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Origin and Quantification of Coupling Between Relative Permeabilities for Two-Phase Flows in Porous Media

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Abstract. An extended formulation of Darcy's two-phase law is developed on the basis of Stokes' equations. It leads, through results borrowed from the thermodynamics of irreversible processes, to a matrix of relative permeabilities. Nondiagonal coefficients of this matrix are due to the viscous coupling exerted between fluid phases, while diagonal coefficients represent the contribution of both fluid phases to the total flow, as if they were alone. The coefficients of this matrix, contrary to standard relative permeabilities, do not depend on the boundary conditions imposed on two-phase flow in porous media, such as the flow rate.

This formalism is validated by comparison with experimental results from tests of two-phase flow in a square cross-section capillary tube and in porous media. Coupling terms of the matrix are found to be nonnegligible compared to diagonal terms. Relationships between standard relative permeabilities and matrix coefficients are studied and lead to an experimental way to determine the new terms for two-phase flow in porous media.

Key words. Coupling, relative permeabilities, porous media flows, two-phase flow, Darcy's law.

0. Nomenclature

Roman letters

(B_i^j)	inverse of the (C_i^j) matrix
B^*	flow constant
$C(z)$	fluid/fluid interface curvature
C_i	curvature of the meniscus Σ_i; $C_i = C(Z_i)$; $i = 1, 2$
(C_i^j)	matrix of flows
$\det(B_i^j)$	determinant of the (B_i^j) matrix
k	intrinsic permeability of the medium
k_i	mobility of phase i, $i = 1, 2$
k_{ri}	relative permeability of phase i, $i = 1, 2$
$k_{r_i}^j$	coefficient of relative permeability matrix $i, j = 1, 2$
k_i^j	coefficient of mobility matrix $i, j = 1, 2$
$\mathcal{L}$	fluid/fluid interface in a cross-section
P_i	pressure exerted by the system upon phase i, $i = 1, 2$
P_c	capillary pressure ($P_c = P_1 - P_2$)
q_i	flow rate of phase i, $i = 1, 2$

Q	absolute value of q_i
$r(z)$	half-side of a cross-section at position z
s_i	saturation of phase i, $i = 1, 2$
S_i	capillary cross-section at position Z_i, $i = 1, 2$
$\mathcal{S}$	solid surface
T	temperature of the system
$\vec{u}_i$	velocity of phase i, $i = 1, 2$
Z_i	position of meniscus Σ_i, $i = 1, 2$

Greek letters

α	function of $\bar{\mu}$ and θ
β	shape factor
γ	interfacial tension of fluid/fluid interface
λ_i	flow constant $i = 1, 2$
μ_i	viscosity of fluid phase i, $i = 1, 2$
$\bar{\mu}$	visocity ratio: $\bar{\mu} = \mu_1/\mu_2$
ρ_i	mass per unit volume of fluid phase i, $i = 1, 2$
θ	contact angle
Σ_i	meniscus i of oil ganglion $i = 1, 2$
ϕ	porosity

Subscripts

1	nonwetting fluid
2	wetting fluid

1. Introduction

The standard description of two-phase flow in porous media is based on the generalization of Darcy's law. This law is rigorous for the flow of one fluid phase in a porous medium. It has been generalized (Wyckoff and Botset, 1936) by considering a permeability reduction factor for each fluid phase, called relative permeability. This term has been introduced on the basis that the porous medium, associated with one fluid phase, is seen as constituting a new porous medium for the other fluid phase. This approach assumes that fluid/solid and fluid/fluid interfaces should act in the same way, which is obviously not true. The viscous coupling effect between fluid phases has to be taken into account.

This idea is not new. Rose (1972, 1974, 1988) promoted it. De Gennes (1983) and, more recently, Auriault and Sanchez-Palencia (1986), Auriault (1987) by use of a method of spatial homogenization, and Whitaker (1986) by use of a method of volume averaging, have established coupled Darcy's laws. This same result has been underlined by the author too (Kalaydjian, 1987) by use of the thermodynamics of irreversible processes. A quantitative estimation of coupling terms for two-phase flow in square cross-section capillary tubes (Kalaydjian and

Legait, 1987b) has shown that they are nonnegligible in front of diagonal terms. This paper describes an experimental approach to achieve this purpose on both microscopic and macroscopic levels. Coupling terms are thus quantified and shown to be nonnegligible, even on a macroscopic scale.

