

**Generalized nonequilibrium capillary relations for two-phase flow through heterogeneous media**Brahim Amaziane,<sup>1,\*</sup> Josipa Pina Milišić,<sup>2,†</sup> Mikhail Panfilov,<sup>3,‡</sup> and Leonid Pankratov<sup>1,4,§</sup><sup>1</sup>*Laboratoire de Mathématiques et leurs Applications, CNRS-UMR 5142, Université de Pau, France*<sup>2</sup>*Faculty of Electrical Engineering and Computing, University of Zagreb, Croatia*<sup>3</sup>*Laboratoire d'Energétique et de Mécanique Théorique et Appliquée, CNRS - Université de Lorraine, France*<sup>4</sup>*B. Verkin Institute for Low Temperature Physics and Engineering, Kharkov, Ukraine*

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For two-phase flow in porous media, the natural medium heterogeneity necessarily gives rise to capillary nonequilibrium effects. The relaxation to the equilibrium is a slow process which should be introduced in macroscopic flow models. Many nonequilibrium models are based on a phenomenological approach. At the same time there exists a rigorous mathematical way to develop the nonequilibrium equations. Its formalism, developed by Bourgeat and Panfilov [*Computational Geosciences* **2**, 191 (1998)], is based on the homogenization of the microscale flow equations over medium heterogeneities. In contrast with the mentioned paper, in which the case of a sufficiently fast relaxation was analyzed, we consider the case of long relaxation, which leads to the appearance of long-term memory on the macroscale. Due to coupling between the nonlinearity and nonlocality in time, the macroscopic model remains, however, incompletely homogenized, in the general case. At the same time, frequently only the relationship for the nonequilibrium capillary pressure is of interest for applications. In the present paper, we obtain such an exact relationship in two different independent forms for the case of long-term memory. This relationship is more general than that obtained by Bourgeat and Panfilov. In addition, we prove the comparison theorem which determines the upper and lower bounds for the macroscopic model. These bounds represent linear flow models, which are completely homogenized. The results obtained are illustrated by numerical simulations.

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**I. INTRODUCTION**

Among the various new approaches used in an attempt to improve the classical theory of two-phase flow in porous media and which are now in fast development, the concept of capillary nonequilibrium plays one of the most important roles. For porous media, it was first introduced by Barenblatt [1] and then developed further by a number of authors. It is closely related to the concept of capillary movement, self-redistribution of phases in space, and medium heterogeneity.

The objective of the present paper is to obtain a general relationship for the nonequilibrium capillary pressure, which is considered now as the main problem of the theory. The phenomenological approaches study only the weak nonequilibrium, which may be described by some relaxation equations. However, even in this case the phenomenological theory is incapable of answering the question of how the nonlinearity of two-phase flow influences the relaxation process and its parameters. The case of high nonequilibrium should be analyzed in a different way. In the present paper we study it by means of the homogenization techniques.

**A. Capillary nonequilibrium, capillary relaxation, and self-redistribution of phases**

For a two-phase fluid, the interfaces between the phases generate the field of surface forces called *the capillary forces* which act on both fluids. The free surface of water in a thin

vertical glass tube immersed in a basin is the simplest classical example illustrating such a situation. It is well known that the level of water in the tube is usually different from the water table in the basin, which proves the existence of a capillary force applied to the interface between water and air in the tube. This force is proportional to the curvature of the interface and may be positive, negative, or nil. For a general interface geometry, the capillary force is applied to any point of the surface and is determined by its local curvature. Thus, a drop of liquid surrounded by another liquid or gas undergoes the action of a vector field of surface capillary forces, as shown in Fig. 1.

If the resulting force is nonzero, as in Fig. 1(b), then the drop is in a nonequilibrium state and will deform its shape with the objective of reaching the equilibrium [Fig. 1(a)]. For a fluid in the bulk, the relaxation to the equilibrium represents a very fast process of drop deformation which may be practically neglected. However, in porous media, the situation is sufficiently different.

In a thin pore tube two fluid phases create the special type of interfaces shown in Fig. 2 and called the menisci. They are the curvilinear surfaces crossing the tube, whose curvature is inversely proportional to the tube radius. Thus, the thinner the tube, the higher the capillary force applied to the meniscus. The equilibrium shape of the phase cluster is determined by the equilibrium between the capillary forces applied to all menisci. In the homogeneous medium shown in Fig. 2, practically any imaginable shape of phase cluster is in equilibrium [as seen, the resulting horizontal and vertical forces are zero for both examples; see Figs. 2(a) and 2(b)].

However, a heterogeneous porous medium causes capillary nonequilibrium. The medium shown in Fig. 3 consists of large and small tubes. As seen, the gray fluid in Fig. 3(a) undergoes a

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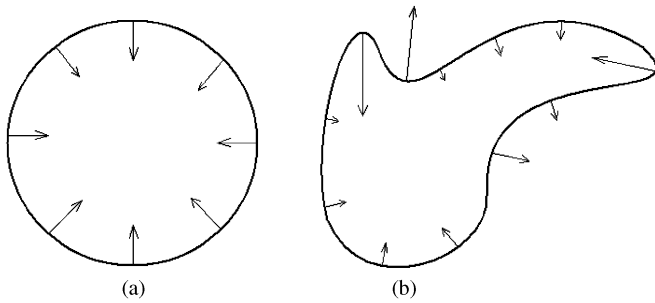


FIG. 1. Capillary forces applied to the interface between two fluids (shown by arrows): (a) equilibrium case and (b) nonequilibrium shape of the liquid drop.

force which pushes it to the right until it reaches the equilibrium configuration shown in Fig. 3(b).

Therefore, the relaxation to the equilibrium in a porous medium state represents the fluid flow within the pore network (and not the simple deformation as in the bulk). As the flow in thin tubes is highly retarded by the friction against the walls, the capillary relaxation in a porous medium is expected to be slow, so it cannot be neglected. Moreover, the equilibrium state seems to be difficult to reach. During the process of relaxation the fluid cluster permanently changes its geometry, which means that the macroscopic hydrodynamic characteristics of both fluids should also change in time. Already in the 1950s it was known that the geometry of a phase cluster may change the flow characteristics at the same fixed phase saturation  $S$  [2]. Thus, the macroscopic characteristics of two-phase flow should depend explicitly on time (and not only on the saturation).

This was confirmed by a number of experimental and numerical data obtained by scientists and petroleum engineers. In a number of papers published long ago, the main characteristic functions of two-phase flow, such as the relative permeabilities (RP) and the capillary pressure  $p_c(S)$  (PC), were shown to be variable in time, with a very slow stabilization [3–5]. These authors calculated numerically RP and PC by using the network models of porous media. The same result was obtained experimentally by Topp *et al.* [6], who measured  $p_c(S)$  curves for quasistatic, steady state, and dynamic conditions. The capillary pressure was shown to be dependent on phase saturation  $S$  and time.

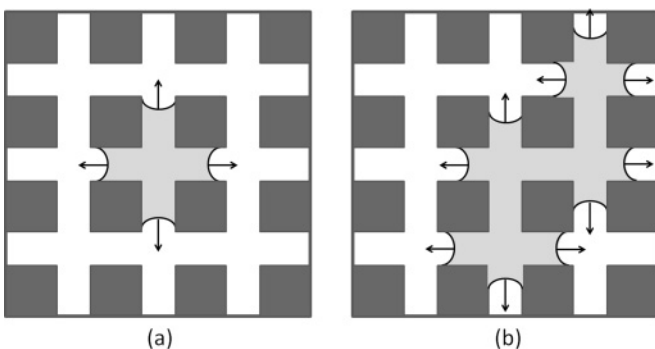
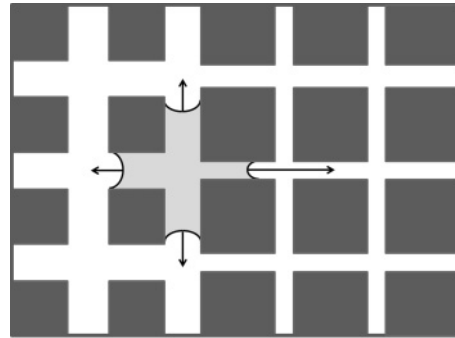
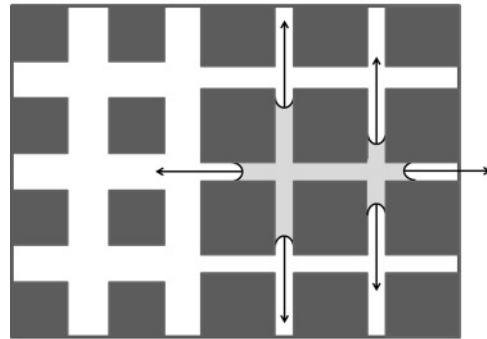


FIG. 2. Gray and white fluids in a homogeneous porous medium. Two different phase clusters are in equilibrium.



(a)



(b)

FIG. 3. Gray and white fluids in a heterogeneous porous medium: (a) nonequilibrium phase repartition and (b) equilibrium phase repartition.

However, this conclusion is in clear contradiction with the classical two-phase theory which is based on the assumption of the local capillary equilibrium. By definition, any equilibrium state is described by thermodynamic relationships. Furthermore, the characteristic functions of two-phase flow are time and rate independent. In the classical theory, they depend on the phase saturation  $S$  only.

Thus, the capillary nonequilibrium causes the self-redistribution of both phases in space. The time of capillary relaxation to the equilibrium state is the characteristic time of fluid self-redistribution within the porous network.

This short physical analysis determines several approaches through which the nonequilibrium might be introduced in the macroscopic flow models: (i) through the introduction of the concept of self-redistribution of phases, (ii) through the introduction of medium heterogeneity, (iii) through the introduction of interface movement, and so on. They are all practically equivalent to one another and, hence, reduce to the similar mathematical formalism.

**B. Relaxation model of self-redistribution of phases in space**

The first series of papers which developed the time-dependent, nonequilibrium capillary relationship was done by Barenblatt [1] with modifications in Refs. [7,8]. The authors accepted that the relative permeabilities and the capillary pressure depend on an apparent saturation  $\sigma$  which corresponds to the equilibrium value that might be reached by the system during the time  $\tau^*$  after phase redistribution in space. Time  $\tau^*$  is the phase redistribution time and is saturation

dependent. The difference between the true saturation  $S$  and  $\sigma$  is determined only by the local rate of saturation variation  $\partial S/\partial t$ , and this dependence was assumed to be linear:

$$S - \sigma = -\tau_{\star} \frac{\partial S}{\partial t}. \quad (\text{I.1})$$

Assuming that the relaxation  $\tau_{\star}$  is sufficiently small (a weak nonequilibrium) one can obtain for capillary pressure or any other function of saturation:

$$p_c(S) = p_c(\sigma) - \tau_{\star} p'_c(\sigma) \frac{\partial S}{\partial t} + \dots, \quad (\text{I.2})$$

or

$$p_c^{\text{dyn}} - p_c^{\text{eq}} = -\tau(S) \frac{\partial S}{\partial t}, \quad (\text{I.3})$$

where  $p_c^{\text{dyn}}$  and  $p_c^{\text{eq}}$  are the nonequilibrium (dynamic) and equilibrium (static) capillary pressures, and  $\tau$  is the new kinetic (or relaxation) time which depends on the saturation.

This fundamental relationship shows that the capillary pressure becomes rate dependent, or “dynamic.”

In Ref. [8] the authors defined the redistribution time as the characteristic time of rearrangement of the phase clusters and consequent modification of their flow properties. Namely, any change of the capillary pressure results in the rearrangement of both phase clusters resulting in changes of their connectivity and their overall hydraulic conductances for the fluids flowing in them. The value of the redistribution time  $\tau$  depends on the saturation. For example, at low saturation, water flows through the narrowest clusters in the corners of pore space, and the capillary pressure is high. Therefore, the time needed to reconfigure water distribution is small and the capillary relaxation time goes to zero as the water saturation approaches its minimal value. On the other side, as the oil saturation is approaching its residual value, the capillary forces weaken and the connectivity of oil grows sparse. Consequently, the redistribution time increases infinitely. Finally, according to Barenblatt and Vinichenko, the relaxation time between the endpoint saturations is roughly inversely proportional to the derivative of capillary pressure, which is close to a constant value.

The theoretical justification of the heuristic relationship (I.3) has been obtained in several papers by using the approach of nonequilibrium thermodynamics: first by Nikolaevsky *et al.* [9], and later by Marle [10]. More precisely, the relationship for entropy production was formulated in the bilinear form, which consists of the products between thermodynamic forces and fluxes, and by assuming the Onsager linear relationships between the forces and fluxes of the same tensor dimension. The development of the relationship for the entropy production is based on the conservation equations and several assumptions regarding the microscopic behavior of the system, which determines the fact that various authors obtain frequently different results. In particular, [11] used the same approach as [10] but obtained a slightly different definition for the relaxation time as well as for the static capillary pressure. He also gave an original interpretation for the kinetic term  $\tau \partial_t S$  by considering it as the contribution of the viscous forces to the pressure drop.

The attempts to define the relaxation time  $\tau$  were performed in Refs. [12–14], but the data obtained concerned a single

porous medium. Using the experimental evidence Stauffer [15] came up with the relationship (I.3). Moreover, he suggested a formula for the coefficient of proportionality which was considered to be constant, whereas  $\tau$  in Eq. (I.3) is saturation dependent. Reference [16] investigated experimental aspects of the dependence of the capillary pressure on the flow rate for two-phase (water-oil) flow in porous media. The authors suggested a relationship identical to (I.3). The dynamic term  $\tau \partial_t S$  was interpreted as the contribution of the viscous forces to the pressure drops, because the parameter  $\tau$  has exactly the dimensions of viscosity.

General principles of construction of nonequilibrium models which remember their history were studied by Entov [17]. The author extended the ideas of Barenblatt [1] concerning the noninstantaneous phase redistribution in space by additionally including its dependence on the direction of fluid displacement. As a result, the process should depend on all its history and should be described by integral operators. However, this paper has not suggested any concrete mathematical model. Note that the dependence of the characteristics of displacement on its history were studied by Dodd and Kiel [18] where the long-term-memory effects were taken into the account in the numerical model of capillary networks.

### C. Relaxation model of active and passive phases

In a series of papers, other physical assumptions were used to develop the nonequilibrium model. For each phase, the existence of an active and passive (trapped in pores) component was assumed, with an exchange between them (i.e., the probable immobilization of the active phase and vice versa).

