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MECHANICS OF MULTIPHASE FLUID FLOWS IN VARIABLY SATURATED POROUS MEDIA

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Abstract—This article proposes a set of flow equations governing the simultaneous movement of aqueous and nonaqueous liquids in variably saturated soils. The basic principles and balance laws of continuum mixture theory, along with thermodynamically admissible constitutive laws and simplifying kinematic assumptions, yield a formulation for isochoric multiphase flows through a nondeforming porous matrix. Cast in terms of familiar quantities, the governing equations are similar in form to the classic Richards equation for each liquid phase. The development suggests new rock-fluid properties that must be measured to characterize multiphase flows in the unsaturated zone.

INTRODUCTION

GROUNDWATER contamination by liquids having limited miscibility with water has attracted increasing scientific and legal attention. While studies of groundwater pollution classically have focussed on single-phase flow and transport, many hazardous substances entering our aquifers are relatively insoluble in water and, hence, flow through porous media as separate, nonaqueous liquid phases. The physics of such flows differ substantially from the physics of single-liquid flows. Especially problematic are simultaneous flows of several liquid phases through the variably saturated zones of soils, which contaminants dumped near the earth's surface often must traverse before reaching saturated aquifers. This paper examines the basic mechanics of multiphase flows in variably saturated soils and proposes an extension of the theory of single-liquid flows to cases where water and nonaqueous liquids flow simultaneously.

Investigations of the mechanics of water flowing in the variably saturated zone date to Richards [1]. Indeed, Richards' formulation is now the most widely used model of water movement in unsaturated soils. Prominent among subsequent investigations of the dynamics of partially saturated flow are articles by Philip [2] and Zaslovsky [3]. Bear *et al.* [4] provide an excellent review of the classical literature in this field. Narasimhan and Witherspoon [5] extend these basic models to include the effects of deformation in the solid porous matrix. More recently, studies by Prévost [6] and Bowen [7, 8] have exploited the continuum theory of mixtures, as developed by Eringen and Ingram [9, 10] and reviewed by Atkin and Craine [11], to derive the partial differential equations governing fluid flows in saturated porous media. The present study also relies on the theory of mixtures but aims at a model of unsaturated media containing two liquid phases under some simplifying assumptions.

In contrast to the physics of saturated porous media and variably saturated media with a single liquid, multiliquid flows in the unsaturated zone have received fairly scant experimental attention. Thus the development that follows amounts to a proposed model and should not be viewed as an *a posteriori* explanation of observations.

KINEMATICS

Consider a mixture comprising four constituents, which we shall label R (rock), W (aqueous liquid), N (nonaqueous liquid), and A (air). These constituents represent four phases of concern in the simultaneous flow of water and oily contaminants in the vadose zones of soils. Our aim is to describe the movements of these constituents and, ultimately, to derive flow equations governing their dynamics.

Corresponding to each constituent α is a body \mathcal{B}^{α} , which is a collection of material points labeled X^{α} . The four bodies \mathcal{B}^{R} , \mathcal{B}^{W} , \mathcal{B}^{N} , \mathcal{B}^{A} form a collection of overlapping continua, so that conceptually each spatial point at which the mixture resides may harbor material points from each of the bodies \mathcal{B}^{α} . Following the standard procedure in mixture

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theory, let us establish for each body a reference configuration, so that we can label each material point X^{α} by its spatial position \mathbf{X}^{α} in that configuration. The *motion* of the body \mathcal{B}^{α} is then the function $\mathbf{x} = \mathbf{x}^{\alpha}(\mathbf{X}^{\alpha}, t)$ giving the spatial coordinates of any material point \mathbf{X}^{α} at any time $t \in [0, \infty)$. Under the hypothesis that each motion is a continuously differentiable function with nonzero Jacobian determinant det $[\partial x_{i}^{\alpha}/\partial X_{j}^{\alpha}]$, the inverse function theorem guarantees the existence of an inverse motion defined at each time t by $\mathbf{X}^{\alpha} = \mathbf{X}^{\alpha}(\mathbf{x}, t)$. Given the motions \mathbf{x}^{α} , we can define the Lagrangian and Eulerian velocities