Capillary displacements are situations for which coupling drag between fluid phases is predominant. This is the case, for instance, and within the framework of oil-recovery processes, for displacement of oil by water in countercurrent flow during spontaneous imbibition (for water-wet porous media), just as it is for the remobilization of oil ganglia by water.

An analysis of possible relationships between the standard and the extended formulation of Darcy's laws gives an experimental way to determine all the coefficients of the matrix of relative permeabilities by using only two experiments performed in porous media, with capillarity being negligible or preponderant.

2. Study at the Pore Level

2.1. THEORY

A porous medium, when considered at the pore level, is constituted by a complex succession of pores and throats (Figure 1), with pore walls being rough and irregular.

To simulate this geometry, a capillary tube with a square cross-section (Legait, 1983a,b) and having an axial constriction has been used (Figure 2a,b). This geometry, because of the corners, allows the simultaneous flow of the two fluid phases in each cross-section. It is preferred to a triangular section (Singhal and Somerton, 1970) because it optimizes saturation of the wetting fluid in a cross-section; but, qualitatively, results do not depend on the geometry of the cross-section.

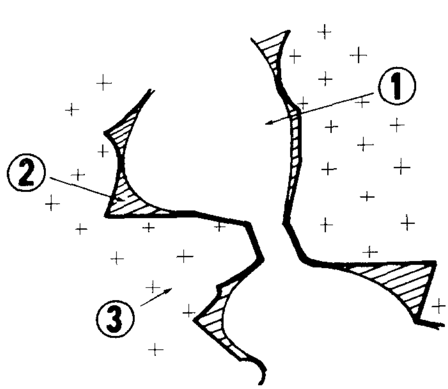

Fig. 1. Fluid distribution at pore scale: ① non-wetting fluid; ② wetting fluid; ③ solid phase.

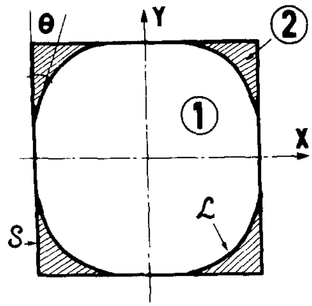

Fig. 2a. Cross-section of the capillary tube.

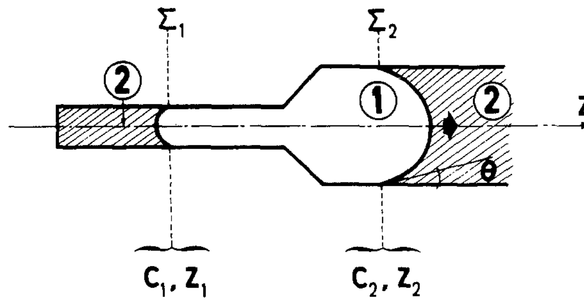

Fig. 2b. Longitudinal section of the capillary tube.

The following assumptions are made to model the flow:

H_1 The capillary tube is slender. z-orthogonal components of fluid phase velocities are thus neglected.
H_2 Flow is slow enough to make inertial and time-dependent terms negligible.
H_3 Fluid/fluid interface curvature is a function only of z. Fluid/fluid interface $\mathcal{L}$ is constituted by four arcs of a circle.

Under these assumptions, the ruling equations are Stokes' equations with standard boundary conditions:

$$i = 1, 2; \quad \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right) u_i = \frac{1}{\mu_i} \cdot \frac{\partial P_i}{\partial z}, \quad \forall (x, y) \in \Omega_i, \tag{1}$$

(B.C.1) $u_1 = u_2 = 0$ along $\mathcal{S}$, the solid surface,
(B.C.2) $\mu_1 \nabla u_1.\mathbf{n} = \mu_2 \nabla u_2.\mathbf{n}$; $u_1 = u_2$ along $\mathcal{L}$,
(B.C.3) $(P_1 - P_2)(z) = \gamma C(z)$,

where Ω_i is the domain occupied by fluid phase i in a cross-section.

Since H_2, pressure gradients are linear functions of the flow rates q_i:

$$q_i = r^4(z) \sum_{j=1,2} C_i^j \frac{1}{\mu_j} \frac{\partial P_j}{\partial z}. \tag{2}$$

Coefficients C_i^j depend on the distribution of fluids in a cross-section (e.g. saturation s_1 and contact angle θ) and viscosity ratio $\bar{\mu}$. They can be numerically calculated by using a finite elements method (Legait, 1983a; Kalaydjian and Legait, 1987a). It must be underlined that matrix (C_i^j) is found to be symmetrical from numerical calculations.

