGAS)

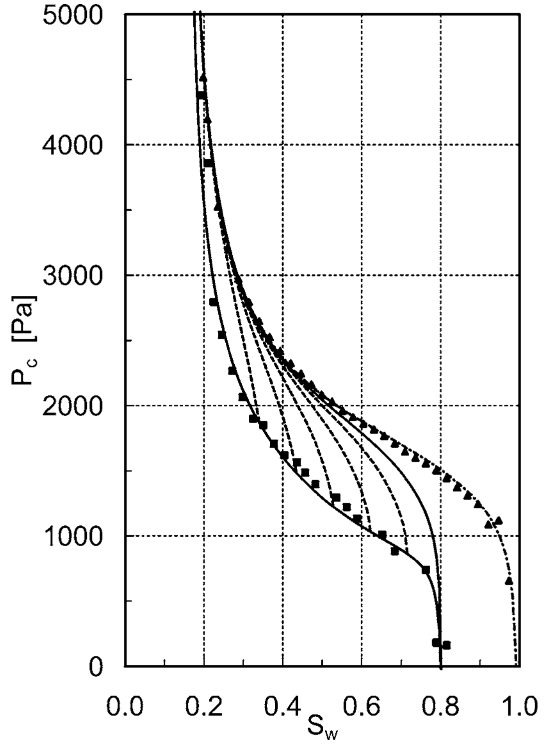

FIG. 1. Hysteresis loop and drainage scanning curves for capillary pressure P_c as function of water saturation $S_{\mathbb{W}}$ fitted to experimental data obtained for a water-wet medium grained sand of porosity $\phi=0.34$. The primary drainage curve is the dashed-dotted line. The main hysteresis loop is the solid line. The dashed lines are drainage scanning curves. All eight curves have the same parameters: $S_{\mathbb{W}\,\mathrm{dr}}=0.15$, $S_{\mathbb{O}\,\mathrm{im}}=0.19$, $\alpha=0.52$, $\beta=0.90$, $\gamma=1.5$, $\delta=3.5$, $\eta_2=4$, $\eta_4=3$, $\Pi_a=1620$ Pa, $\Pi_b=25$ Pa, and $P_2^*=2500$ Pa, $P_4^*=400$ Pa. The five scanning curves start from the boundary imbibition curve at $S_{\mathbb{W}}=0.3,0.4,0.5,0.6,0.7$.

at the Universität Stuttgart. They are described in more detail in Ref. [28]. The parameters for all the curves shown in all four figures are $S_{\mathbb{W}\,\mathrm{dr}}=0.15$, $S_{\mathbb{O}\,\mathrm{im}}=0.19$, $\alpha=0.52$, $\beta=0.90$, $\gamma=1.5$, $\delta=3.5$, $\eta_2=4$, $\eta_4=3$, $\Pi_a=1620$ Pa, $\Pi_b=25$ Pa, and $P_2^*=2500$ Pa, $P_4^*=400$ Pa.

If it is further assumed that the medium is isotropic and that the matrices R_1, R_3 have the form

$$R_1 = R_1^* \phi_1^{-\kappa_{\mathbb{W}}} \mathbf{1}, \tag{47a}$$

$$R_3 = R_3^* \phi_3^{-\kappa_{\mathbb{O}}} \mathbf{1} \tag{47b}$$

then the relative permeability functions are obtained from Eqs. (46). The result for the special case $\kappa_{\mathbb{W}}=\kappa_{\mathbb{O}}=0$ is shown in Figs. 3 and 4. The parameters R_1^*, R_3^* are chosen such that $R_1^*=2\phi^2\mu_{\mathbb{W}}/k$ and $R_3^*=2\phi^2\mu_{\mathbb{O}}/k$, where $\mu_{\mathbb{W}}, \mu_{\mathbb{O}}$ are the fluid viscosities and k is the absolute permeability of the medium. All other parameters for the relative permeability functions shown in Figs. 3 and 4 are identical to those of the capillary pressure curves in Figs. 1 and 2.

Note that Figs. 1–4 show a total of 30 different scanning curves, 5 drainage and 5 imbibition scanning curves each for P_c, $k^r_{\mathbb{W}}$, and $k^r_{\mathbb{O}}$. In addition, a total of nine different bounding curves are displayed, namely the primary drainage, secondary drainage, and secondary imbibition curve for P_c, $k^r_{\mathbb{W}}$, and $k^r_{\mathbb{O}}$. Three more bounding curves, namely primary imbibition for P_c, $k^r_{\mathbb{W}}$, and $k^r_{\mathbb{O}}$ starting from $S_{\mathbb{W}}=0$, are not shown because they are difficult to obtain experimentally for a water-wet sample. Of course, the number of scanning curves can be increased indefinitely. All of these curves have the same values of the constitutive parameters. There is less than one parameter per curve. The curves shown in the figures exhibit the full range of hysteretic phenomena known from experiment. Nevertheless, it should be kept in mind that these curves are obtained only under special approximations, and when these are not valid, such curves do not exist.