$$\mathbf{v}^{\alpha} = \begin{cases} \frac{\partial \mathbf{x}^{\alpha}}{\partial t} (\mathbf{X}^{\alpha}, t) = \mathbf{v}^{\alpha} (\mathbf{X}^{\alpha}, t) & \text{(Lagrangian),} \\ \frac{\partial \mathbf{x}^{\alpha}}{\partial t} (\mathbf{X}^{\alpha} (\mathbf{x}^{\alpha}, t), t) = \mathbf{v}^{\alpha} (\mathbf{x}^{\alpha}, t) & \text{(Eulerian),} \end{cases}$$

as well as other Lagrangian and Eulerian quantities describing each body's motion.

Also associated with each body \mathcal{B}^{α} is a nonnegative scalar function M^{α} defined on measurable subsets V of the spatial configuration $\mathbf{x}(\mathcal{B}^{\alpha})$ at any time $t \ge 0$. Physically, the value $M^{\alpha}(V, t)$ is the mass of phase α contained in the set V of spatial points at time t. If, at each time t, M^{α} is absolutely continuous with respect to Lebesgue measure on three-dimensional Euclidean space, then, by the Radon-Nikodym theorem, there must be some scalar function $\xi^{\alpha}: \mathbf{x}(\mathcal{B}^{\alpha}) \times [0, \infty) \rightarrow [0, \infty)$ such that

$$M^{\alpha}(V, t) = \int_{V} \xi^{\alpha}(\mathbf{x}, t) \, \mathrm{d}v.$$

The function ξ^{α} is the bulk mass density of α , giving the mass of phase α per unit volume of mixture.

By analogy with the mass density, we can also define the volume fraction occupied by phase α at a given point in the spatial configuration of \mathcal{B}^{α} . To each \mathcal{B}^{α} associate a nonnegative scalar function F^{α} , whose domain at any time *t* is the collection of measurable subsets *V* of $\mathbf{x}(\mathcal{B}^{\alpha})$, such that $F^{\alpha}(V, t)$ gives the volume in *V* occupied by phase α . It is clear that $0 \leq F^{\alpha}(V) \leq \int_{V} dv$. If F^{α} is absolutely continuous with Lebesgue measure on Euclidean three-space, then we have a scalar function ϕ^{α} : $\mathbf{x}(\mathcal{B}^{\alpha}) \times [0, \infty) \rightarrow [0, 1]$ such that

$$F^{\alpha}(V, t) = \int_{V} \phi^{\alpha}(\mathbf{x}, t) \, \mathrm{d}v.$$

The function ϕ^{α} is the volume fraction of phase α , and the collection $\{\phi^{R}, \phi^{W}, \phi^{N}, \phi^{A}\}$ must obey the constraint $\sum \phi^{\alpha} = 1$.

Given the functions ξ^{α} and ϕ^{α} for each phase, we can define the *intrinsic mass densities*. These are $\rho^{\alpha} = \xi^{\alpha}/\phi^{\alpha}$, which are meaningful quantities only where $\phi^{\alpha} \neq 0$, that is, where material from phase α is actually present. The function ρ^{α} gives the mass of phase α per unit volume of phase α . Having established the functions ξ^{α} , ϕ^{α} and ρ^{α} and the phase velocities \mathbf{v}^{α} , we can define a variety of quantities useful in describing the motions of the phases. Table 1 summarizes these definitions.