2.2. EXPERIMENTAL

Spontaneous capillary displacement of a nonwetting fluid ganglion by a wetting fluid is typically due to viscous interaction between fluid phases. The experiment consists of first positioning the capillary tube horizontally (to avoid gravity effects) and then filling it with water. Then an oil-ganglion is positioned in such a way that one of its menisci is in the section of half-side R_1 and the other in the section of half-side R_2 ($R_2 > R_1$) (Figure 2a). Since the fine section has been closed, oil-ganglion moves spontaneously towards the big section because of the differential of capillary pressures (proportional to curvature). This countercurrent displacement (oil and water moving in opposite directions. and total flow being nul) ends when both of the curvatures become equal. The measurements concern the positions of one of the menisci as a function of time.

2.3. ESTIMATION OF VISCOUS COUPLING ON THE PORE SCALE

Integration of Equation (2) along the ganglion, leads to:

$$\gamma[C_2 - C_1] = q_1 \int_{z_1}^{z_2} \frac{B_1^1 \mu_1 - B_2^1 \mu_2}{r^4(z)}\, \mathrm{d}z + q_2 \int_{z_1}^{z_2} \frac{B_1^2 \mu_1 - B_2^2 \mu_2}{r^4(z)}\, \mathrm{d}z. \tag{3}$$

Matrix (B_i^j) is the inverse of matrix (C_i^j), while C_i is the curvature of the meniscus Σ_i and is given by the following result (Mayer and Stowe, 1965):

$$C_i = \frac{F(\theta)}{R_i},$$

$$F(\theta) = \frac{\theta + \cos^2(\theta) - \pi/4 - \sin(\theta)\cos(\theta)}{\cos(\theta) - \sqrt{\pi/4 - \theta + \sin(\theta)\cos(\theta)}}. \tag{4}$$

Considering that the total flow is nul, Equation (3) can be used to calculate, for a given position of the ganglion, flow rate Q ($Q = |q_1| = |q_2|$), from assumptions as to fluid distribution in a cross-section, e.g. concerning contact angle values θ. For this experiment, the contact angle must be 13° in order to match theoretical and experimental results (Figure 3). For other pairs of fluid with different viscosity ratios varying from 1 to 100, contact angles vary between 10° and 15°. These values are in good agreement with experimental values of contact angles estimated in porous media for displacements in imbibition (Ngan and Dussan, 1982).

With the modelling thus being validated, Equation (2) may be interpreted in terms of relative permeabilities since it gives a relationship between flow rates and pressure gradients. By taking average values of each velocity u_i of fluid phase i in the domain Ω_i, this formalism does not lead to the standard formulation of Darcy's laws but to an extended formulation of these laws by means of a matrix of relative permeabilities $(k_{r_i^j})$ also referred to as the *matrix of interaction*:

$$q_1 = \frac{k.k_{r_1^1}}{\mu_1}\left(-\frac{\partial P_1}{\partial z}\right) + \frac{k.k_{r_1^2}}{\mu_2}\left(-\frac{\partial P_2}{\partial z}\right),$$

$$q_2 = \frac{k.k_{r_2^1}}{\mu_1}\left(-\frac{\partial P_1}{\partial z}\right) + \frac{k.k_{r_2^2}}{\mu_2}\left(-\frac{\partial P_2}{\partial z}\right). \tag{5}$$

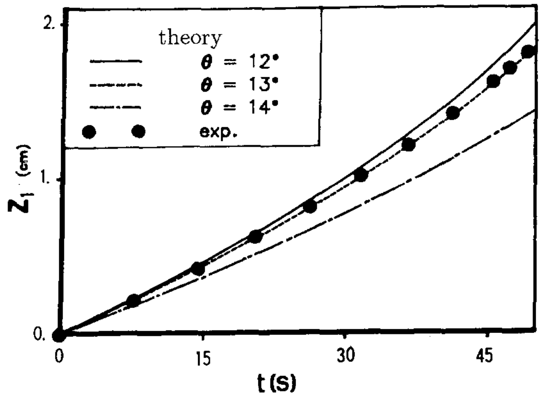

Fig. 3. Kinetics of counter-current flow, theory versus experiment, influence of contact angle.