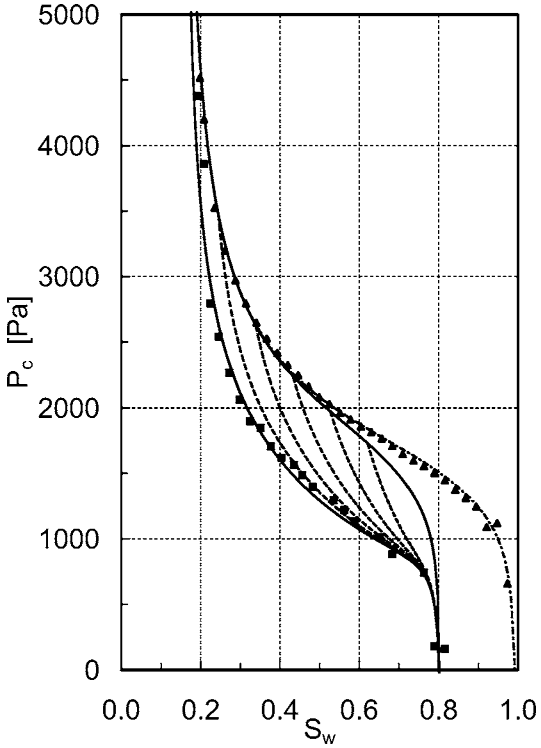

FIG. 2. Hysteresis loop for capillary pressure P_c as function of water saturation $S_{\mathbb{W}}$ as in Fig. 1. The imbibition scanning curves start from the secondary drainage curve. Parameters and line styles are identical to those in Fig. 1.

V. CONCLUSION

The present paper has shown that the experimental observations on hysteresis and process dependence of capillary pressure and relative permeabilities can be reproduced within a macroscopic phenomenological theory. The approach is based on the distinction between percolating and nonpercolating fluid phases introduced in Refs. [22–24]. Contrary to what was expected, the introduction of interfacial areas can be avoided at the expense of some rather drastic constitutive assumptions. In spite of these assumptions, the present theory is more general than the traditional theory because the

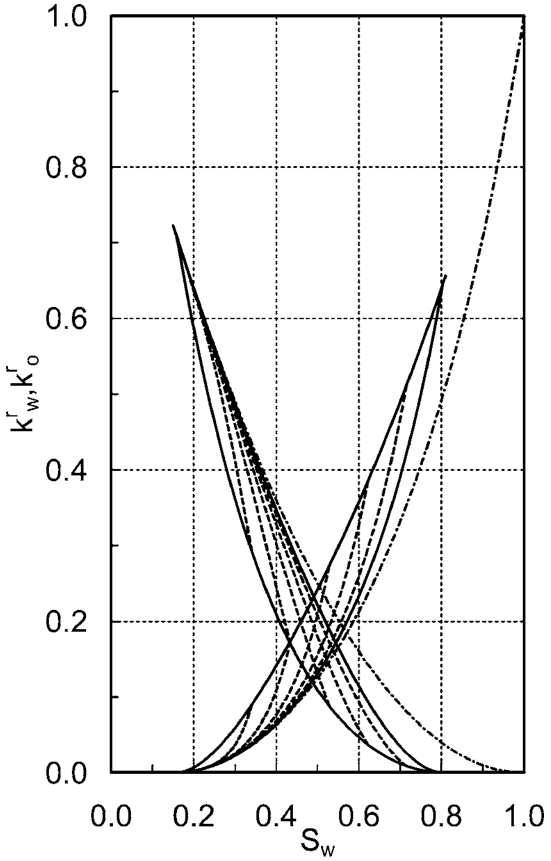

FIG. 3. Hysteresis loop and drainage scanning curves for relative permeabilities $k^r_{\mathbb{W}}, k^r_{\mathbb{O}}$ as function of water saturation $S_{\mathbb{W}}$. Parameters and line styles are identical to those for capillary pressure in Figs. 1 and 2. Here $\kappa_{\mathbb{W}}=\kappa_{\mathbb{O}}=0$ and the parameters R_1^*, R_3^* are chosen such that $R_1^*=2\phi^2\mu_{\mathbb{W}}/k$ and $R_3^*=2\phi^2\mu_{\mathbb{O}}/k$.

latter can be obtained as a special case. In this special case, capillary pressure and relative permeability functions can be identified and are found to show similar hysteresis and process dependence as observed in experiment.

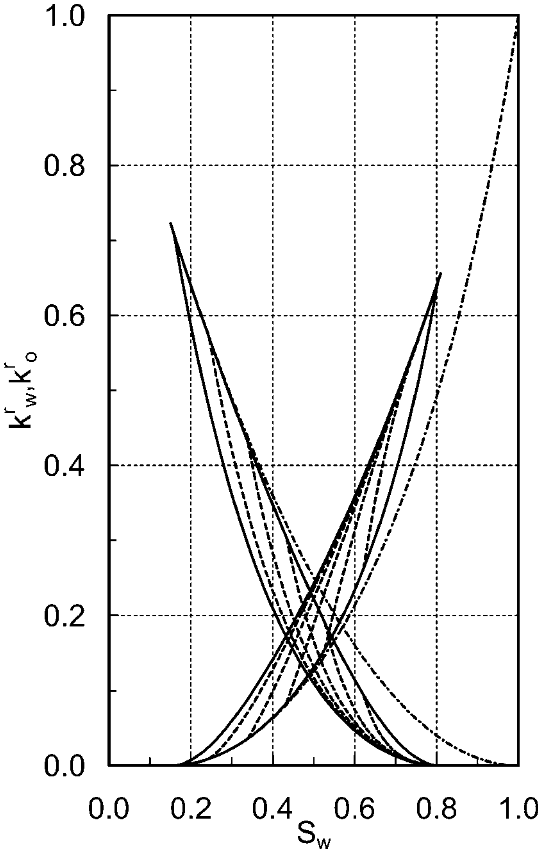

FIG. 4. Same as Fig. 3 with imbibition scanning curves starting from the secondary drainage curve. Parameters and line styles are identical to those in Fig. 3.

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