Barenblatt and Entov [19] assumed this exchange process to be noninstantaneous and its rate to be proportional to the difference between the active and passive saturations. After mathematical developments, it was shown that this approach yields the same formal mathematical relationship (I.3).

Thus, different physical mechanisms lead to the same mathematical model. The physical interpretation of the parameter  $\tau$  becomes the exchange time.

The extremal version of this model was proposed by Zarubin [20], who eliminated entirely the concept of RP and PC and considered the exchange terms between the passive and active components as an alternative to them.

The same ideas were presented by Bulygin [21] in terms of the dynamic effective porosity with respect to the active component. This dynamic porosity was studied experimentally by Pleshinskii *et al.* [22], but the data obtained were insufficient to estimate the exchange time  $\tau$ . The main defect of this model consists of assuming the same time  $\tau$  for phase mobilization and trapping. However, these two processes are nonsymmetric.

Another approach to obtain long-term memory was developed by Buevich and Mambetov [23] as the extension of the exchange model. Instead of the passive and active phase components, each phase was presented as the system of several clusters of different scales. Between the clusters there exists an exchange process. Only the exchange between the neighboring cluster was kept. Each exchange was described by the kinetic law. It was expected that the macroscale model

would give integrodifferential equations. However, only the general methodology was presented.

A more general model of the same class was suggested by Hilfer *et al.* in Refs. [24–26] who introduced the connected and disconnected elements of the same phase and the exchanges between them. Since this concept is more general than the active and passive phases, it enabled the authors to relate the macroscopic two-phase-flow theory to the percolation theory.

#### D. Relaxation model with transport of interfaces

As mentioned above, saturation alone is insufficient to describe the geometry of phase repartition in space. The second parameter might be the specific area of the interface between the phases, usually noted by  $a$ . In the Ref. [27], a system with a number of interfaces was examined within the framework of a phenomenological approach, and a new idea was suggested to examine the highly dispersed interface as an individual continuum, while formulating the mass- and momentum-balance equations for it. To homogenize such a system with multiple discontinuities, the author applied the technique of distributions. In a particular case, the author obtained the known model with cross relative permeabilities, which proved the validity of this new approach.

In Refs. [28–30], the authors introduced two theoretical concepts simultaneously: the additional continuum of Marle representing the ensemble of interfaces, and Barenblatt’s concept of the capillary nonequilibrium. By applying the technique of nonequilibrium thermodynamics, the authors obtained the new model of two-phase flow which contained the same relationship (I.3), but in which the relaxation time and the capillary pressures depend on the saturation  $S$  and the specific area  $a$ :

$$p_c^{\text{dyn}} - p_c^{\text{stat}} = -\tau(S, a) \frac{\partial S}{\partial t}. \quad (\text{I.4})$$

For the specific area  $a$ , the authors obtained the new momentum-balance equation. The function  $a$  was also studied on the basis of capillary network modeling. It was shown that this function is related with the saturation, but not in a unique way. The authors gave estimates of  $\tau$  for air-water systems (soil type: sand; type of experiment: drainage), as well as for oil-water systems (soil type: sandstone or limestone; type of experiment: imbibition) by interpreting their own data and the data obtained by several authors in a period from 1967 to 1993.

The authors showed that their model was free of contradictions proper for other publications based on the relaxation model (I.3).

The more developed form of the model with transport of interfaces was suggested in Ref. [31] where the system of interfaces was considered as the new continuum (*M continuum*) having specific physical properties and generating the vector field of capillary forces associated with each individual meniscus. Therefore, the concept of capillarity changed its topological dimension. Such a model is qualitatively different from the approach related to capillary nonequilibrium.

#### E. Relaxation model of flow in heterogeneous medium

The processes of fluid self-redistribution in space or the exchange between passive and active components of fluid

have the same origin: medium heterogeneity on a small scale. In particular, it is sufficient to consider a medium consisting of two components: highly permeable fractures and low-permeability inclusions (“blocks”). The higher the difference in permeability of block and fractures, the higher the capillary nonequilibrium will be.

Consequently, the general nonequilibrium model may be obtained by homogenizing the flow in a heterogeneous medium with highly different permeabilities of medium components.

Such model was developed by Panfilov [32] and Bourgeat and Panfilov [33] by the asymptotic homogenization of two-phase flow in the medium described below. The blocks formed a periodic field with a period  $\varepsilon$  which was much smaller than the macroscopic size of the domain (a double-porosity medium). The heterogeneity degree was described by two parameters: the ratio  $\omega$  of the low and high permeability and the ratio  $\delta$  of the mean value of the capillary pressure in blocks versus that in fractures. The case of high heterogeneity,  $\omega \sim \varepsilon^2$ , is characterized by a significant non-neglecting nonequilibrium, which leads to the appearance of the long-term memory. In order to avoid the problem of coupling between the nonlinearity of two-phase flow equations and the long-term memory, the authors considered the case of low nonequilibrium, intermediate between  $\omega \sim 1$  and  $\omega \sim \varepsilon^2$ ; more exactly, the following class of media was studied:

$$\omega \ll 1, \quad \varkappa \ll \delta \ll 1, \quad \text{with } \varkappa \equiv \frac{\varepsilon^2}{\omega}. \quad (\text{I.5})$$

For this case they deduced the completely homogenized model by formal asymptotic expansions. In particular, the following nonequilibrium relationship between capillary pressure in fractures and blocks was obtained:

$$\frac{1}{\delta} p_c^{\text{I}}(S^{\text{I}}) - p_c^{\text{II}}(S^{\text{II}}) = -\tau_*(S^{\text{I}}) \frac{\partial S^{\text{I}}}{\partial t}, \quad (\text{I.6})$$

where  $S^{\text{I}}$  and  $S^{\text{II}}$  are the macroscopic water saturation in blocks and fractures, respectively, and capillary pressure  $p_c$  is dimensionless (divided by the mean value of capillary pressure in blocks and fractures, respectively) and of order unity.

Taking into account the fact that the process in blocks is in nonequilibrium with respect to that in fractures, Eq. (I.6) is evidently identical to (I.3).

The capillary relaxation term  $\tau_*$  depends on  $S^{\text{I}}$  and is proportional to  $\varkappa = \varepsilon^2/\omega$ . It is defined through a cell problem which has an explicit analytical solution when the block shape is spherical.

In the same paper, the exact relationship for the relaxation time was obtained. The hypothesis of Barenblatt and Vinichenko [8] mentioned in Sec. IB was not confirmed: the dependence of  $\tau$  on the saturation was shown to be a nonmonotonic function increasing to infinity when the system becomes single phase.

#### F. Models with long-term memory: complete and incomplete homogenization

The mentioned relaxation models of various types describe the case of weak nonequilibrium which leads to the appearance of short-term memory in macroscopic models. This means

that, strictly speaking, the model of (1.3) is valid only for short relaxation time  $\tau$ . Physically, this means that the process remembers only the last instants of its history, but not the entire history. The question that remains open, however, is what mathematical model of capillary nonequilibrium for the case of long-term memory should be proposed when the process depends on the overall history. In this case the appearance of some integrodifferential relationships is expected.

Long memory may appear in the model of flow in a heterogeneous medium if the ratio  $\omega$  between the block and the fracture permeabilities were very small:  $\omega \sim \varepsilon^2$ . For instance, such a model appears in the case of single-phase flow in double-porosity media [34,35]. This case corresponds to a delay of the process in blocks that is longer than the case described in Sec. IE. Attempts to develop a similar model for two-phase flow have been undertaken in Refs. [36–38]. The formal mathematical results were obtained, but in the form which will we call “incompletely homogenized.”

We call a model *incompletely homogenized* if the fast  $y$  and slow  $x$  variables are not totally separated. In contrast, a *completely homogenized* model contains functions dependent only on slow variables, while the coefficients are calculated only through cell problems independent of slow variables.

The typical example of the incomplete homogenization corresponds to the case when the zero approximation of the asymptotic expansion over  $\varepsilon$  depends on  $y$ :  $S^\varepsilon(x, y, t) = S_0(x, y, t) + \varepsilon \cdots$ . In this case, the macroscopic model will contain the function  $S_0(x, y, t)$  whose dependence on  $y$  is determined through a cell problem also containing the slow variable  $x$ . The completely homogenized model might be obtained if a transition from the function  $S_0(x, y, t)$  to the averaged function  $\widehat{S}(x, t)$  would be possible. Unfortunately, for nonlinear problems, such a transition frequently does not exist.

An incompletely homogenized model, when rigorously justified, is sufficient for mathematics, but it is useless for physics in the majority of cases just due to the fact that the microscopic and macroscopic scales are not separated and, as the result, an incompletely homogenized model is not simpler than the original microscopic equations. The difference between completely and incompletely homogenized models is the main difficulty which causes some incomprehension between physicists and mathematicians.

In the case of double-porosity media, we will show that the limit term of the asymptotic expansions  $S_0(x, y, t)$  is given by two functions:  $S_0^{\parallel}(x, t)$  in fractures, which is the homogenized value over fractures, and  $S_0^{\perp}(x, y, t)$ , which is incompletely homogenized in blocks. The saturation in blocks is described by a nonlinear boundary-value cell problem for a nonlinear diffusion equation which is interpreted in mechanics and engineering as the *capillary imbibition problem*. This problem has no analytical solutions, nor explicit integral representations that could be used to transit from  $S_0(x, y, t)$  to the averaged saturation in blocks  $\widehat{S}^{\perp}(x, t)$ .

In these cases the unique way to develop completely homogenized models consists of introducing some approximations or analyzing some asymptotic cases.

We will develop two types of complete models: the asymptotic case, which corresponds to the weak nonequilibrium, and the linearized case, which corresponds to the upper and lower bounds of the nonlinear homogenized model.

### G. Models with long-term memory: linearization and approximate solutions

An example of linearization for the long-memory case is given in Ref. [39]. The author linearized the boundary-value imbibition problem obtained for  $S_0^{\perp}(x, y, t)$  in blocks and solved it analytically through Green’s functions, which leads to the completely homogenized model. The admissibility of the linearization was justified by the comparison of numerical results obtained for the exact incompletely homogenized model and the linearized approximation. It was shown that the simulation results assess any loss in accuracy. Moreover, it was shown that the simplified model approximates the capillary effects quite well, which is important for our analysis.

Such a linear problem in blocks was solved and used to close the macroscopic flow model in Ref. [40].

Much more recent papers in the field of petroleum engineering were devoted to finding good approximations for the solution of the problem in blocks. The main attention focused on the exchange term between blocks and fractures and not on the capillary nonequilibrium relationship, although these two concepts are closely related with one another. Indeed, the exchange rate  $q$  is equal to  $\phi^1 \partial_t S^1$  where  $S^1$  is the mean saturation in a block. Using the main formula (1.3), we obtain the relationships for the difference between the equilibrium and dynamic capillary pressure:  $p_c^{\text{dyn}} - p_c^{\text{stat}} = -q\tau/\phi^1$ .

The relationship for the exchange rate  $q$  was obtained from the approximative analytical solution of the linearized one-dimensional (1D) problem of imbibition into a block. Such a solution of the Dirichlet problem for the linear diffusion equation represents a Fourier series of a function exponentially decreasing in time. The simplest approximation consists of keeping only the main eigenmode, which leads to the relationship suggested by Skvortsov [41]:

$$q(x, t) \simeq N_0 \frac{\exp(-\lambda_0 [t - t_0(x)])}{\sqrt{t - t_0(x)}}, \quad t > t_0(x),$$

where  $\lambda_0$  is the main eigenvalue of the Laplace operator, and  $N_0$  is a constant parameter. Function  $t_0(x)$  represents the moment at which the displacement front reaches the point  $x$ . This function may be obtained as the solution of the Buckley-Leverett problem in a homogeneous medium without exchange processes.

Other correlations were suggested in Refs. [42,43]. They represent  $q$  as the difference between two exponential functions responsible respectively for two inverse processes: the transfer of the cumulative oil mass from the block center towards the interface, and the transfer of the same oil mass in the opposite direction, which have different eigentimes. In Ref. [44] the rate  $q$  was presented in the form of the convolution of the mentioned difference between two exponential functions with the derivative  $\partial_t S$ .

The simplest exponential correlation was suggested in Ref. [45].

The development of the nonequilibrium theory was successful because it explained several phenomena. For instance, the nonequilibrium theory forecasted that oil recovery from reservoir might be a nonmonotonic function of the recovery rate as it was observed in practice. On the other hand, this was

not explained by the classical theory, which is invariant with respect to the flow rate.

### H. Some theoretical and numerical results concerning dynamic capillary pressure

Recently, the mathematical study of the immiscible two-phase flow model in porous media with a dynamic capillary pressure-saturation relationship started to attract the interest of the scientific community. These studies are based either on the full two-phase flow model, or on Richards' equation which is the limiting case of two-phase flow when the viscosity and density of one phase are assumed to be zero (flow of water and air).

First, we mention a series of papers devoted to Richards' model with the nonequilibrium term in the form of the third-order mixed derivative:

$$\partial_t s = \operatorname{div}(k(s)[-P'_c(s)\nabla s + \tau \nabla \partial_t s]). \quad (\text{I.7})$$

In Ref. [46], Mikelić used entropy techniques to prove the existence of an appropriate weak solution of equation (I.7) with square-integrable first derivatives in time and space and square-integrable time derivatives of the gradient, for any time interval. The existence of a weak solution to a closely related model was proven by Cancès *et al.* [47]. The existence of traveling wave solutions for equation (I.7) was extensively studied (see, e.g., Refs. [48–50]).

The capillarity limit for the linear relaxation model (the relaxation time  $\tau$  is constant) was studied by Duijn *et al.* [51]. It leads to the Buckley-Leverett equation with discontinuous solutions which do not satisfy Oleinik's entropy condition [52]. Next, the results obtained were extended for linear higher-order terms in Ref. [53] where the existence of traveling-wave solutions in the nonlinear and degenerate case was also investigated. Cases considered may lead to nonsmooth traveling waves, as well as to a discontinuous capillary pressure.

The homogenization of the pseudoparabolic equation in periodic media was studied by Peszynska *et al.* [54]. They obtained upscaled limits by asymptotic expansion and two-scale convergence in various linear cases for both the classical binary medium model and the highly heterogeneous case. Moreover, the double-porosity limit of Richards' equation with dynamic capillary pressure is obtained.