To describe the rates of change of various quantities with time, we need to introduce material derivatives. As is usual in mixture theory, if f is a Lagrangian quantity, so that $f = f(\mathbf{X}^{\alpha}, t)$, then the material derivative of f with respect to phase α is the time rate of change of f following a fixed material point \mathbf{X}^{α} in phase α :

$$\frac{\frac{\partial f}{\partial t}}{Dt}(\mathbf{X}^{\alpha}, t) = \frac{\partial f}{\partial t}(\mathbf{X}^{\alpha}, t).$$

On the other hand, if f is an Eulerian quantity, implying $f = f(\mathbf{x}^{\alpha}, t)$, then

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Table 1. Definitions of mass-related quantities

SYMBOL	DEFINITION	NAME
ρ ^α	$\xi^{\alpha}/\phi^{\alpha}$	Intrinsic mass density
ρ	$\sum_{\alpha} \phi^{\alpha} \rho^{\alpha}$	Overall mixture density
Ϋ́	$(1/\rho) \sum_{\alpha} \phi^{\alpha} \rho^{\alpha} y^{\alpha}$	Barycentric velocity
ν ^α	y ^a - y	Diffusion velocity
φ	$1 - \phi^R$	Porosity
S^{α} ($\alpha = W, N, A$)	ϕ^{lpha}/ϕ	Saturation of fluid α

$$\frac{\frac{\partial f}{\partial t}}{Dt}(\mathbf{x}^{\alpha}, t) = \frac{\partial f}{\partial t}(\mathbf{x}^{\alpha}, t) + \mathbf{v}^{\alpha}(\mathbf{x}^{\alpha}, t) \cdot \nabla f(\mathbf{x}^{\alpha}, t),$$

where the operator ∇ signifies the gradient with respect to spatial position; in Cartesian coordinates

$$\nabla = \left(\frac{\partial}{\partial x_1}, \frac{\partial}{\partial x_2}, \frac{\partial}{\partial x_3}\right).$$

We can also associate with the mixture a barycentric material derivative, given by

$$\frac{D}{Dt} = \frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla,$$

where v is the barycentric velocity defined in Table 1. The operator D/Dt is related to $\overset{\alpha}{D}/Dt$ as follows:

$$\frac{\overset{\alpha}{D}}{Dt}=\frac{D}{Dt}+\nu^{\alpha}\cdot\nabla,$$

where $\nu^{\alpha} = \mathbf{v}^{\alpha} - \mathbf{v}$ is the diffusion velocity, also defined in Table 1. Finally, we shall encounter intensive variables Ψ^{α} appearing in mass-weighted sums over all phases, as in the equation

$$\rho\Psi = \sum_{\alpha} \phi^{\alpha} \rho^{\alpha} \Psi^{\alpha},$$

defining the "mixture property" Ψ . When working with sums of this sort, we shall find the following identity useful:

$$\rho \, \frac{D\Psi}{Dt} = \sum_{\alpha} \phi^{\alpha} \rho^{\alpha} \frac{\overset{\alpha}{D}\Psi^{\alpha}}{Dt} - \nabla \cdot \sum_{\alpha} \phi^{\alpha} \rho^{\alpha} \Psi^{\alpha} \nu^{\alpha}. \tag{1}$$

BALANCE LAWS

The equations governing multiphase contaminant flows in the unsaturated zone arise from the balance laws for mixtures, modified by constitutive assumptions and restrictions imposed by the Clausius–Duhem inequality of thermodynamics. We shall stipulate that

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the multiphase mixtures of interest are isothermal and have no heat sources, so that it will not be necessary to solve an energy balance equation explicitly. However, the mass, momentum, angular momentum, and energy balances, together with an entropy inequality corresponding to the second law of thermodynamics, are all essential to the complete dynamic specification of the systems. The mixture balance laws, in their primitive form, assert relationships among certain integrals over material volumes and their bounding surfaces. A standard sequence of arguments reduces these integral laws to differential forms involving sums of densities, fluxes, and sources over all constituents in the mixture. Then, by introducing constituent exchange terms, one can reduce the differential balance laws for the mixture to differential laws for each constituent. Since Bowen [7] reviews this development, the present section simply states the balance laws and gives their particular forms under appropriate assumptions about the mixture.