Diagonal terms $k_{r_1}^1$ and $k_{r_2}^2$ represent the permeability available to the flow of each fluid phase if it were alone, while nondiagonal terms $k_{r_1}^2$ and $k_{r_2}^1$ represent the viscous coupling effect exerted between fluid phases. They are linked by the following equation:

$$\frac{k_{r_1}^2}{\mu_2} = \frac{k_{r_2}^1}{\mu_1}. \tag{6}$$

This result may be justified from Onsager's reciprocity equations, assuming that the thermodynamics of irreversible processes is applicable. This assumption is possible if thermodynamic relationships are assumed to remain identical on microscopic and macroscopic scales. Equation (6) may also be justified by the generalization, on the macroscopic scale, of the result obtained on the microscopic one (Equation (2)), using calculations for the capillary tube. This generalization comes from the linearity of the equations involved.

Comparison between Equations (2) and (5) explicitly determines the coefficients of the matrix:

$$k_{r_i}^i = \frac{-B^* B_j^j}{\det(B_i^j)},$$

$$k_{r_i}^j = \frac{B^* B_i^j}{\det(B_i^j)} \quad (i, j = 1, 2;\ i \neq j). \tag{7}$$

It is then possible to calculate the variations of the relative permeability matrix coefficients versus the state of saturation in a cross-section for a given contact angle and a given viscosity ratio. These results are shown in Figure 4a for a zero contact angle and for a viscosity ratio equal to one. Results obtained with values of saturation s_1 of less than 0.785, for which there is no longer any contact between the fluid/fluid interface and the solid surface, have to be carefully

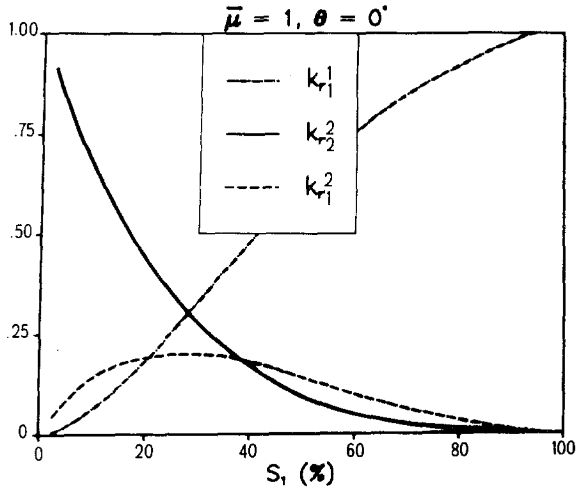

Fig. 4a. Relative permeability matrix ($\bar{\mu} = 1$). Calculations refer to flow in a capillary tube.

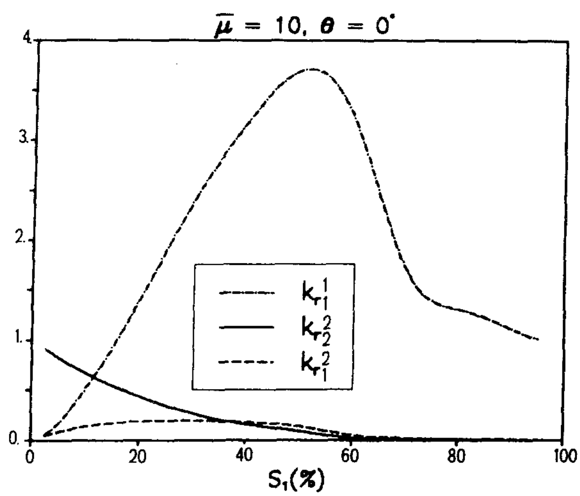

Fig. 4b. Relative permeability matrix ($\bar{\mu} = 10$. Calculations refer to flow in a capillary tube.

extrapolated because of considerations of stability. But, what can be seen is that, in any case, coupling terms are never negligible compared to diagonal terms. Otherwise, it could be said that coupling terms are definitely negligible. This kind of displacement is an overestimation of what could be viscous coupling in porous media given the size of the fluid/fluid interface.

A similar calculation can be made for a viscosity ratio greater than one (Figure 4b). In this figure the diagonal term k_{r1}^1 exceeds one, which can be attributed to a lubrication effect (Danis and Jacquin, 1983).

3. Study on a Macroscopic Level

3.1. RELATIONSHIPS BETWEEN STANDARD TERMS AND COEFFICIENTS FOR THE RELATIVE PERMEABILITY MATRIX

Since the two approaches (the standard one and the one using a matrix of interaction) are available to describe two-phase flow in porous media, it may be useful to examine the possible relationships between the parameters involved in these descriptions. This is achieved in the scalar case by writing the evolution equation verified by the saturation in both of the descriptions.