Numerical treatment of dynamic capillary pressure effects in heterogeneous porous media were studied by Helmig *et al.* [55]. The authors were able to integrate the dynamic capillary function into the finite volume discretization scheme. The paper of Peszynska and Yi [56] considered numerical aspects of the Richards' equation as well as the full two-phase flow model. For the discretization, they used two classes of methods: a cell-centered finite differences method and a locally conservative Eulerian-Lagrangian method. Moreover, they discussed convergence of the methods and extensions to heterogeneous porous media with different rock types. Finally, they concluded that, in convection-dominated cases and for large dynamic effects, some instabilities may arise.

### I. Results obtained in present paper

In the present paper we study the capillary nonequilibrium by using the approach based on introducing the medium

heterogeneity. As shown in Sec. I A, the medium heterogeneity necessarily causes the capillary nonequilibrium. We consider a more complicated case than the case (I.5) analyzed in Ref. [33], which causes the appearance of the long-term memory. For this case, the macroscopic model was proven in Refs. [57,58]. These models, being mathematically strictly justified, remain, however, incompletely homogenized, and thus play a limited role for physics.

We present the following results:

(i) Two general nonequilibrium relationships are presented for the macroscopic dynamic capillary pressure and its relationship with equilibrium capillary pressure. We also present the exact method of calculating this relationship which requires solving three cell problems.

(ii) We show that the nonequilibrium relationship of Bourgeat and Panfilov [33] results from our general relationship as a particular asymptotic case which corresponds to the small relaxation time.

(iii) The theorem of comparison for the macroscopic nonlinear model is presented, which establishes its lower and upper bounds. These bounds represent the linear mathematical system of equations. The completely homogenized macroscopic model for these bounds proves explicitly the long-memory effects and allows the explicit calculation of the relaxation kernel.

(iv) We show a number of numerical simulations that compare our homogenized results with the direct numerical data, as well as with the results of Bourgeat and Panfilov [33].

## II. MICROSCOPIC MODEL

### A. Medium structure

We consider a reservoir  $\Omega \subset \mathbb{R}^d$  ( $d = 2, 3$ ) which is assumed to be a bounded, connected domain with a periodic microstructure scaled by a parameter  $\varepsilon$  that represents the ratio of the period to the whole region. We assume that  $\varepsilon > 0$  is a small parameter tending to zero. The unit cell of this microstructure is  $Y = (0, 1)^d$  and consists of two subdomains (corresponding to two types of rock): a connected subdomain  $Y^{\text{II}}$  ("the fracture") and another subdomain  $Y^{\text{I}}$  ("the block") which is surrounded by  $Y^{\text{II}}$ .

We denote by  $\Gamma$  the interface between the two subdomains in  $Y$  and  $\Gamma_T \stackrel{\text{def}}{=} \Gamma \times (0, T)$ . Thus,  $Y = Y^{\text{I}} \cup Y^{\text{II}} \cup \Gamma$ .

Let  $\mathbf{1}^{\text{I}}(y)$  be the characteristic function of  $Y^{\text{I}}$  extended  $Y$ -periodically to the whole  $\mathbb{R}^d$ . Then the medium  $\Omega$  contains two subdomains  $\Omega^{\varepsilon, \text{I}}$  and  $\Omega^{\varepsilon, \text{II}}$ , which represent two different types of rock and satisfy the conditions

$$\begin{aligned} \Omega^{\varepsilon, \text{II}} &\subset \left\{ x \in \Omega : \mathbf{1}^{\varepsilon, \text{II}} \left( \frac{x}{\varepsilon} \right) = 1 \right\}, \\ \Omega^{\varepsilon, \text{I}} &= \Omega \setminus \overline{\Omega^{\varepsilon, \text{II}}}. \end{aligned} \quad (\text{II.1})$$

We introduce the notation

$$\Omega_T \stackrel{\text{def}}{=} \Omega \times (0, T), \quad \Omega_T^{\varepsilon, \ell} \stackrel{\text{def}}{=} \Omega^{\varepsilon, \ell} \times (0, T), \quad (\text{II.2})$$

$$\Gamma^\varepsilon \stackrel{\text{def}}{=} \partial \Omega^{\varepsilon, \text{I}} \cap \partial \Omega^{\varepsilon, \text{II}} \cap \Omega,$$

$$\Gamma_T^\varepsilon \stackrel{\text{def}}{=} \Gamma^\varepsilon \times (0, T), \quad (\text{II.3})$$

where  $T > 0$  is fixed and  $\ell = \text{I, II}$ .

### B. Flow equations

The main assumptions of this paper are

- (i) The flow is two phase and completely immiscible.
- (ii) Both phases are incompressible, so that their densities and viscosities are constant.
- (iii) The porous medium is nondeformable.
- (iv) The system is locally in capillary equilibrium; that is, the difference of phase pressures is equal to the local capillary pressure function at any point on the microscale.

Under these assumptions, the mass conservation for each phase is formulated in the classical way (see, e.g., Refs. [41,59–61]); the momentum conservation equations are formulated through Darcy-Muskat's equations in  $\Omega_T$ :

$$\begin{aligned} 0 &= \phi^\varepsilon \left( \frac{x}{\varepsilon} \right) \frac{\partial s^\varepsilon}{\partial t} + \operatorname{div} \vec{v}_w^\varepsilon, \\ 0 &= -\phi^\varepsilon \left( \frac{x}{\varepsilon} \right) \frac{\partial s^\varepsilon}{\partial t} + \operatorname{div} \vec{v}_o^\varepsilon, \\ \vec{v}_w^\varepsilon &= -K^{\varepsilon, \varkappa} \left( \frac{x}{\varepsilon} \right) \lambda_w \left( s^\varepsilon, \frac{x}{\varepsilon} \right) (\nabla p_w^\varepsilon - \gamma_w \vec{e}_g), \\ \vec{v}_o^\varepsilon &= -K^{\varepsilon, \varkappa} \left( \frac{x}{\varepsilon} \right) \lambda_o \left( s^\varepsilon, \frac{x}{\varepsilon} \right) (\nabla p_o^\varepsilon - \gamma_o \vec{e}_g), \end{aligned} \quad (\text{II.4})$$

$$\begin{aligned} \lambda_w \left( s^\varepsilon, \frac{x}{\varepsilon} \right) &\stackrel{\text{def}}{=} \frac{k_w \left( s^\varepsilon, \frac{x}{\varepsilon} \right)}{\mu_w}, \\ \lambda_o \left( s^\varepsilon, \frac{x}{\varepsilon} \right) &\stackrel{\text{def}}{=} \frac{k_o \left( s^\varepsilon, \frac{x}{\varepsilon} \right)}{\mu_o}, \end{aligned}$$

and the condition of the local capillary equilibrium,

$$p_o^\varepsilon - p_w^\varepsilon = p_c \left( s^\varepsilon, \frac{x}{\varepsilon} \right). \quad (\text{II.5})$$

The conditions on the interface  $\Gamma_T^\varepsilon$  are those of continuity of the normal phase fluxes and pressures:

$$\begin{aligned} [\vec{v}_w^\varepsilon \cdot \vec{\nu}] &= 0, \quad [\vec{v}_o^\varepsilon \cdot \vec{\nu}] = 0, \quad [p_w^\varepsilon] = 0, \quad [p_o^\varepsilon] = 0, \end{aligned} \quad (\text{II.6})$$

where  $p_i^\varepsilon$  is the pressure of the phase  $i = w, o$ ,  $s^\varepsilon \equiv s_w^\varepsilon$  is the water saturation (the volumetric fraction of water in porous volume),  $s_o^\varepsilon = 1 - s^\varepsilon$  is the oil saturation,  $\vec{v}_i^\varepsilon$  is the Darcy velocity,  $\gamma_i$  is the given gravity number,  $\mu_i$  is the given dynamic viscosity,  $\vec{e}_g$  is the unit vector of the gravity field,  $k_w(s^\varepsilon, x)$  and  $k_o(s^\varepsilon, x)$  are the given functions of the relative permeability of water and oil, respectively,  $p_c(s^\varepsilon, x)$  is a given local capillary pressure,  $\phi^\varepsilon(x)$  is the given medium porosity,  $K^{\varepsilon, \varkappa}(x)$  is the given absolute permeability tensor, the symbol  $[\cdot]$  means a jump, and  $\vec{\nu}$  is the unit vector outer normal to  $\Gamma^\varepsilon$ .

All equations are assumed to be formulated in dimensionless form through dimensionless rescaled variables. The scale for the space coordinates is the domain macroscopic length  $L$ , the scale for the pressure is the characteristic pressure difference  $\Delta P$  reached at the domain boundary, the scales for the absolute permeability and porosity are their arithmetic mean values in fractures  $\bar{K}^\parallel$  and  $\bar{\phi}^\parallel$ , and the scale for the viscosities is the mean value between the viscosity of water and oil  $M$ . Finally, the dimensionless gravity numbers are defined as  $\gamma_i = \rho_i L g / \Delta P$  where  $\rho_i$  is the true density with dimension

and  $g$  is the absolute value of the acceleration due to gravity. The time scale is

$$t_* \stackrel{\text{def}}{=} \frac{L^2 \bar{\phi} M}{K \Delta P}, \quad (\text{II.7})$$

which is the characteristic intrinsic time of perturbation propagation over length  $L$  through the porous medium having average parameters and saturated by an average fluid.

Each function  $f^\varepsilon \equiv s^\varepsilon, p_w^\varepsilon, p_o^\varepsilon, \vec{v}_w^\varepsilon, \vec{v}_o^\varepsilon$  is defined as

$$f^\varepsilon \stackrel{\text{def}}{=} f^{\varepsilon, \text{I}}(x) \mathbf{1}^{\varepsilon, \text{I}}(x) + f^{\varepsilon, \text{II}}(x) \mathbf{1}^{\varepsilon, \text{II}}(x),$$

where  $\mathbf{1}^{\varepsilon, \ell}(x) = \mathbf{1}^\ell \left( \frac{x}{\varepsilon} \right)$  is the characteristic function of the subdomain  $\Omega^{\varepsilon, \ell}$ ,  $\ell = \text{I, II}$ .

### C. Boundary and initial conditions

For the sake of simplicity, we assume:

- (i)  $\Omega^{\varepsilon, \text{I}} \cap \partial \Omega = \emptyset$  (i.e., the boundary  $\partial \Omega$  passes only through fractures),
- (ii) the boundary consists of two parts  $\partial \Omega = \partial \Omega_{\text{inj}} \cup \partial \Omega_{\text{imp}}$ , such that  $\partial \Omega_{\text{inj}} \cap \partial \Omega_{\text{imp}} = \emptyset$ .

For  $t \in (0, T)$ , the boundary conditions are given by

$$\begin{aligned} p_w^{\varepsilon, \text{II}}(x, t) &= p_{w*}(x, t), \\ s^{\varepsilon, \text{II}}(x, t) &= S_*(x, t), \quad x \in \partial \Omega_{\text{inj}}, \\ \vec{v}_w^{\varepsilon, \text{II}} \cdot \vec{\nu} &= \vec{v}_o^{\varepsilon, \text{II}} \cdot \vec{\nu} = 0, \quad x \in \partial \Omega_{\text{imp}}, \end{aligned} \quad (\text{II.8})$$

where the function  $p_{w*}$  determines the pressure of the water injection in the reservoir, while  $S_*$  determines the fraction of water in the injected fluid.

Finally, the initial conditions read

$$\begin{aligned} s^{\varepsilon, \text{II}}(x, 0) &= S_0^{\text{II}}(x) \quad \text{in } \Omega^{\varepsilon, \text{II}}, \\ s^{\varepsilon, \text{I}}(x, 0) &= S_0^{\text{I}}(x) \quad \text{in } \Omega^{\varepsilon, \text{I}}. \end{aligned} \quad (\text{II.9})$$

If these initial values do not verify the local capillary equilibrium, then the formulated problem describes the relaxation of an initially nonequilibrium system to an equilibrium state.

### D. Main conditions on parameters

We assume that all physical medium properties are different in two domains and verify the following conditions:

#### A.1: Double-porosity conditions

*Porosity.* The function  $\phi = \phi(y)$  is  $Y$  periodic and given by

$$\phi^\varepsilon(x) \stackrel{\text{def}}{=} \begin{cases} \phi^\text{I} & \text{in } \Omega^{\varepsilon, \text{I}} \\ \phi^\text{II} & \text{in } \Omega^{\varepsilon, \text{II}}, \end{cases} \quad (\text{II.10})$$

where  $\phi^\text{I}$  and  $\phi^\text{II}$  are positive parameters that do not depend on  $\varepsilon$ .

*Absolute permeability.* The ratio between the absolute permeabilities of block and fractures is  $\varepsilon^2/\varkappa$ , where  $\varkappa$  is an independent parameter. Then the permeability tensor may be

presented as

$$K^{\varepsilon, \varkappa}(x) \stackrel{\text{def}}{=} \begin{cases} \varepsilon^2 K^{\varkappa, \text{I}} \stackrel{\text{def}}{=} \frac{\varepsilon^2}{\varkappa} K^{\text{I}} \mathbb{I} & \text{in } \Omega^{\varepsilon, \text{I}} \\ K^{\text{II}} \mathbb{I} & \text{in } \Omega^{\varepsilon, \text{II}}, \end{cases} \quad (\text{II.11})$$

where  $\mathbb{I}$  is the unit tensor, while  $K^{\text{I}}$ ,  $K^{\text{II}}$ , and  $\varkappa$  are positive parameters that do not depend on  $\varepsilon$ .

We will deal with the classical double porosity medium, or  $\varepsilon^2$  medium, when  $\varkappa \sim 1$  [which is different from (I.5)].

*Capillary pressure.* The ratio between the mean values of the capillary pressure curves in fractures and blocks is  $\delta$ , so the function  $p_c$  may be presented as

$$p_c\left(s^\varepsilon, \frac{x}{\varepsilon}\right) \stackrel{\text{def}}{=} \begin{cases} p_c^{\text{I}, \delta} \stackrel{\text{def}}{=} \delta^{-1} p_c^{\text{I}}(s^{\varepsilon, \text{I}}) & \text{in } \Omega^{\varepsilon, \text{I}} \\ p_c^{\text{II}}(s^{\varepsilon, \text{II}}) & \text{in } \Omega^{\varepsilon, \text{II}}, \end{cases} \quad (\text{II.12})$$

with functions  $p_c^\ell(s)$  ( $\ell = \text{I}, \text{II}$ ) of order unity.