The differential form of the mass balance for any phase α is

$$\frac{\overset{\alpha}{D}}{Dt}(\phi^{\alpha}\rho^{\alpha})+\phi^{\alpha}\rho^{\alpha}\nabla\cdot\mathbf{v}^{\alpha}=r^{\alpha},$$

where r^{α} signifies the rate of exchange of mass into phase α from other phases as a result of chemical reactions, phase changes, adsorption, dissolution, and the like. To be consistent with the global mass balance for the mixture, the exchange terms r^{α} must satisfy $\sum_{\alpha} r^{\alpha} = 0$. We shall simplify matters by allowing no interphase mass transfer, so

that each $r^{\alpha} = 0$, and the mass balance reduces to

$$\frac{\overset{\alpha}{D}}{Dt}(\phi^{\alpha}\rho^{\alpha})+\phi^{\alpha}\rho^{\alpha}\nabla\cdot\mathbf{v}^{\alpha}=0.$$
(2)

It is worth noting that exchange terms may be present in many contaminant flows of practical interest, where dissolution and microbial degradation of organic liquids may be significant (Schwille [12]). In these cases one must retain r^{α} in eqn (2).

The primitive differential momentum balance is

$$\frac{\overset{\alpha}{D}}{Dt}(\phi^{\alpha}\rho^{\alpha}\mathbf{v}^{\alpha})+\phi^{\alpha}\rho^{\alpha}\mathbf{v}^{\alpha}\nabla\cdot\mathbf{v}^{\alpha}-\nabla\cdot\mathbf{t}^{\alpha}-\phi^{\alpha}\rho^{\alpha}\mathbf{b}^{\alpha}=\hat{\mathbf{p}}^{\alpha}.$$

Here \mathbf{t}^{α} denotes the stress tensor for phase α , \mathbf{b}^{α} signifies the rate at which body forces contribute to the momentum density, and $\hat{\mathbf{p}}^{\alpha}$ represents the exchange of momentum into phase α from other phases. As in the mass balances, the exchange terms must obey the restriction $\sum_{\alpha} \hat{\mathbf{p}}^{\alpha} = \mathbf{0}$. By expanding the primitive momentum balance and eliminating

terms that sum to zero according to the mass balance, one finds

$$\phi^{\alpha}\rho^{\alpha}\frac{\overset{\alpha}{D}\mathbf{v}^{\alpha}}{Dt}-\nabla\cdot\mathbf{t}^{\alpha}-\phi^{\alpha}\rho^{\alpha}\mathbf{b}^{\alpha}=\hat{\mathbf{p}}^{\alpha}.$$
(3)

In the mechanics of single continua it is well known that the primitive balance of angular momentum reduces, in the absence of body couples, to the symmetry of the stress tensor, $\mathbf{t} - \mathbf{t}^{T} = \mathbf{0}$. Here \mathbf{t}^{T} denotes the transpose of the stress tensor \mathbf{t} . For mixtures the analogous argument leads to a weaker statement, namely,

$$\mathbf{t}^{\alpha} - (\mathbf{t}^{\alpha})^{\mathrm{T}} = \mathbf{M}^{\alpha}$$

where \mathbf{M}^{α} stands for the exchange of angular momentum into phase α from other phases.

Let us assume that angular momentum exchanges are absent. In this case the angular momentum balance for any constituent α reduces to $\mathbf{t}^{\alpha} - (\mathbf{t}^{\alpha})^{T} = \mathbf{0}$, that is, the stress tensor of each phase is symmetric.