In the standard case, for nonnul total flow, saturation s_1 is governed by the following equation, where $k_i = k_{ri}/\mu_i$, $i = 1, 2$

$$\frac{u^0}{\phi}\frac{\partial}{\partial x}\left[\varphi(s_1, x) - \psi(s_1, x)\frac{\partial s_1}{\partial x}\right] + \frac{\partial s_1}{\partial t} = 0, \tag{8}$$

with

- $u^0 = u_1 + u_2$,
- $\varphi = \dfrac{k_1}{k_1 + k_2} + \dfrac{k_1 k_2}{k_1 + k_2}\dfrac{k(x)}{u^0}(\rho_1 - \rho_2) g$
- $\psi = \dfrac{k_1 k_2}{k_1 + k_2}\dfrac{k(x)}{u^0}\dfrac{\mathrm{d}P_c}{\mathrm{d}s_1}$

while, in the case where coupling terms are explicitly taken into account, this equation becomes

$$\frac{u^{*0}}{\phi}\frac{\partial}{\partial x}\left[\varphi^*(s_1, x) - \psi^*(s_1, x)\frac{\partial s_1}{\partial x}\right] + \frac{\partial s_1}{\partial t} = 0, \tag{9}$$

with

- $u^{*0} = u_1^* + u_2^*$,
- $\varphi^* = \dfrac{k_1^1 + k_1^2}{\Sigma} + \dfrac{\Delta}{\Sigma}\dfrac{k(x)}{u^{*0}}(\rho_1 - \rho_2) g$,
- $\psi^* = \dfrac{\Delta}{\Sigma}\dfrac{k(x)}{u^{0*}}\dfrac{\mathrm{d}P_c}{\mathrm{d}s_1}$,

where coefficients k_i^j $(i, j = 1, 2)$ equal to $k_{r_i^j}/\mu_j$ and Δ and Σ are defined by

$$\Delta = \det\begin{pmatrix} k_1^1 & k_1^2 \\ k_1^2 & k_2^2 \end{pmatrix}, \tag{10}$$

$$\Sigma = \begin{pmatrix} k_1^1 & k_1^2 \\ k_1^2 & k_2^2 \end{pmatrix} : \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix}. \tag{11}$$

Both of these approaches would be equal if the parameters of Equations (8) and

(9) were simultaneously identical, e.g.

$$u^0 = u^{*0}, \qquad \varphi = \varphi^*, \qquad \psi = \psi^*$$

which would lead to

$$u^0 = u^{*0}, \qquad k_1 = \frac{\Delta}{k_2^2 + k_1^2}, \qquad k_2 = \frac{\Delta}{k_1^1 + k_1^2}. \tag{12}$$

But the equations of this system are not verified simultaneously (Kalaydjian, 1988). More precisely, assuming the last two equations of system (12) to be true, it is not possible (Appendix 1) to obtain equality between u^0 and u^{*0}. But they lead to the following equation for the coupling term k_1^2

$$k_1^2 = \sqrt{(k_1 - k_1^1)(k_2 - k_2^2)}. \tag{13}$$

Relatiohships between parameters can be established for a nonnul total flow only if there is no capillary pressure effect (for high flow velocities, for instance)

$$k_1 = k_1^1 + k_1^2, \qquad k_2 = k_1^2 + k_2^2. \tag{14}$$

In return, for zero total flow (countercurrent conditions), by very similar calculations it is possible to find parameters for both of the approaches, which lead to the same evolution equation. Relationships existing between parameters are again given by the last two equations of (12).

In passing, an interesting result stems from Equations (12) and (14), since it can easily be shown that standard mobilities are not equal for the two displacements. Mobilities estimated in countercurrent conditions are less than those estimated in cocurrent conditions for a high flow rate.