Physically, the capillary pressure is inversely proportional to the square root of the absolute permeability (the lower the pore size, the higher the capillary pressure); that is,  $p_c \sim 1/\sqrt{K}$ . Hence, it follows that  $\delta \sim \varepsilon$ . We will assume however, as in Ref. [33], that the parameter  $\delta$  is independent of  $\varepsilon$ , which covers the most general probable case.

As the dimensionless capillary pressure behaves as  $p_c^{\text{II}} \sim 1$ , then the mean capillary pressure in fractures is of the same order as the macroscopic pressure drop  $\Delta P$ . In other words, the macroscopic capillary number (the ratio between the macroscopic pressure drop and the mean capillary pressure) in fractures is of order unity.

Functions  $p_c^\ell(s)$  ( $\ell = \text{I}, \text{II}$ ) in Eq. (II.12) belong to the space  $C^1((0, 1]; \mathbb{R}^+)$  and satisfy the following conditions:

$$\frac{dp_c^\ell}{ds}(s) < 0 \quad \text{in } (0, 1] \quad \text{and} \quad p_c^\ell(s = 1) = 0.$$

*Remark 1.* In contrast to the functions  $p_o^\varepsilon, p_w^\varepsilon$ , the saturation  $s^\varepsilon$  may have a jump at the interface  $\Gamma^\varepsilon$ . Namely, from the transmission conditions (II.6), the following holds for the phase pressure:

$$p_c^{\text{II}}(s^{\varepsilon, \text{II}}) = p_c^{\text{I}, \delta}(s^{\varepsilon, \text{I}}) \quad \text{on } \Gamma_T^\varepsilon, \quad (\text{II.13})$$

which gives the discontinuity of the saturation at the interface.

### I. A.2: Behavior of the relative permeability or mobility

The relative phase mobilities are defined as

$$\lambda_j\left(s^\varepsilon, \frac{x}{\varepsilon}\right) \stackrel{\text{def}}{=} \begin{cases} \lambda_j^{\text{I}}(s^{\varepsilon, \text{I}}) & \text{in } \Omega^{\varepsilon, \text{I}} \\ \lambda_j^{\text{II}}(s^{\varepsilon, \text{II}}) & \text{in } \Omega^{\varepsilon, \text{II}}, \end{cases} \quad (\text{II.14})$$

with  $j = w, o$ .

Functions  $\lambda_w^\ell(s)$  and  $\lambda_o^\ell(s)$  ( $\ell = \text{I}, \text{II}$ ) belong to the space  $C([0, 1]; \mathbb{R}^+)$  and satisfy the following properties:

- (i)  $\lambda_w^\ell(s)$  increases while  $\lambda_o^\ell(s)$  decreases within  $[0, 1]$ ;
- (ii)  $0 \leq \lambda_w^\ell(s), \lambda_o^\ell(s) \leq 1$  for  $s \in [0, 1]$ ;
- (iii)  $\lambda_w^\ell(0) = \lambda_o^\ell(1) = 0$ ;
- (iv) there exists values  $s_*$  and  $s^*$ , called the *percolation thresholds*, such that  $\lambda_w(s) \equiv 0$  when  $0 \leq s \leq s_*$ , and  $\lambda_o(s) \equiv 0$  when  $s^* \leq s \leq 1$ . This means that each phase remains

immobile if its own saturation is too low (due to the loss of the phase connectivity).

- (v) there exists a value  $L_0 > 0$  such that

$$\lambda^\ell(s) \stackrel{\text{def}}{=} \lambda_w^\ell(s) + \lambda_o^\ell(s) \geq L_0 > 0 \quad \text{for } s \in [0, 1]. \quad (\text{II.15})$$

Physically, this means that both fluids cannot be simultaneously immobile.

### A.3: Moderate gravity number

We assume that both gravity numbers  $\gamma_w$  and  $\gamma_o$  are of order unity, which means physically that the gravity force is assumed to be of the same order as the external pressure drop  $\Delta P$  and the mean capillary pressure in fracture.

### A.4: Nondegeneration of two-phase flow

For the sake of simplicity, we will assume that a constant  $\mathbf{c}_0$  exists such that

$$0 < \mathbf{c}_0 < S_0^{\text{I}}, \quad S_0^{\text{II}}, \quad S_* < 1 - \mathbf{c}_0. \quad (\text{II.16})$$

## III. HOMOGENIZED MODEL

Using the results of Ref. [58] we can formulate the limit of problem (II.4)–(II.6), (II.8) and (II.9) as  $\varepsilon \rightarrow 0$ . The small parameters  $\delta, \varkappa$  are fixed in the homogenization process.

### A. Formulation of homogenization equations

The homogenized model is formulated in terms of two series of macroscopic functions; one part of them being independent of the fast variable  $y$ :

$$S^{\text{II}}(x, t), \quad P_w^{\text{II}}(x, t), \quad P_o^{\text{II}}(x, t), \quad P_c^{\text{II}}(x, t),$$

while the other part remains dependent on  $y$ :

$$S^{\text{I}}(x, y, t), \quad P_w^{\text{I}}(x, y, t), \quad P_o^{\text{I}}(x, y, t), \quad P_c^{\text{I}}(x, y, t),$$

which are the two-scale limits of the microscopic variables:

$$\begin{aligned} s^{\varepsilon, \text{II}} &= S^{\text{II}}(x, t) + \varepsilon \dots, & s^{\varepsilon, \text{I}} &= S^{\text{I}}(x, y, t) + \varepsilon \dots, \\ p_w^{\varepsilon, \text{II}} &= P_w^{\text{II}}(x, t) + \varepsilon \dots, & p_w^{\varepsilon, \text{I}} &= P_w^{\text{I}}(x, y, t) + \varepsilon \dots, \\ p_o^{\varepsilon, \text{II}} &= P_o^{\text{II}}(x, t) + \varepsilon \dots, & p_o^{\varepsilon, \text{I}} &= P_o^{\text{I}}(x, y, t) + \varepsilon \dots, \end{aligned}$$

$$\begin{aligned} p_c^{\text{II}}(s) &= P_c^{\text{II}}(s) + \varepsilon \dots, \\ p_c^{\text{I}, \delta}(s) &= P_c^{\text{I}, \delta}(s) + \varepsilon \dots = \delta^{-1} P_c^{\text{I}}(s) + \varepsilon \dots. \end{aligned}$$

The homogenized problem for (II.4)–(II.9) can be obtained by various methods. For example, it is obtained in Refs. [57, 58] by the method of two-scale convergence (see Ref. [62] for the method). According to the mentioned papers, the vector function  $\langle p_w^\varepsilon, p_o^\varepsilon, s^\varepsilon \rangle$ , which satisfies (II.4)–(II.9), conditions A.1–A.4, and several additional conditions, converges in the two-scale sense to the vector function  $\langle P_w^{\text{I}}, P_o^{\text{I}}, P_w^{\text{II}}, P_o^{\text{II}}, S^{\text{I}}, S^{\text{II}} \rangle$ , which is a weak solution to problem (III.1)–(III.8).



In  $\Omega_T$ ,

$$\begin{aligned} \widehat{\phi}^{\text{II}} \frac{\partial S^{\text{II}}}{\partial t} - \text{div} (\mathbb{K} \lambda_w^{\text{II}}(S^{\text{II}}) (\nabla P_w^{\text{II}} - \gamma_w \vec{e}_g)) &= -\widehat{\phi}^{\text{I}} \frac{\partial \langle S^{\text{I}} \rangle_1}{\partial t}, \\ -\widehat{\phi}^{\text{II}} \frac{\partial S^{\text{II}}}{\partial t} - \text{div} (\mathbb{K} \lambda_o^{\text{II}}(S^{\text{II}}) (\nabla P_o^{\text{II}} - \gamma_o \vec{e}_g)) &= \widehat{\phi}^{\text{I}} \frac{\partial \langle S^{\text{I}} \rangle_1}{\partial t}, \end{aligned} \quad (\text{III.1})$$

$$P_c^{\text{II}}(S^{\text{II}}) = P_o^{\text{II}} - P_w^{\text{II}}.$$

In  $Y^{\text{I}} \times \Omega_T$ ,

$$\begin{aligned} \phi^{\text{I}} \frac{\partial S^{\text{I}}}{\partial t} - \text{div}_y (K^{\infty, \text{I}} \lambda_w^{\text{I}}(S^{\text{I}}) \nabla_y P_w^{\text{I}}) &= 0, \\ -\phi^{\text{I}} \frac{\partial S^{\text{I}}}{\partial t} - \text{div}_y (K^{\infty, \text{I}} \lambda_o^{\text{I}}(S^{\text{I}}) \nabla_y P_o^{\text{I}}) &= 0, \end{aligned} \quad (\text{III.2})$$

$$P_c^{\text{I}, \delta}(S^{\text{I}}) = P_o^{\text{I}} - P_w^{\text{I}}.$$

On  $\Gamma \times \Omega_T$ ,

$$P_w^{\text{I}}(x, y, t) = P_w^{\text{II}}(x, t), \quad P_o^{\text{I}}(x, y, t) = P_o^{\text{II}}(x, t). \quad (\text{III.3})$$

Herein,  $|Y^\ell|$  denotes the measure of the set  $Y^\ell$ ,

$$\langle u \rangle_\ell \stackrel{\text{def}}{=} \frac{1}{|Y^\ell|} \int_{Y^\ell} u(y) dy, \quad \ell = \text{I, II}, \quad (\text{III.4})$$

are the average porosities, and the effective permeability tensors are given by their entries:

$$\begin{aligned} \widehat{\phi}^{\text{I}} &\stackrel{\text{def}}{=} \phi^{\text{I}} |Y^{\text{I}}|, \quad \widehat{\phi}^{\text{II}} \stackrel{\text{def}}{=} \phi^{\text{II}} |Y^{\text{II}}|, \\ \mathbb{K}_{ij} &\stackrel{\text{def}}{=} \int_{Y^{\text{II}}} K^{\text{II}} [\nabla_y \xi_i + \vec{e}_i] \cdot [\nabla_y \xi_j + \vec{e}_j] dy, \end{aligned} \quad (\text{III.5})$$

for  $i, j = 1, \dots, d$ , where  $\xi_j$  is the solution of the following problem:

$$\begin{aligned} -\Delta_{yy} \xi_j &= 0 \quad \text{in } Y^{\text{II}}, \\ \nabla_y \xi_j \cdot \vec{\nu} &= -\vec{e}_j \cdot \vec{\nu} \quad \text{on } \Gamma, \\ y &\mapsto \xi_j(y), \quad Y \text{ periodic}, \end{aligned} \quad (\text{III.6})$$

with  $\vec{e}_j$  being the  $j$ th coordinate vector.

The initial conditions read

$$\begin{aligned} S^{\text{II}}(x, 0) &= S_0^{\text{II}}(x) \quad \text{in } \Omega, \\ S^{\text{I}}(x, y, 0) &= S_0^{\text{I}}(x, y) \quad \text{in } \Omega \times Y^{\text{I}}. \end{aligned} \quad (\text{III.7})$$

The boundary conditions for system (III.1) are given by

$$\begin{aligned} P_w^{\text{II}}(x, t) &= p_{w*}, \quad S^{\text{II}}(x, t) = S_* \quad \text{at } \partial\Omega_{\text{inj}} \times (0, T), \\ \vec{V}_w \cdot \vec{\nu} &= \vec{V}_o \cdot \vec{\nu} = 0 \quad \text{at } \partial\Omega_{\text{imp}} \times (0, T), \end{aligned} \quad (\text{III.8})$$

where  $\partial\Omega_{\text{inj}}$  and  $\partial\Omega_{\text{imp}}$  are two parts of the boundary  $\partial\Omega$  that correspond to the fluid injection into the domain and to the impermeability, respectively. Finally, the macroscopic flow velocities are defined as

$$\vec{V}_j = -\mathbb{K} \lambda_j^{\text{II}}(S^{\text{II}}) (\nabla P_j^{\text{II}} - \gamma_j \vec{e}_g), \quad j = w, o. \quad (\text{III.9})$$

As seen, the model obtained is not entirely homogenized since the problem (III.2), formulated through the fast variable  $y$ , also contains, however, the slow variable  $x$  through the boundary condition (III.3). So the cell problem (III.2) must be solved for various points  $x$ , which determines the coupling between the slow and the fast variables.

### B. Equivalent formulation of problem in blocks (III.2)

Problem (III.2) and (III.3) may be reformulated with respect to the single function  $S^{\text{I}}$  by eliminating the pressures:

$$\begin{aligned} \phi^{\text{I}} \frac{\partial S^{\text{I}}}{\partial t} &= \frac{1}{\varepsilon \delta} \text{div}_y (K^{\text{I}} D^{\text{I}}(S^{\text{I}}) \nabla_y S^{\text{I}}) = 0 \quad \text{in } \Omega_T \times Y^{\text{I}}, \\ S^{\text{I}}(x, y, t) &= s^{\text{I}}(S^{\text{II}}) \quad \text{in } \Omega_T \times \Gamma, \\ S^{\text{I}}(x, y, 0) &= S_0^{\text{I}} \quad \text{in } \Omega \times Y^{\text{I}}. \end{aligned} \quad (\text{III.10})$$

The capillary diffusion is defined as

$$D^{\text{I}, \delta}(s) \stackrel{\text{def}}{=} \frac{\lambda_w^{\text{I}}(s) \lambda_o^{\text{I}}(s)}{\lambda_w^{\text{I}}(s) + \lambda_o^{\text{I}}(s)} \frac{dP_c^{\text{I}, \delta}(s)}{ds} \quad (\text{III.11})$$

and

$$D^{\text{I}}(s) = \frac{1}{\delta} D^{\text{I}, \delta}(s) \stackrel{\text{def}}{=} \frac{\lambda_w^{\text{I}}(s) \lambda_o^{\text{I}}(s)}{\lambda_w^{\text{I}}(s) + \lambda_o^{\text{I}}(s)} \frac{dP_c^{\text{I}}(s)}{ds}.$$

The symbol  $s^{\text{I}}(S^{\text{II}})$  means that the the saturation in blocks is in the equilibrium with  $S^{\text{II}}$ . Consequently, it is the solution of the following nonlinear algebraic equation:

$$P_c^{\text{I}, \delta}(s^{\text{I}}(x, t)) = P_c^{\text{II}}(S^{\text{II}}(x, t)). \quad (\text{III.12})$$

The differential equation (III.10) results directly from the lemma (IV.6), which is proven further.

The latter relationship results from (III.3) and the last equation in systems (III.1) and (III.2).