The primitive form of the differential energy balance is

$$\frac{\alpha}{Dt} \left(\phi^{\alpha} \rho^{\alpha} E^{\alpha} + \frac{1}{2} \phi^{\alpha} \rho^{\alpha} \mathbf{v}^{\alpha} \cdot \mathbf{v}^{\alpha} \right) + \phi^{\alpha} \rho^{\alpha} (E^{\alpha} + \frac{1}{2} \mathbf{v}^{\alpha} \cdot \mathbf{v}^{\alpha}) \nabla \cdot \mathbf{v}^{\alpha} - \nabla \cdot (\mathbf{q}^{\alpha} + \mathbf{t}^{\alpha} \cdot \mathbf{v}^{\alpha}) - \phi^{\alpha} \rho^{\alpha} h^{\alpha} - \phi^{\alpha} \rho^{\alpha} \mathbf{h}^{\alpha} \cdot \mathbf{v}^{\alpha} = \epsilon^{\alpha}$$

In this equation, E^{α} is the internal energy of phase α per unit mass, \mathbf{q}^{α} is the heat flux vector in phase α , and h^{α} is the rate of contribution to the total energy per unit mass from heat sources. The term $\frac{1}{2}\phi^{\alpha}\rho^{\alpha}\mathbf{v}^{\alpha}\cdot\mathbf{v}^{\alpha}$ clearly accounts for kinetic energy; $-\nabla \cdot (\mathbf{t}^{\alpha}\cdot\mathbf{v}^{\alpha})$ is the rate of working and heating attributable to stress, and $-\phi^{\alpha}\rho^{\alpha}\mathbf{b}^{\alpha}\cdot\mathbf{v}^{\alpha}$ represents the rate of working of body forces. The quantity ϵ^{α} on the right of the energy balance again stands for the rate of exchange of energy into phase α from other phases, subject to the restriction $\sum \epsilon^{\alpha} = 0$.

As with the momentum balance, it is possible to eliminate certain terms from the primitive energy balance by observing that their sum is proportional to the left side of the mass balance (2). Furthermore, one can notice that several "mechanical energy" terms in the energy balance also appear when one forms the dot product of the momentum balance (3) with v^{α} . Using the mass and momentum balances in this way to simplify the energy balance yields a "thermal energy balance"

$$\phi^{\alpha}\rho^{\alpha}\frac{\overset{\alpha}{D}E^{\alpha}}{Dt}-\mathbf{t}^{\alpha}:\nabla\mathbf{v}^{\alpha}-\nabla\cdot\mathbf{q}^{\alpha}-\phi^{\alpha}\rho^{\alpha}h^{\alpha}=\epsilon^{\alpha}-\hat{\mathbf{p}}^{\alpha}\cdot\mathbf{v}^{\alpha}.$$
(4)

For our purposes the most useful energy balance is not the balance equation for each phase but rather the overall balance for the mixture. To get this equation, simply sum equation (4) over all phases α . Bearing in mind the identity (1) we find, after simplifying,

$$\frac{D}{Dt}\sum_{\alpha}\phi^{\alpha}\rho^{\alpha}E^{\alpha} + (\nabla \cdot \mathbf{v})\sum_{\alpha}\phi^{\alpha}\rho^{\alpha}E^{\alpha} - \sum_{\alpha}\mathbf{t}^{\alpha}:\nabla\mathbf{v}^{\alpha} + \nabla \cdot \sum_{\alpha}\left(\phi^{\alpha}\rho^{\alpha}E^{\alpha}\nu^{\alpha} - \mathbf{q}^{\alpha}\right) - \sum_{\alpha}\phi^{\alpha}\rho^{\alpha}h^{\alpha} = \sum_{\alpha}\mathbf{p}^{\alpha}\cdot\nu^{\alpha}.$$

Now define the inner part E_I of the total internal energy as

$$E_I = \frac{1}{\rho} \sum_{\alpha} \phi^{\alpha} \rho^{\alpha} E^{\alpha}$$

and the total heat flux q and total heat source h as

$$\mathbf{q} = \sum_{\alpha} (\mathbf{q}^{\alpha} - \phi^{\alpha} \rho^{\alpha} E^{\alpha} \boldsymbol{\nu}^{\alpha}), \qquad h = \frac{1}{\rho} \sum_{\alpha} \phi^{\alpha} \rho^{\alpha} h^{\alpha}.$$