By using relationships obtained for these two kinds of displacement, the following equations between the two couples (k_1, k_2) and (k'_1, k'_2), respectively characterizing a flow in cocurrent conditions with no capillary effect and a countercurrent displacement with zero total flow, and a unique triplet (k_1^1, k_2^2, k_1^2) are derived

$$k_1^2 = \sqrt{(k_1 - k_1^1)(k_2 - k_2^2)}, \qquad k'_1 = \frac{\Delta}{k_1^2 + k_2^2}, \qquad k'_2 = \frac{\Delta}{k_1^1 + k_1^2}. \tag{15}$$

Starting from Equations (15), it is possible (Appendix 2) to express coefficients k_1^1, k_2^2 and k_1^2 as a function of the standard mobilities

$$k_1^1 = \frac{k_1 k_2}{k_1 + k_2} + \frac{\Delta}{k_1 + k_2}\left(1 - \frac{k_1}{k'_1} + \frac{k_1}{k'_2}\right), \tag{16}$$

$$k_2^2 = \frac{k_1 k_2}{k_1 + k_2} + \frac{\Delta}{k_1 + k_2}\left(1 + \frac{k_2}{k'_1} - \frac{k_2}{k'_2}\right), \tag{17}$$

$$k_1^2 = \frac{(k'_2 - k_2)(k_1 - k'_1)}{\dfrac{(k'_2 - k_2)k'_1}{k'_2} + \dfrac{(k'_1 - k_1)k'_2}{k'_1}}. \tag{18}$$

Equations (17) and (18) associated with Equation (10) lead to the following relationship between Δ and k_1^2

$$\Delta = k_1^2(k_1' + k_2') + k_1' k_2'. \tag{19}$$

Thus, by knowing the pairs (k_1, k_2) and (k_1', k_2'), the method for determining the coefficients of the matrix of interaction consists first in calculating k_1^2 by Equation (18) then in calculating Δ by Equation (19). Likewise, diagonal terms are obtained by Equations (16) and (17).

3.2. EXPERIMENTAL

Very few studies have dealt with experimental studies of displacements in co- and countercurrent conditions for the same pair of fluids in the same porous medium. Lelièvre (1966) has done such a study, though applied to flow in an artificial porous medium, and more recently experiments performed in natural porous media (Bourbiaux and Kalaydjian, 1988) have shown the same trend, e.g. relative permeabilities are smaller when measured in countercurrent conditions than in cocurrent conditions (Figure 5a). This result cannot be explained within the framework of the standard theory of relative permeabilities. But it provides an opportunity to estimate coupling terms for two-phase flow in porous media. By

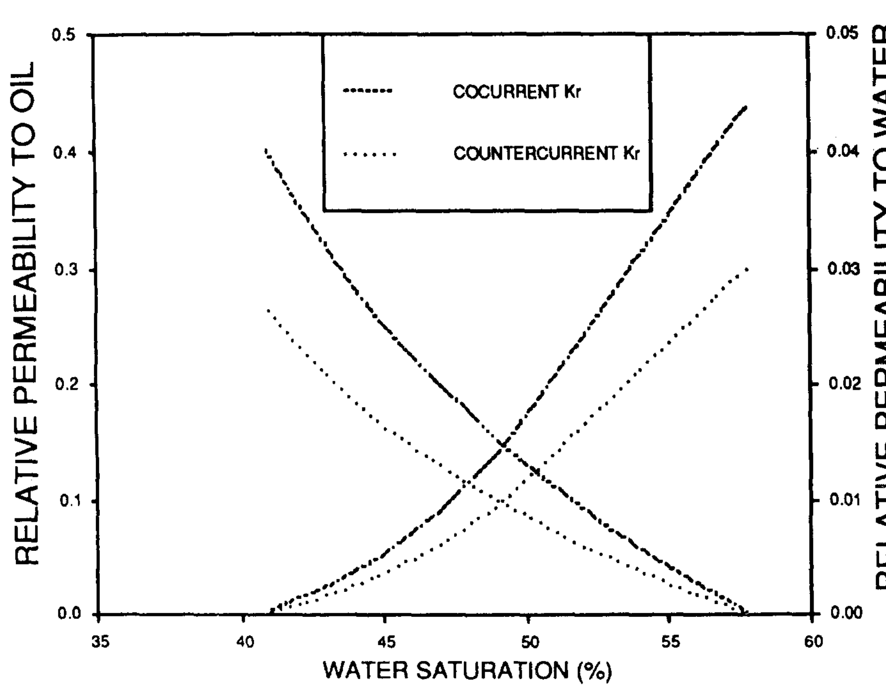

Fig. 5a. Comparison of relative permeabilities measured in natural porous media, in co- and countercurrent conditions (from Bourbiaux and Kalaydjian, 1988).