### C. Appearance of long-term memory

The macroscopic model formulated proves memory effects in the implicit form. Just in order to show the mentioned effects explicitly, in this section we consider the case when the coefficients of the problem (III.10) are weakly variable, so the model can be linearized. We will also linearize the boundary condition by taking

$$s^{\text{I}}(S^{\text{II}}) = a S^{\text{II}}(x, t) + b.$$

By introducing a new function  $U = S^{\text{I}} - s^{\text{I}}$ , we easily obtain the similar differential equation for  $U$  as in Eq. (III.10) but with the right-hand side  $f(x, t) \equiv -\partial_t S^{\text{I}} = -a \partial_t S^{\text{II}}$  and homogeneous boundary and initial conditions. The solution of such a problem represents the convolution of the Green's function  $G$  of the diffusion operator with the function  $f: U = \int_0^t G(t - \tau) f(\tau) d\tau$ . Then  $S^{\text{I}} = a S^{\text{II}} + b - a \int_0^t G(t - \tau) \partial_\tau S^{\text{II}} d\tau$ . Substituting this explicit relationship into equation (III.1), we obtain the following for the exchange term in the right-hand sides:

$$a \widehat{\phi}^{\text{I}} \left\{ \frac{\partial S^{\text{II}}(x, \tau)}{\partial t} - \frac{\partial}{\partial t} \int_0^t \langle G(t - \tau) \rangle_1 \frac{\partial S^{\text{II}}}{\partial \tau}(x, \tau) \right\} d\tau, \quad (\text{III.13})$$

which contains a convolution operator.

Finally, from (III.1) follows the system of completely homogenized integrodifferential equations formulated with respect to the three functions  $P_w^{\text{II}}$ ,  $P_o^{\text{II}}$ , and  $S^{\text{II}}$ . The kernel of the integral operators represents the average Green's function of the diffusion operator in a block that does not allows for reducing the convolution integral to a differential operator of higher order. The appearance of such operators physically

means the long-term memory (i.e., the dependence of the process on its overall history).

#### IV. GENERAL THEOREM ON NONEQUILIBRIUM CAPILLARY PRESSURE

##### A. Two nonequilibrium relationships for capillary pressure

The presented homogenized model enables us to obtain the main result of the paper.

*Theorem IV.1.* Under our standing assumptions, the following two nonequilibrium relationships between the capillary pressures hold true in  $\Omega_T$ :

$$\frac{1}{\delta} \langle P_c^I(S^I) \rangle_I - P_c^I(S^I) = -\varkappa \left\langle \frac{\phi^I \partial S^I}{\partial t} \zeta \right\rangle_I \quad (\text{IV.1})$$

and

$$\frac{1}{\delta} P_c^I \left( \langle S^I \rangle_I - \varkappa \delta \left\langle \phi^I \frac{\partial S^I}{\partial t} \psi \right\rangle_I \right) = P_c^I(S^I), \quad (\text{IV.2})$$

where the functions  $\zeta$  and  $\psi$  are the solutions of the following cell problems:

$$\begin{aligned} \nabla_y \left( K^I \frac{\lambda_o^I(S^I) \lambda_w^I(S^I)}{\lambda^I(S^I)} \nabla_y \zeta \right) &= 1 \quad \text{in } Y^I, \\ \zeta &= 0 \quad \text{on } \Gamma. \end{aligned} \quad (\text{IV.3})$$

and

$$\begin{aligned} \nabla_y [K^I D^I(S^I) \nabla_y \psi] &= 1 \quad \text{in } Y^I, \\ \psi &= 0 \quad \text{on } \Gamma. \end{aligned} \quad (\text{IV.4})$$

Theorem IV 1 is proven in Appendix.

*Remark 2.* Due to the assumption (II.16) and the principle of maximum, the coefficients of differential equations in (IV.3) and (IV.4) are positive definite, which ensures the correctness of the corresponding problems.

*Corollary.* From (IV.1) and (IV.2) follows

$$\begin{aligned} \langle P_c^{I,\delta}(S^I) \rangle_{Y^I} &= -\varkappa \left\langle \frac{\phi^I \partial S^I}{\partial t} \zeta \right\rangle_{Y^I} \\ &+ P_c^{I,\delta} \left( \langle S^I \rangle_{Y^I} - \varkappa \delta \left\langle \phi^I \frac{\partial S^I}{\partial t} \psi \right\rangle_{Y^I} \right). \end{aligned} \quad (\text{IV.5})$$

##### B. Physical interpretation of relationships obtained

As the process in the fracture is always in equilibrium, the obtained relationship (IV.1) may be rewritten as

$$P_c^{\text{dyn}}(S^{\text{dyn}}) - P_c^{\text{eq}}(S^{\text{eq}}) = -\frac{\partial \langle a S^{\text{mic}} \rangle}{\partial t},$$

where  $P_c^{\text{dyn}}$  and  $P_c^{\text{eq}}$  are dynamic and equilibrium macroscale capillary pressures, respectively,  $S^{\text{dyn}}$  and  $S^{\text{eq}}$  are the nonequilibrium and equilibrium saturations, respectively,  $S^{\text{mic}}$  is the microscale saturation,  $a$  is a coefficient, and angular brackets mean averaging over heterogeneity. The previous expression represents the generalization of the Barenblatt relationship (I.3).

##### C. Lemma on zero total flux in blocks

First of all, we reveal a fundamental property of problem (III.2), which consists of the following result:

*Lemma IV.1.* The total flux at any point of the block  $Y^I$  is zero; that is,

$$\lambda^I \nabla_y P_w^I + \lambda_o^I(S^I) \nabla_y P_c^{I,\delta} = 0 \quad \text{in } Y^I \times \Omega_T. \quad (\text{IV.6})$$

The proof of Lemma IV.1 is moved to the Appendix.

The physical interpretation of this property is very clear. If one phase enters in the block, then another phase has to leave it (as the fluids are incompressible). In addition, all functions at the block boundary  $\Gamma$  do not depend on  $y$ . Consequently, the fluxes are identical at any point along the boundary. Thus, the total flux as the vector will be zero at any points. Such a process is called the *counter-current capillary imbibition*.

#### V. COMPLETELY HOMOGENIZED CASE I: SHORT-TERM MEMORY

##### A. Weakly nonequilibrium capillary pressure

Let us consider the case of a small parameter  $\varkappa$ . From the problem (III.10) it follows that the smallness of  $\varkappa$  is the same as the large-time asymptotics of the process at  $\varkappa \sim 1$ . So, the asymptotics  $\varkappa \rightarrow 0$  and  $t \rightarrow \infty$  are expected to lead to the same results.

According to the definition of  $\varkappa$  (II.11), it means that the ratio between the block and fracture permeability is

$$\frac{K^I}{K^I} \sim \frac{\varepsilon^2}{\varkappa} \gg \varepsilon^2.$$

Therefore, we deal with a medium with a lower heterogeneity than the classic  $\varepsilon^2$  double-porosity medium. In particular, this might be the case of  $K^I/K^I \sim \varepsilon$ . Let the parameter  $\delta$  be of order unity, which means that the heterogeneity with respect to capillary effects is low.

The fact that the degree of medium heterogeneity is not very high means that the flow rate in the blocks will be not much lower than that in the fractures. Consequently, the delay in block behavior is not expected to be very high. Thus, we deal with the case of low nonequilibrium.

All functions in Eqs. (IV.1) and (IV.2) may be expanded in asymptotic series over  $\varkappa$ :

$$\begin{aligned} S^I(x, y, t) &= S_0^I(x, t) + \varkappa S_1^I(x, y, t) + \varkappa^2 \dots, \\ S^I(x, y, t) &= S_0^I(x, t) + \varkappa S_1^I(x, t) + \varkappa^2 \dots, \\ \zeta(y, x, t) &= \zeta_0(y, x, t) + \varkappa \dots. \end{aligned} \quad (\text{V.1})$$

The fact that the zero-order term  $S_0^I(x, t)$  does not depend on the fast variable  $y$  is justified by the weak nonequilibrium and, consequently, by the sufficiently fast stabilization of the saturation field in blocks. Moreover, the fact that the first-order term  $S_1^I(x, t)$  does not depend on  $y$  is due to the fact that the fluctuations over the mean values in fractures are very low. In Ref. [33] such a structure of the asymptotic expansions was proven analytically.

Then relationship (IV.1) gives the following expansion:

$$\begin{aligned} & \frac{1}{\delta} \langle P_c^I(S_0^I(x,t) + \varkappa S_1^I(x,y,t)) \rangle_I \\ &= P_c^{\text{II}}(S_0^{\text{II}} + \varkappa S_1^{\text{II}}(x,y,t)) - \varkappa \langle \phi^I \zeta_0 \rangle_I \frac{\partial S_0^I(x,t)}{\partial t} + O(\varkappa^2). \end{aligned} \quad (\text{V.2})$$

Let us introduce the average saturations:

$$\begin{aligned} \widehat{S}^I &= S_0^I(x,t) + \varkappa \langle S_1^I(x,y,t) \rangle_I, \\ \widehat{S}^{\text{II}} &= S_0^{\text{II}}(x,t) + \varkappa S_1^{\text{II}}(x,t). \end{aligned}$$

Next, for the first term in Eq. (V.2), one obtains

$$\begin{aligned} & \langle P_c^I(S_0^I(x,t) + \varkappa S_1^I(x,y,t)) \rangle_I \\ &= \langle P_c^I(\widehat{S}^I(x,t) + \varkappa (S_1^I(x,y,t) - \langle S_1^I(x,y,t) \rangle_I)) \rangle_I \\ &= P_c^I(\widehat{S}^I(x,t)) + \varkappa (\langle S_1^I(x,y,t) \rangle_I - \langle S_1^I(x,y,t) \rangle_I) \\ &= P_c^I(\widehat{S}^I(x,t)). \end{aligned}$$

Then (V.2) yields

$$\frac{1}{\delta} P_c^I(\widehat{S}^I) - P_c^{\text{II}}(\widehat{S}^{\text{II}}) = -\varkappa \langle \phi^I \zeta_0 \rangle_I \frac{\partial \widehat{S}^I}{\partial t} + O(\varkappa^2). \quad (\text{V.3})$$

The function  $\zeta_0$  is obtained from the problem (IV.3):

$$\zeta_0 = -\frac{\lambda^I(S_0^I)}{\lambda_w^I(S_0^I)\lambda_o^I(S_0^I)} \varphi(y),$$

where  $\varphi(y)$  is the solution of the following problem:

$$\begin{aligned} \nabla_y(K^I \nabla_y \varphi) &= -1 \quad \text{in } Y^I, \\ \varphi &= 0 \quad \text{on } \partial Y^I. \end{aligned} \quad (\text{V.4})$$

Definitely, relationship (V.3) becomes

$$\frac{1}{\delta} P_c^I(\widehat{S}^I) - P_c^{\text{II}}(\widehat{S}^{\text{II}}) = \tau_\star(\widehat{S}^I) \frac{\partial \widehat{S}^I}{\partial t}, \quad (\text{V.5})$$

with

$$\tau_\star \stackrel{\text{def}}{=} \varkappa \frac{\langle \phi^I \varphi \rangle_I \lambda^I(\widehat{S}^I)}{\lambda_w^I(\widehat{S}^I)\lambda_o^I(\widehat{S}^I)}. \quad (\text{V.6})$$

Expression (V.6) is exactly the nonequilibrium relationship (38c) of Bourgeat and Panfilov obtained in Ref. [33].

In the mentioned paper relationship (V.6) was obtained even for  $\delta \ll 1$ , i.e. for higher degree of heterogeneity with respect to capillary effects, when the structure of asymptotic expansions becomes more complicated. The necessary condition of validity of relationship (V.6) is

$$\varkappa \ll \delta \ll 1 \quad (\text{V.7})$$

In a similar way the same relationship may be obtained from (IV.2).

Thus, the weak nonequilibrium, which corresponds to sufficiently weak nonstationary effects within the blocks, leads to appearance of a short-term memory in the averaged system (with small relaxation time  $\tau_\star$ ).

## B. Homogenized model

According to Ref. [33], the homogenized model (III.1) in the considered case (V.7) becomes

$$\begin{aligned} \widehat{\phi} \frac{\partial \widehat{S}^{\text{II}}}{\partial t} - \text{div}(\mathbb{K} \lambda_w^{\text{II}}(\widehat{S}^{\text{II}}) (\nabla P_w^{\text{II}} - \gamma_w \vec{e}_g)) &= -\phi^I \frac{\partial \widehat{S}^I}{\partial t}(x,t), \\ -\widehat{\phi} \frac{\partial \widehat{S}^{\text{II}}}{\partial t} - \text{div}(\mathbb{K} \lambda_o^{\text{II}}(\widehat{S}^{\text{II}}) (\nabla P_o^{\text{II}} - \gamma_o \vec{e}_g)) &= \phi^I \frac{\partial \widehat{S}^I}{\partial t}(x,t), \end{aligned} \quad (\text{V.8})$$

$$P_c^{\text{II}}(\widehat{S}^{\text{II}}) = P_o^{\text{II}} - P_w^{\text{II}}.$$

This system, being completed with the nonequilibrium relationship (V.6), represents the closed completely homogenized model with respect to the functions  $\widehat{S}^{\text{II}}$ ,  $\widehat{S}^I$ ,  $P_o^{\text{II}}$ , and  $P_w^{\text{II}}$ .

## VI. UPPER AND LOWER BOUNDS FOR NONLINEAR MODEL: COMPLETELY HOMOGENIZED CASE 2

For the problem in the block, it is possible to obtain the useful result which is usually called the theorem of comparison. It establishes the upper and lower bounds for the macroscopic nonlinear model. The advantage of these bounds is that they represent linear models. Due to this, they may be represented in completely homogenized form.

### A. Theorem of comparison

For the problem (III.10), let us introduce the upper and the lower bounds for the nonlinear coefficient  $D^{I,\delta}(S^I)$ :

$$D_M \stackrel{\text{def}}{=} \max_{s \in [0,1]} D^{I,\delta}(s), \quad D_m \stackrel{\text{def}}{=} \min_{s \in [0,1]} D^{I,\delta}(s), \quad (\text{VI.1})$$

and two linear bounds for the nonlinear function  $s^I(S^{\text{II}})$  which may be defined in various ways; for instance, as

$$\begin{aligned} s_j(S^{\text{II}}) &= \alpha_j(S^{\text{II}} - s_\star), \quad j = m, M \\ \alpha_j &= \begin{cases} \frac{dP_c^{\text{II}}}{dS^{\text{II}}} \left( \frac{dP_c^{I,\delta}}{dS^I} \right)^{-1} \Big|_{S^{\text{II}}=s_\star} & \text{for } j = M \\ \frac{1}{1-s_\star}, & \text{for } j = m, \end{cases} \end{aligned} \quad (\text{VI.2})$$

where  $s_\star$  is the percolation threshold mentioned in property A.2 in Sec. II D. Their plot is shown in Fig. 4.

Notice that, due to (II.16) and the definition A.3 of the capillary diffusion, it holds that  $D_m, D_M > 0$ .

Therefore, for any  $x, y, t$  one has

$$\begin{aligned} lD_m &\leq D^{I,\delta}(S^I) \leq D_M, \\ s_m(S^{\text{II}}) &\leq s^I(S^{\text{II}}) \leq s_M(S^{\text{II}}). \end{aligned} \quad (\text{VI.3})$$

Using these four bounds we can formulate four linear cell problems in a block for four new functions  $S_{i,j}^I(x, y, t)$ ,  $i, j = M, m$ :

$$\begin{aligned} \phi^I \frac{\partial S_{i,j}^I}{\partial t} - \nabla_y(K^{\varkappa,1} D_i \nabla_y S_{i,j}^I) &= 0 \quad \text{in } \Omega_T \times Y^I, \\ S_{i,j}^I &= s_j(S^{\text{II}}) \quad \text{in } \Omega_T \times \Gamma, \\ S_{i,j}^I(x, y, 0) &= S_0^I(x) \quad \text{in } \Omega \times Y^I. \end{aligned} \quad (\text{VI.4})$$

Finally, we can formulate the comparison theorem.

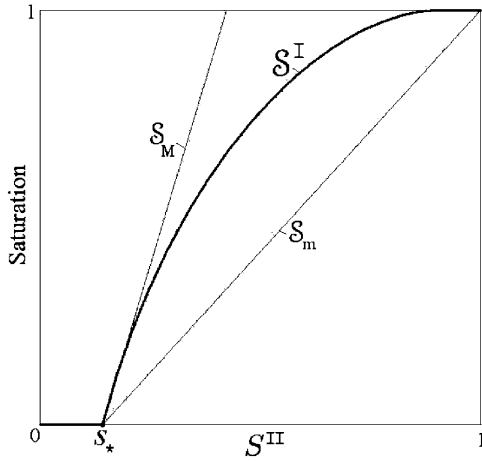


FIG. 4. Equilibrium saturation in blocks  $s^I$  and its upper and lower linear bounds as functions of saturation in fractures  $S^{II}$ .

*Theorem VI.1.* Let  $S^I$  and  $S_{i,j}^I$  be the solutions of the cell problems (III.10), and (VI.4), respectively. Then the following inequalities hold true in  $\Omega_T \times Y^I$ :

$$\begin{aligned} \text{if } S_0^I \leq s^I \text{ then } S_{m,m}^I &\leq S^I \leq S_{M,M}^I, \\ \text{if } S_0^I \geq s^I \text{ then } S_{M,m}^I &\leq S^I \leq S_{m,m}^I. \end{aligned} \quad (\text{VI.5})$$

Thus, in any case, the solution of the nonlinear problem (III.10) is located between the solutions of four linear problems (VI.4).

The proof of theorem VI.1, being too technical, is moved out of the main text to the Appendix.

### B. Physical meaning of theorem of comparison

Figure 5 illustrates the typical behavior of two functions,  $S_{M,M}^I(y,t)$  and  $S^I(y,t)$ , over a square block, as the numerical solution of two cell problems, (VI.4) and (III.10).

The initial and boundary data were:  $S_0^I = 0$ ,  $s^I = s_M = 0.3$  so that, according to the principle of maximum,  $S_0^I \leq S^I \leq s^I$ .

These functions are calculated numerically as the numerical solution of the problems (III.10) and (VI.4) for the following values of parameters:  $D^{1,\delta}(s) = s(1-s) + 0.1$ ,  $D_M = 0.25 + 0.1 = \max\{D^{1,\delta}(s)\}$ . The domain is represented by a unit square. The lower surface (b) corresponds to the solution of the nonlinear problem (III.10), while the upper surface (a) is

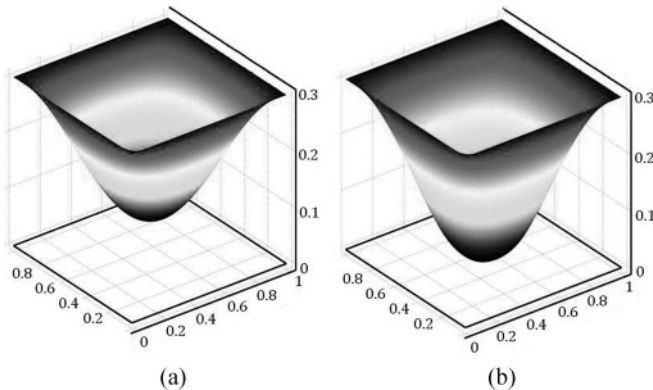


FIG. 5. Illustration to the theorem of comparison: functions (a)  $S_{M,M}^I(y,t)$  and (b)  $S^I(y,t)$  for a fixed time  $t$ . The case  $S_0^I \leq s^I$ .

the solution of the linear problem (VI.4) with maximal value of the diffusion coefficient, which corresponds to the theorem of comparison.

Such behavior explains the meaning of the theorem. Indeed, the process in a cell represents a monotonic tendency of the initially perturbed saturation field which has the shape of a mountain to become plane and equivalent to its uniform boundary value. The saturation function, which has the higher rate of dissipation, will be flatter. The rate of such a dissipation is proportional to the diffusion parameter. So, for the maximum diffusion parameter, the saturation surface will be closer to a plane. Due to this the surface  $S_{M,M}^I(y,t)$  will be always situated above  $S^I(y,t)$  (i.e.,  $S_{M,M}^I \geq S^I$ ). This result is valid even in the case  $s^I = s_M$ , which is shown in Fig. 5. Obviously it will be more assured if we move the whole left-hand figure up (i.e., apply the strict inequality  $s^I < s_M$ ).

In a similar way, for the case  $S_0^I \geq s^I$ , the function  $S_{M,m}^I$ , which corresponds to the maximal diffusion and has the maximal rate of dissipation, will be situated below the surface  $S^I$  (i.e.,  $S_{M,m}^I \leq S^I$ ).

### C. Completely homogenized model for linear bounds

Let us consider the homogenized equations (III.1) in which we will replace function  $S^I$  defined through the nonlinear problem (III.10) by  $S_{i,j}^I$  defined through the linear problem (VI.4). Then we obtain four macroscopic models for various values of  $i$  and  $j$ . Each of them may be reduced to the completely homogenized model, for  $i,j = M,m$ :

$$\begin{aligned} \widehat{\phi}^I \frac{\partial S_{i,j}^{II}}{\partial t} - \text{div}(\mathbb{K} \lambda_w^I(S_{i,j}^{II})(\nabla P_{w;i,j}^{II} - \gamma_w \vec{e}_g)) \\ = \widehat{\phi}^I \alpha_j \int_0^t \frac{\partial \langle \mathbf{G}_i \rangle_1(t-\tau)}{\partial t} \frac{\partial S_{q,i}^{II}}{\partial \tau} d\tau, \\ - \widehat{\phi}^I \frac{\partial S_{i,j}^{II}}{\partial t} - \text{div}(\mathbb{K} \lambda_o^I(S_{i,j}^{II})(\nabla P_{o;i,j}^{II} - \gamma_o \vec{e}_g)) \\ = -\widehat{\phi}^I \alpha_j \int_0^t \frac{\partial \langle \mathbf{G}_i \rangle_1(t-\tau)}{\partial t} \frac{\partial S_{q,i}^{II}}{\partial \tau} d\tau, \\ P_c^I(S_{i,j}^{II}) = P_{o;i,j}^{II} - P_{w;i,j}^{II}. \end{aligned} \quad (\text{VI.6})$$

Herein the average porosity  $\widehat{\phi}$  and the effective permeability tensor  $\mathbb{K}$  are given by the same equations (III.5) and (III.6). The kernel of the integrodifferential operator,  $\mathbf{G}_i$ , is the Green's function defined for  $i = M,m$  as

$$\begin{aligned} \omega \frac{\partial \mathbf{G}_i}{\partial t} - D_i \Delta_{yy} \mathbf{G}_i &= -\delta(x-y)\delta(t-\tau) \quad \text{in } \Omega_T \times Y^I, \\ \mathbf{G}_i(x,y,t) &= 0 \quad \text{in } \Omega_T \times \Gamma; \\ \mathbf{G}_i(x,y,0) &= 0 \quad \text{in } \Omega \times Y^I, \end{aligned} \quad (\text{VI.7})$$

where  $\omega \stackrel{\text{def}}{=} \phi^I / K^{\infty,I}$ .

The presented system of integrodifferential equations is closed with respect to the three functions  $P_{w;i,j}^{II}(x,t)$ ,  $P_{o;i,j}^{II}(x,t)$ , and  $S_{i,j}^{II}(x,t)$ . This model is completely homogenized, because system (VI.6) does not contain fast variables any longer, while the cell problem (VI.7) is independent of slow variables.

According to the theorem of comparison, we can expect that the solution of the nonlinear model (III.1) and (III.10)

is bounded by a combination constructed from four linear models (VI.6):

$$S_{\min}^{\parallel}(x,t) \leq S^{\parallel}(x,t) \leq S_{\max}^{\parallel}(x,t), \quad (\text{VI.8})$$

where

$$S_{\min}^{\parallel} \stackrel{\text{def}}{=} \min_{i,j=m,M} \{S_{i,j}^{\parallel}\}, \quad S_{\max}^{\parallel} \stackrel{\text{def}}{=} \max_{i,j=m,M} \{S_{i,j}^{\parallel}\}. \quad (\text{VI.9})$$

The nonequilibrium relationship between two average capillary pressures, (IV.1) or (IV.2), becomes useless for the linear bounds.

For completeness, we show how one can obtain the completely homogenized model (VI.6). The linear problem (VI.4) can be solved analytically by means of a Green's function. By introducing a new function:  $U = S_{i,j}^I - s_j$ , we easily obtain a similar differential equation for  $U$  as in Eq. (III.10), but with the right-hand side being  $f(x,t) \equiv -\partial_t s_j = -\alpha_j \partial_t S_{i,j}^{\parallel}$  and the following homogeneous boundary and initial conditions (it is assumed that the boundary and initial functions are compatible;  $s_j|_{t=0} = S_0^I$ ):

$$\begin{aligned} \phi^I \partial_t U &= \nabla_y (K^{\varkappa,1} D_i \nabla_y U) - \alpha_j \partial_t S_{i,j}^{\parallel}, \quad y \in Y^I, \\ U(x,y,t) &= 0, \quad y \in \Gamma, \\ U(x,y,t) &= 0, \quad t = 0. \end{aligned}$$

The solution of such a problem represents the convolution of the Green's function  $\mathbf{G}_i$  of the diffusion operator with the function  $f$  [i.e.,  $U = \int_0^t \mathbf{G}_i(t-\tau) f(\tau) d\tau$ , where  $\mathbf{G}_i$  is defined as the solution of (VI.7)]. Then we obtain

$$\begin{aligned} S_{i,j}^I(x,y,t) &= \alpha_j [S_{i,j}^{\parallel}(x,t) - s_*] \\ &\quad - \alpha_j \int_0^t \mathbf{G}_i(x,y,t-\tau) \frac{\partial S_{i,j}^{\parallel}}{\partial \tau}(x,\tau) d\tau. \end{aligned} \quad (\text{VI.10})$$

Substituting this explicit relationship in equations (III.1) instead of  $S^I$  we obtain (VI.6).

## VII. NUMERICAL SIMULATIONS

### A. Data description

The theorem of convergence determines strictly the domain of the validity of the result obtained. However, several restrictions on the parameters have no clear physical meaning and are sufficiently difficult to control in practice. Another way to check the validity is to calculate numerically the microscopic problem and all homogenized parameters. In the present paper we will analyze only the validity of the general nonequilibrium relationship for the capillary pressure, (IV.1) and compare it with the simpler asymptotic equation (V.6). For this, it is sufficient to calculate the cell problem in blocks (III.10) under a given boundary function  $s^{\parallel}(x,t)$ .

The simulation was performed by applying the finite element method and an implicit approximation with respect to time. The block was a homogeneous two-dimensional square of area 0.8. The initial saturation in blocks  $S_0^I$  is assumed to be constant in space. The following closure relationships were assumed:

$$\begin{aligned} P_c^{\parallel}(s) &= \alpha \sqrt{-\ln s}, \quad P_c^{1,\delta}(s) = \frac{\alpha}{\delta} \sqrt{-\ln s}, \\ \lambda_w^I(s) &= s^2, \quad \lambda_o^I(s) = (1-s)^2. \end{aligned}$$

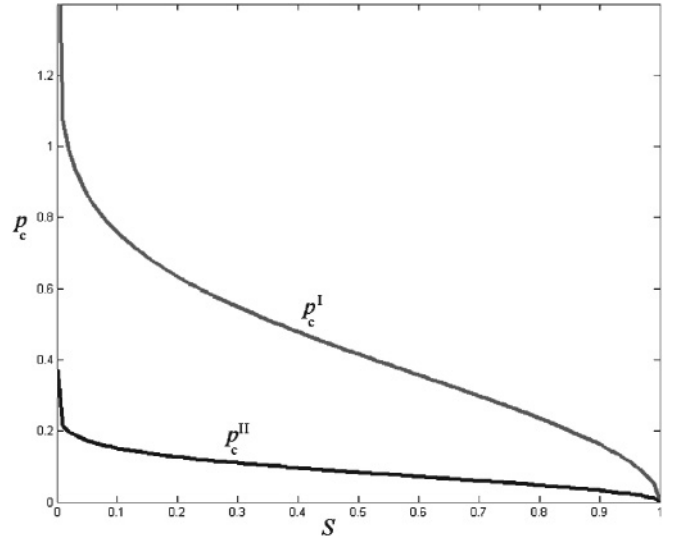


FIG. 6. Microscopic capillary pressure-saturation functions for blocks and fractures.