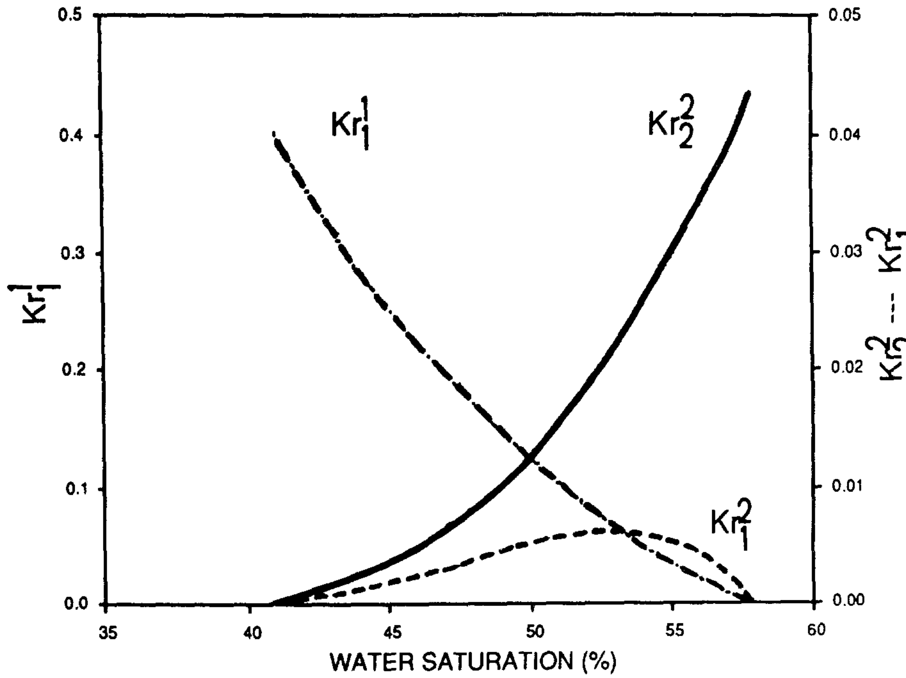

Fig. 5b. Calculation of the relative permeability matrix associated with curves of Figure 5a.

using the method presented above, the coefficients of the matrix of interaction can be calculated. Coupling terms appear to be smaller than for flow in a capillary tube, but cannot however, be neglected in the face of diagonal terms (Figure 5b).

4. Conclusions

The flow of two immiscible and incompressible fluid phases has been studied on the pore level under Stokes' approximation. This study leads to a formulation similar to an extended two-phase Darcy's law involving a symmetrical matrix of mobilities (and therefore three terms of mobilities) instead of the two standard scalar ones. Coupling terms (nondiagonal terms) represent the viscous coupling exerted between fluid phases. On the macroscopic level, the analysis of the relationships between the standard formulation of two-phase flow and the extended formulation developed in this paper provides an experimental way of determining the coefficients of the matrix, which remain identical as macroscopic boundary conditions, such as flow rate, are changed. This determination is based on two experiments. The first one is a cocurrent displacement without any capillary effect; the second one is a countercurrent flow (with zero total flow). Application to flow in porous media shows that coupling terms are nonnegligible and give an explanation for discrepancies measured between standard co- and countercurrent mobilities.

Appendix 1

For a nonnul total flow, the standard approach and the approach using a matrix of interaction are equivalent if and only if the following three equations are simultaneously verified:

$$u^0 = u^{*0}, \qquad k_1 = \frac{\Delta}{k_2^2 + k_1^2}, \qquad k_2 = \frac{\Delta}{k_1^1 + k_1^2}.$$

In the following, all the coefficients are assumed to be positive.

Identity between u^0 and u^{*0} leads to

$$k_1\left(\frac{\partial P_1}{\partial x} - \rho_1 g\right) + k_2\left(\frac{\partial P_2}{\partial x} - \rho_2 g\right)$$

$$= (k_1^1 + k_1^2)\left(\frac{\partial P_1}{\partial x} - \rho_1 g\right) + (k_1^2 + k_2^2)\left(\frac{\partial P_2}{\partial x} - \rho_2 g\right).$$