The nonequilibrium is introduced into the system through the boundary value  $S^{\parallel}(x,t)$ , if it is selected in such a way that

$$P_c^{\parallel}(S^{\parallel}) \neq P_c^{1,\delta}(S_0^I). \quad (\text{VII.1})$$

The shape of the capillary pressure-saturation functions is shown in Fig. 6 for  $\delta = 0.2$ . Several parameters were fixed:  $\delta = 0.8$ ,  $\alpha = 0.1$ , and  $K^I/\phi^I = 2$ . The selected value of  $\delta$  corresponds to the moderate difference between the microscopic capillary pressure-saturation curves in the blocks and in the fractures.

We consider two main scenarios of variation of the boundary function: (i) the monotonic-in-time boundary condition, which corresponds to the permanent injection of water into the reservoir initially saturated with oil, and (ii) the nonmonotonic variation in time of the saturation  $S^{\parallel}$ , which corresponds to the alternated injection of water and oil. In engineering practice, the water injection is called *imbibition*, while the oil injection is *drainage*. Within the framework of each scenario we analyzed the case of high nonequilibrium,  $\varkappa \sim 1$ , and the case of low nonequilibrium,  $\varkappa \ll 1$ .

### B. Example 1: monotonic perturbation

The first example corresponds to the monotonic perturbation carried out through the boundary function  $S^{\parallel}$ . This is the case of a simple history. In the initial state, the medium was occupied by oil (the initial water saturation was 0.2), after which one starts to inject water in fractures at constant saturation 0.9. The imposed variation of the saturation in fractures is shown in Fig. 7 for two cases:  $\varkappa = 0.1$  and  $\varkappa = 1$ . The same figure also contains the saturation in the block which was calculated by numerically solving the problem (III.10).

The difference between the average capillary pressure in blocks and in fractures equals

$$\langle P_c^{1,\delta}(S^I) \rangle_1 - P_c^{\parallel}(S^{\parallel}). \quad (\text{VII.2})$$

It was calculated in three different ways and are all presented in Fig. 8. First of all we calculated (VII.2) by numerically solving

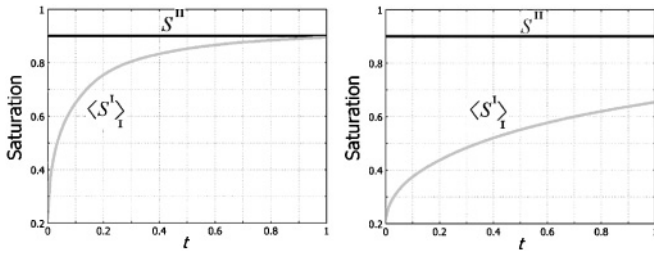


FIG. 7. Variation of saturation in fractures and blocks for (a) weak nonequilibrium,  $\varkappa = 0.1$  and (b) high nonequilibrium,  $\varkappa = 1$ .

the problem in a block (III.10) and by averaging the numerical data obtained for the function  $P_c^{1,\delta}(S^I)$ . We call these data the “exact” data. They are presented by red dashed curves.

Second, we calculated the mentioned difference by using the relationship (IV.1). For this, we numerically solved the cell problems (IV.3) and (IV.4) to obtain the functions  $\zeta(y)$  and  $\psi(y)$ . The data for the factor  $\partial_t S^I$  were obtained from the numerical solution of problem (III.10). These results are presented by the black curve in Fig. 8.

Third, we calculated the difference (VII.2) by using the asymptotic equation obtained in Ref. [33] (the blue curve).

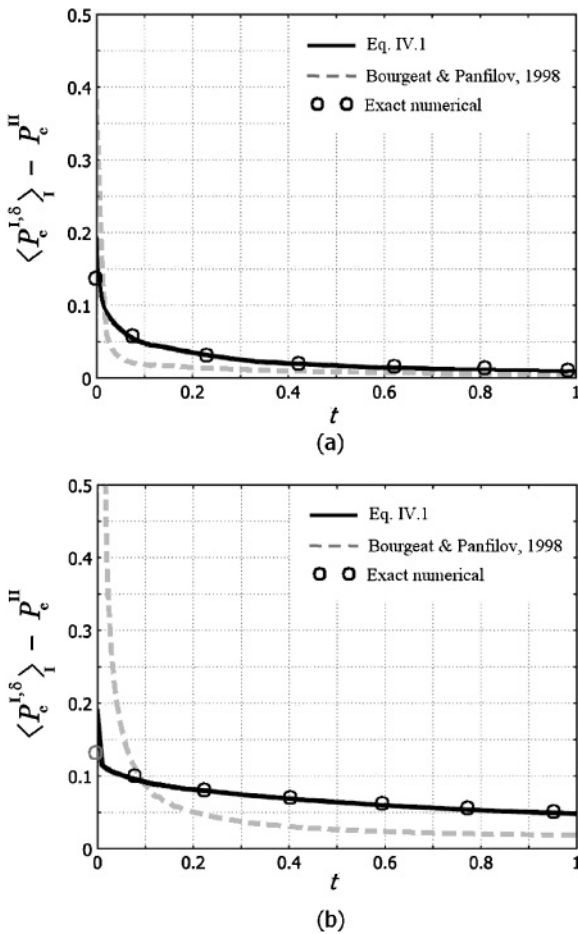


FIG. 8. Difference between average capillary pressures in fractures and blocks for (a) weak nonequilibrium,  $\varkappa = 0.1$ , and (b) high nonequilibrium,  $\varkappa = 1$  calculated in three different ways.

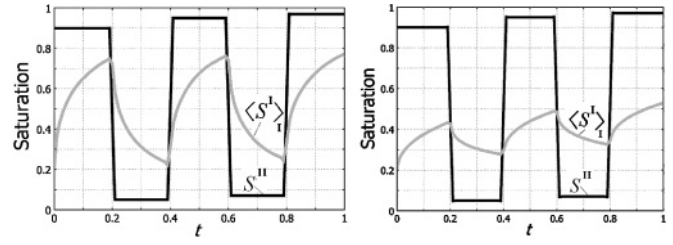


FIG. 9. Variation of saturation in fractures and blocks for (a) weak nonequilibrium,  $\varkappa = 0.1$  and (b) high nonequilibrium,  $\varkappa = 1$ .

One sees that the relationship (IV.1) obtained in the present paper gives good results in both cases. The asymptotic relationship of [33] rapidly converges to the exact solution in the case of weak nonequilibrium [Fig. 8(a)], but gives non-neglecting quantitative error in the case of high nonequilibrium [Fig. 8(b)].

### C. Example 2: nonmonotonic perturbation

In the second example we consider a complicated history, when the system was perturbed several times. At the initial state the medium was occupied essentially by oil at water saturation 0.2. At  $t = 0$  one starts to inject water in fractures at constant saturation 0.9. At  $t = 0.2, 0.4, 0.6,$  and  $0.8$

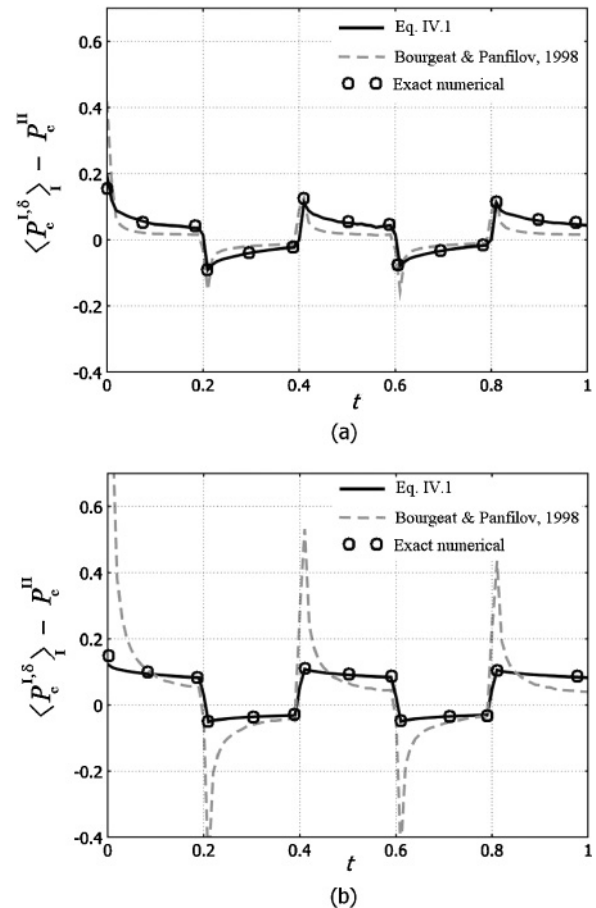


FIG. 10. Difference between average capillary pressures in fractures and blocks for (a) weak nonequilibrium,  $\varkappa = 0.1$  and (b) high nonequilibrium,  $\varkappa = 1$ , calculated in three different ways.

one sharply changes the boundary conditions by imposing the values 0.05, 0.95, 0.07, and 0.97, respectively, to  $S^{\text{II}}$ , alternating the imbibition and drainage. The variation of the saturation in fractures  $S^{\text{I}}$  is shown in Fig. 9 for two cases:  $\varkappa = 0.1$  and  $\varkappa = 1$ . The saturation in the block, which is also presented in these figures, was calculated by solving numerically the problem (III.10).

Due to repetitive perturbations, the system does not have sufficiently time to reach the equilibrium in both cases.

The difference between the averaged capillary pressure (VII.2) calculated in three different ways is presented in Fig. 10 relationship (IV.1) gives good results with respect to the exact solution in both cases, as in the previous sections. The asymptotic relationship (V.6) is satisfactory for the case of weak nonequilibrium, but causes significant quantitative error for the case of high nonequilibrium.

As mentioned in Sec. V A, the asymptotic relationship (V.6) is invalid for small times after perturbation. If the perturbation is significant and repetitive, as in the considered case, then the asymptotic relationship (V.6) becomes invalid in the vicinity of perturbation points. If one simply cuts in all zones corresponding to small times after perturbation, then the obtained solution may be considered as satisfactory for approximate calculations.

The memory effects are clearly manifested in terms of the average capillary pressure  $\bar{P}_c$  as the function of the average saturation  $\bar{S}$  which are defined as

$$\bar{S} \equiv \langle S \rangle_{\text{I}} \theta + S^{\text{II}}(1 - \theta), \quad \bar{P}_c \equiv \langle P_c^{\text{I}} \rangle_{\text{I}} \theta + P_c^{\text{II}}(1 - \theta).$$

Figure 11 illustrates the behavior of the mean capillary pressure for the analyzed example of nonmonotonic perturbation when the initial imbibition is alternated with the secondary drainage, and next with the secondary imbibition and the ternary drainage. The hysteresis behavior is the explicit proof of the appearance of the long-term memory. The mean capillary pressure is not a unique function of saturation, but depends on the overall history of the process.

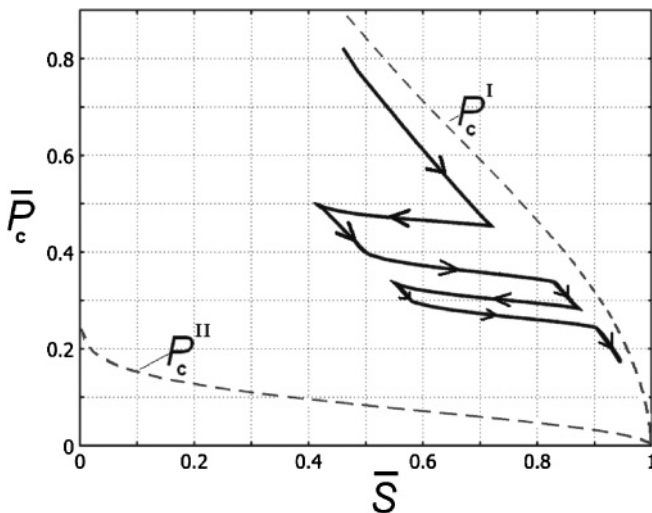


FIG. 11. Hysteresis behavior of the average capillary pressure depending on the process direction for an alternation of imbibition and drainage (black curve).

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## APPENDIX: PROOFS OF SOME TECHNICAL RESULTS

### 1. Proof of Theorem IV.1

(1) Let  $\zeta^*$  be an arbitrary function such that  $\zeta^* = 0$  on  $\Gamma$ . Then, from the first equation of system (III.2), it follows that

$$\int_{Y^{\text{I}}} \phi^{\text{I}} \frac{\partial S^{\text{I}}}{\partial t} \zeta^* dy = - \int_{Y^{\text{I}}} K^{\varkappa, \text{I}} \lambda_w^{\text{I}} \nabla_y P_w^{\text{I}} \cdot \nabla_y \zeta^* dy. \quad (\text{A1})$$

Using (IV.6) we obtain

$$\begin{aligned} \int_{Y^{\text{I}}} \phi^{\text{I}} \frac{\partial S^{\text{I}}}{\partial t} \zeta^* dy &= \int_{Y^{\text{I}}} \frac{K^{\varkappa, \text{I}} \lambda_w^{\text{I}} \lambda_o^{\text{I}}}{\lambda^{\text{I}}} \nabla_y P_c^{\text{I}, \delta} \cdot \nabla_y \zeta^* dy \\ &= \int_{Y^{\text{I}}} \frac{K^{\varkappa, \text{I}} \lambda_w^{\text{I}} \lambda_o^{\text{I}}}{\lambda^{\text{I}}} \nabla_y (P_c^{\text{I}, \delta} - P_c^{\text{II}}) \cdot \nabla_y \zeta^* dy, \end{aligned}$$

because  $P_c^{\text{II}}$  is independent of  $y$ . Integrating by part the last term we obtain

$$\begin{aligned} \int_{Y^{\text{I}}} \phi^{\text{I}} \frac{\partial S^{\text{I}}}{\partial t} \zeta^* dy &= \int_{\Gamma} \frac{K^{\varkappa, \text{I}} \lambda_w^{\text{I}} \lambda_o^{\text{I}}}{\lambda^{\text{I}}} (P_c^{\text{I}, \delta} - P_c^{\text{II}}) \frac{\partial \zeta^*}{\partial \nu} dy \\ &\quad - \int_{Y^{\text{I}}} (P_c^{\text{I}, \delta} - P_c^{\text{II}}) \nabla_y \left( \frac{K^{\varkappa, \text{I}} \lambda_w^{\text{I}} \lambda_o^{\text{I}}}{\lambda^{\text{I}}} \nabla_y \zeta^* \right) dy. \end{aligned}$$

The first term disappears due to (III.12). In the second term we assume that the function  $\zeta^* = \zeta$  which verifies the problem (IV.3). Then we obtain

$$\int_{Y^{\text{I}}} \phi^{\text{I}} \varkappa \frac{\partial S^{\text{I}}}{\partial t} \zeta dy = - \int_{Y^{\text{I}}} (P_c^{\text{I}, \delta} - P_c^{\text{II}}) dy, \quad (\text{A2})$$

which yields (IV.1).