Then, by using the equations for k_1 and k_2 as function of the coefficients of the matrix, we find

$$\left[\frac{\Delta}{k_1^2 + k_2^2} - (k_1^1 + k_1^2)\right]\left(\frac{\partial P_1}{\partial x} - \rho_1 g\right) +$$

$$+ \left[\frac{\Delta}{k_1^1 + k_1^2} - (k_1^2 + k_2^2)\right]\left(\frac{\partial P_2}{\partial x} - \rho_2 g\right) = 0,$$

which is equivalent to

$$k_1^2 . \Sigma . \left[\frac{1}{k_1^2 + k_2^2}\left(\frac{\partial P_1}{\partial x} - \rho_1 g\right) + \frac{1}{k_1^1 + k_1^2}\left(\frac{\partial P_2}{\partial x} - \rho_2 g\right)\right] = 0.$$

Thus, since $\Delta \neq 0$ (theory of irreversible processing indicating that $\Delta > 0$ (Glansdorff and Prigogine 1971))

$$k_1^2 . \frac{\Sigma}{\Delta} . \left[k_1\left(\frac{\partial P_1}{\partial x} - \rho_1 g\right) + k_2\left(\frac{\partial P_2}{\partial x} - \rho_2 g\right)\right] = 0$$

or

$$k_1^2 . \frac{\Sigma}{\Delta} . u^0 = 0.$$

But, since $\Sigma > 0$ (because of the assumption made above) and since the total flow is nonnul, this equation implies that k_1^2 is nul, which is not true.

Therefore, there is no equivalence between the two approaches in the case of a nonnul total flow with capillary effects.

Appendix 2

Relationships between mobilities in co- and countercurrent flow and coefficients of the matrix of mobilities are given by the following equations:

$$k_1^2 = \sqrt{(k_1 - k_1^1)(k_2 - k_2^2)}, \tag{A2.1}$$

$$k_1' = \frac{\Delta}{k_1^2 + k_2^2}, \tag{A2.2}$$

$$k_2' = \frac{\Delta}{k_1^1 + k_1^2}, \tag{A2.3}$$

where Δ is expressed in terms of the matrix of mobilities (see Equation 10) by

$$\Delta = k_1^1 k_2^2 - (k_1^2)^2. \tag{A2.4}$$

From Equations (A2.2) and (A2.3), we find

$$k_1^1 = -k_1^2 + \frac{\Delta}{k_2'}, \tag{A2.5}$$

$$k_2^2 = -k_1^2 + \frac{\Delta}{k_1'} \cdot \tag{A2.6}$$

Moreover, by taking the square of (A2.1) and by using the Equation (A2.4) for Δ, we find

$$\Delta = k_1 k_2^2 + k_2 k_1^1 - k_1 k_2 . \tag{A2.7}$$

Then, by introducing Equations (A2.5) and (A2.6) into Equation (A2.7), the following equation is derived:

$$k_1\left(\frac{\Delta}{k_1'} - k_1^2\right) + k_2\left(\frac{\Delta}{k_2'} - k_1^2\right) - k_1 k_2 = \Delta ,$$

which leads to

$$k_1^2 = -\frac{k_1 k_2}{k_1 + k_2} + \frac{\Delta}{k_1 + k_2}\left(\frac{k_1}{k_1'} + \frac{k_2}{k_2'} - 1\right). \tag{A2.8}$$

From Equations (A2.5), (A2.6) and (A2.8), the following equations of k_1^1 and k_2^2 can be obtained:

$$k_1^1 = \frac{k_1 k_2}{k_1 + k_2} + \frac{\Delta}{k_1 + k_2}\left(1 - \frac{k_1}{k_1'} + \frac{k_1}{k_2'}\right), \tag{A2.9}$$

$$k_2^2 = \frac{k_1 k_2}{k_1 + k_2} + \frac{\Delta}{k_1 + k_2}\left(1 + \frac{k_2}{k_1'} - \frac{k_2}{k_2'}\right). \tag{A2.10}$$

On the other hand, by introducing Equations (A2.5) and (A2.6) into (A2.5), the following equation between Δ and k_1^2 is derived:

$$\Delta = k_1^2(k_1' + k_2') + k_1' k_2' . \tag{A2.11}$$

Therefore, the combination of Equations (A2.8) and (A2.11) leads to the equation for k_1^2:

$$k_1^2 = \frac{(k_2' - k_2)(k_1 - k_1')}{\frac{(k_2' - k_2)k_1'}{k_2'} + \frac{(k_1' - k_1)k_2'}{k_1'}} . \tag{A2.12}$$

Equations (A2.9), (A2.10) and (A2.12) thus give the equations for the coefficients k_i^j as a function of standard mobilities determined in co- and countercurrent conditions.

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