(2) The second relationship (IV.2) is obtained in the following way: First of all, the relationship (A2) may be formulated in the equivalent form

$$\begin{aligned} \int_{Y^{\text{I}}} \phi^{\text{I}} \frac{\partial S^{\text{I}}}{\partial t} \zeta^* dy &= \int_{Y^{\text{I}}} \frac{K^{\varkappa, \text{I}} \lambda_w^{\text{I}} \lambda_o^{\text{I}}}{\lambda^{\text{I}}} \frac{dP_c^{\text{I}, \delta}}{dS^{\text{I}}} \nabla_y (S^{\text{I}} - s^{\text{I}}) \cdot \nabla_y \zeta^* dy \\ &= - \int_{Y^{\text{I}}} K^{\varkappa, \text{I}} D^{\text{I}, \delta}(S^{\text{I}}) \nabla_y (S^{\text{I}} - s^{\text{I}}) \cdot \nabla_y \zeta^* dy, \end{aligned} \quad (\text{A3})$$

because  $s^{\text{I}}$  is independent of  $y$ . Integrating by parts we obtain

$$\int_{Y^{\text{I}}} \phi^{\text{I}} \frac{\partial S^{\text{I}}}{\partial t} \zeta^* dy = \frac{1}{\delta} \int_{Y^{\text{I}}} (S^{\text{I}} - s^{\text{I}}) \nabla_y (K^{\varkappa, \text{I}} D^{\text{I}, \delta} \nabla_y \zeta^*) dy. \quad (\text{A4})$$

Assuming that  $\zeta^* = \psi$ , we have

$$\varkappa \delta \left\langle \phi^{\text{I}} \frac{\partial S^{\text{I}}}{\partial t} \psi \right\rangle_{Y^{\text{I}}} = \langle S^{\text{I}} \rangle_{Y^{\text{I}}} - s^{\text{I}}. \quad (\text{A5})$$

Then,

$$P_c^{1,\delta} \left( \langle S^1 \rangle_{Y^1} - \varkappa \delta \left\langle \phi^1 \frac{\partial S^1}{\partial t} \psi \right\rangle_{Y^1} \right) = P_c^{1,\delta} (s^1),$$

and, using (III.12), we obtain (IV.2).

## 2. Proof of Lemma on zero total flux in blocks IV.1

(1) From the two first equations of (III.2) one obtains

$$\nabla_y (K^{\varkappa,1} \lambda^1(S^1) \nabla_y P_w^1 + K^{\varkappa,1} \lambda_o^1(S^1) \nabla_y P_c^{1,\delta}) = 0. \quad (\text{A6})$$

(2) Let  $\psi(x, y, t)$  be so that  $\psi = 0$  on  $\Gamma$ . Then the last relationship multiplied by  $\psi$  and integrated over  $Y^1$  becomes

$$\begin{aligned} 0 &= \int_{Y^1} K^{\varkappa,1} \lambda^1 \left( \nabla_y P_w^1 + \frac{\lambda_o^1}{\lambda^1} \nabla_y P_c^{1,\delta} \right) \nabla_y \psi dy \\ &= \int_{Y^1} K^{\varkappa,1} \lambda^1 \nabla_y P^1 \cdot \nabla_y \psi dy, \end{aligned} \quad (\text{A7})$$

where

$$P^1 \stackrel{\text{def}}{=} P_w^1 - \int_s^1 \frac{\lambda_o^1(\xi)}{\lambda^1(\xi)} \frac{dP_c^{1,\delta}}{d\xi} d\xi$$

is the global pressure [60]. Let us define the new function  $\psi$  as  $\psi = P^1 - P_\Gamma^1$ , where  $P_\Gamma^1 \stackrel{\text{def}}{=} P^1|_{y \in \Gamma}$ . According to (III.12), the boundary saturation  $s^1$  does not depend on  $y$ ; thus, the value  $P_\Gamma^1$  also is independent of  $y$ . Then we obtain from (A7) the following:

$$0 = \int_{Y^1} K^{\varkappa,1} \lambda^1 \nabla_y P^1 \cdot (\nabla_y P^1 - P_\Gamma^1) dy = \int_{Y^1} K^{\varkappa,1} \lambda^1 |\nabla_y P^1|^2 dy. \quad (\text{A8})$$

Using property (II.15), we obtain

$$0 = \int_{Y^1} K^{\varkappa,1} \lambda^1 |\nabla_y P^1|^2 dy \geq K_{\min}^{\varkappa,1} L_0 \int_{Y^1} |\nabla_y P^1|^2 dy, \quad (\text{A9})$$

which means that  $\nabla_y P^1 \equiv 0$ . This proves relationship (IV.6).

## 3. Proof of Theorem of comparison VI.1

We restrict ourselves to the proof of the right-hand side of the first inequality in Eq. (VI.5). Other inequalities can be proven by using the same arguments.

First, let us formulate the cell problems (III.10) and (VI.4) in the equivalent form:

$$\begin{aligned} \phi^1 \frac{\partial S^1}{\partial t} - K^{\varkappa,1} \Delta_{yy} \beta^{1,\delta}(S^1) &= 0 \quad \text{in } \Omega_T \times Y^1, \\ S^1 &= S^1(S^\mathbb{I}) \quad \text{in } \Omega_T \times \Gamma, \\ S^1(x, y, 0) &= S_0^1(x) \quad \text{in } \Omega \times Y^1. \end{aligned} \quad (\text{A10})$$

$$\begin{aligned} \phi^1 \frac{\partial S_{i,j}^1}{\partial t} - K^{\varkappa,1} \Delta_{yy} \beta_i(S_{i,j}^1) &= 0 \quad \text{in } \Omega_T \times Y^1, \\ S_{i,j}^1 &= s_j(S^\mathbb{I}) \quad \text{in } \Omega_T \times \Gamma, \\ S_{i,j}^1(x, y, 0) &= S_0^1(x) \quad \text{in } \Omega \times Y^1. \end{aligned} \quad (\text{A11})$$

where

$$\beta^{1,\delta}(s) \stackrel{\text{def}}{=} \int_0^s D^{1,\delta} d\xi = - \int_0^s \frac{\lambda_w^1 \lambda_o^1}{\lambda^1} \frac{dP_c^{1,\delta}}{d\xi} d\xi, \quad (\text{A12})$$

$$\beta_M(s) \stackrel{\text{def}}{=} \int_0^s D_M d\xi, \quad \beta_m(s) \stackrel{\text{def}}{=} \int_0^s D_m d\xi. \quad (\text{A13})$$

(1) Consider the case  $S_0^1 \leq s^1$ . From the principle of maximum and minimum for parabolic equation, it results that the solution of (A10) and (A11) may be only located between the boundary and the initial values

$$S_0^1 \leq S^1 \leq s^1. \quad (\text{A14})$$

(2) Consider the weak formulation of the cell problem (A10). It reads

$$\begin{aligned} \int_{\Omega_t \times Y^1} \phi^1 \frac{\partial S^1}{\partial \tau} \varphi dx dy d\tau \\ + \int_{\Omega_t \times Y^1} K^{\varkappa,1} \nabla_y \beta^{1,\delta}(S^1) \cdot \nabla_y \varphi dx dy d\tau = 0, \end{aligned} \quad (\text{A15})$$

where  $\Omega_t \stackrel{\text{def}}{=} \Omega \times (0, t)$ . The corresponding weak formulation of the cell problem (A11) for  $i, j = M$  reads

$$\begin{aligned} 0 &= \int_{\Omega_t \times Y^1} \phi^1 \frac{\partial S_{M,M}^1}{\partial \tau} \varphi dx dy d\tau \\ &+ \int_{\Omega_t \times Y^1} K^{\varkappa,1} \nabla_y \beta_M(S_{M,M}^1) \cdot \nabla_y \varphi dx dy d\tau. \end{aligned} \quad (\text{A16})$$

(3) Taking the difference of equations (A16) and (A15) we obtain

$$\begin{aligned} 0 &= \int_{\Omega_t \times Y^1} \phi^1 \frac{\partial}{\partial \tau} (S_{M,M}^1 - S^1) \varphi dx dy d\tau \\ &+ \int_{\Omega_t \times Y^1} \nabla_y (\beta_M(S_{M,M}^1) - \beta^{1,\delta}(S^1)) \cdot K^{\varkappa,1} \nabla_y \varphi dx dy d\tau. \end{aligned} \quad (\text{A17})$$

Since  $s^1$  and  $s_M$  do not depend on  $y$ , then we rewrite equation (A17) as follows:

$$\begin{aligned} 0 &= \int_{\Omega_t \times Y^1} \phi^1 \frac{\partial}{\partial \tau} (S_{M,M}^1 - S^1) \varphi dx dy d\tau \\ &+ \int_{\Omega_t \times Y^1} \nabla_y \Phi \cdot K^{\varkappa,1} \nabla_y \varphi dx dy d\tau, \end{aligned} \quad (\text{A18})$$

with

$$\begin{aligned} \Phi &\stackrel{\text{def}}{=} \beta_M(S_{M,M}^1) - \beta_M(s_M(S^\mathbb{I})) \\ &- [\beta^{1,\delta}(S^1) - \beta^{1,\delta}(s^1(S^\mathbb{I}))] \\ &= \int_{s_M}^{S_{M,M}^1} D_M d\xi - \int_{s^1}^{S^1} D^{1,\delta} d\xi \\ &= \int_{s_M}^{S_{M,M}^1} (D_M - D^{1,\delta}) d\xi + \int_{s_M}^{S^1} D^{1,\delta} d\xi + \int_{s^1}^{S_{M,M}^1} D^{1,\delta} d\xi. \end{aligned} \quad (\text{A19})$$



Integrating by parts in the second term, we get:

$$0 = \int_{\Omega_t \times Y^1} \phi^I \frac{\partial}{\partial \tau} (S_{M,M}^I - S^I) \phi dx dy d\tau - \int_{\Omega_t \times Y^1} \Phi \nabla_y (K^{z,I} \nabla_y \phi) dx dy d\tau. \quad (A20)$$

(4) Assume now that

$$S_{M,M}^I(x, y, t) \leq S^I(x, y, t), \quad (A21)$$

then, according to (A14) and (VI.3) we reveal that all three terms in the right-hand side of (A19) are nonpositive. Thus:

$$\Phi \leq 0. \quad (A22)$$

(5) Let us introduce the function  $\varphi^*$ , solving the following problem for all  $(y, t) \in \Omega_t$ :

$$\begin{aligned} \nabla_y (K^{z,I} \nabla_y \varphi^*) &= S^* \quad \text{in } Y^1, \\ \varphi^* &= 0 \quad \text{on } \Gamma, \end{aligned} \quad (A23)$$

with

$$S^* \stackrel{\text{def}}{=} \begin{cases} S_{M,M}^I - S^I & \text{when } S_{M,M}^I < S^I \\ 0 & \text{when } S_{M,M}^I \geq S^I. \end{cases} \quad (A24)$$

Notice that

$$\varphi^* \equiv 0 \quad \text{when } S_{M,M}^I \geq S^I. \quad (A25)$$

In equation (A18) we set  $\varphi = \varphi^*$ . Then (A20) becomes

$$\int_{\Omega_t \times Y^1} \phi^I \frac{\partial}{\partial \tau} (S_{M,M}^I - S^I) \varphi^* dx dy d\tau = \int_{\Omega_t \times Y^1} \Phi S^* dx dy d\tau. \quad (A26)$$

Taking into account (A25) and (A22), we get the following inequality:

$$\int_{\Omega_t \times Y^1} \phi^I \frac{\partial}{\partial \tau} (S_{M,M}^I - S^I) \varphi^* dx dy d\tau \geq 0 \quad \text{for all } S^*. \quad (A27)$$

(6) Let us develop inequality (A27) by using (A23):

$$\begin{aligned} 0 &\leq \int_{\Omega_t \times Y^1} \phi^I \frac{\partial S^*}{\partial \tau} \varphi^* dx dy d\tau \\ &= \int_{\Omega_t \times Y^1} \phi^I \frac{\partial}{\partial \tau} [\nabla_y (K^{z,I} \nabla_y \varphi^*)] \varphi^* dx dy d\tau \\ &= - \int_{\Omega_t \times Y^1} \phi^I K^{z,I} \frac{\partial}{\partial \tau} (\nabla_y \varphi^*) \cdot \nabla_y \varphi^* dx dy d\tau \\ &= - \int_{\Omega_t \times Y^1} \phi^I K^{z,I} \frac{\partial}{\partial \tau} \left( \frac{1}{2} |\nabla_y \varphi^*|^2 \right) dx dy d\tau \\ &= - \frac{1}{2} \int_{\Omega \times Y^1} \phi^I K^{z,I} |\nabla_y \varphi^*(t)|^2 dx dy \\ &\quad + \frac{1}{2} \int_{\Omega \times Y^1} \phi^I K^{z,I} |\nabla_y \varphi^*(0)|^2 dx dy. \end{aligned}$$

Thus, we get the following inequality:

$$\int_{\Omega \times Y^1} \phi^I K^{z,I} |\nabla_y \varphi^*(t)|^2 dx dy \leq \int_{\Omega \times Y^1} \phi^I K^{z,I} |\nabla_y \varphi^*(0)|^2 dx dy. \quad (A28)$$

Since it follows from (A23) that

$$\varphi^*(0) \equiv 0, \quad (A29)$$

then from (A28) we get finally

$$\int_{\Omega \times Y^1} \phi^I K^{z,I} |\nabla_y \varphi^*(t)|^2 dx dy \leq 0. \quad (A30)$$

Thus, taking into account (A23) and (A30), we have that, for any  $t \in [0, T]$ ,

$$\varphi^* \equiv 0 \quad \text{in } \Omega_T \times Y^1. \quad (A31)$$

This means that

$$S_{M,M}^I \geq S^I \quad \text{in } \Omega_T \times Y^1, \quad (A32)$$

which contradicts the original assumption (A21). Thus, this assumption is invalid, and the true result is

$$S_{M,M}^I \geq S^I. \quad (A33)$$

The left-hand side of the first inequality (VI.5) is proven. The right-hand side may be proven in the same way. Theorem VI. 1 is proven.

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