With these definitions the overall energy balance reduces to

$$\rho \frac{DE_l}{Dt} - \sum_{\alpha} \mathbf{t}^{\alpha} \cdot \mathbf{v}^{\alpha} - \nabla \cdot \mathbf{q} - \rho h = \sum_{\alpha} \mathbf{p}^{\alpha} \cdot \boldsymbol{\nu}^{\alpha}.$$
(5)

Finally, certain thermodynamic restrictions on the behavior of the mixture follow

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from the Clausius-Duhem inequality governing entropy changes. There is apparently no universally accepted form of this entropy inequality; Atkin and Craine [11], for example, review the history of this controversy. Passman *et al.* [13] also discuss the entropy inequality, noting some of the less satisfactory aspects of the mixture inequality as compared with the entropy inequality valid for single-constituent continua. The version adopted here is essentially that used in Bowen's [8] development for fluid flow in incompressible porous media. In differential form this law states that

$$\sum_{\alpha} \left[\frac{D}{Dt} \left(\phi^{\alpha} \rho^{\alpha} \eta^{\alpha} \right) + \phi^{\alpha} \rho^{\alpha} \eta^{\alpha} \nabla \cdot \mathbf{v}^{\alpha} - \frac{1}{T^{\alpha}} \nabla \cdot \mathbf{q}^{\alpha} - \frac{1}{T^{\alpha}} \phi^{\alpha} \rho^{\alpha} h^{\alpha} \right] \ge 0,$$

where η^{α} is the entropy per unit mass in the α phase, and T^{α} is the temperature of the α phase, assumed positive. Let us henceforth assume that the phases in the mixture share a constant, spatially uniform temperature T. By defining the total entropy of the mixture as

$$\eta = \frac{1}{\rho} \sum_{\alpha} \phi^{\alpha} \rho^{\alpha} \eta^{\alpha}$$

and using the identity (1), we can then rewrite the primitive inequality as

$$\rho \, \frac{D\eta}{Dt} - \nabla \cdot \sum_{\alpha} \left(\frac{1}{T} \, \mathbf{q}^{\alpha} - \phi^{\alpha} \rho^{\alpha} \eta^{\alpha} \nu^{\alpha} \right) - \sum_{\alpha} \frac{1}{T} \, \phi^{\alpha} \rho^{\alpha} h^{\alpha} \ge 0.$$

Now we can use the energy balance (5) to substitute for the last term on the left of this inequality. After some simplification, this yields

$$-\sum_{\alpha} \phi^{\alpha} \rho^{\alpha} \frac{\overset{\alpha}{D} A^{\alpha}}{Dt} + \sum_{\alpha} \mathbf{t}^{\alpha} : \nabla \mathbf{v}^{\alpha} + \sum_{\alpha} \hat{\mathbf{p}}^{\alpha} \cdot \boldsymbol{\nu}^{\alpha} \ge 0.$$
(6)

The quantity A^{α} appearing in this inequality is the Helmholtz free energy, defined as $A^{\alpha} = E^{\alpha} - T\eta^{\alpha}$.

CONSTITUTIVE ASSUMPTIONS

To apply the balance laws, which have rather general validity, to specific mixtures such as variably saturated porous media, we need to make some assumptions restricting the class of materials to which these laws apply. The restrictions of interest include certain kinematic assumptions regarding the nature of the motions, internal constraints on the possible responses of the bodies, and constitutive laws giving functional relationships between various quantities appearing in the balance equations. The constitutive laws also serve a mathematical purpose in closing the deficit between equations and unknowns in the mechanical formulation of the theory.

Let us assume first that the fluids flow isochorically, so that $D \rho^{\alpha}/Dt = 0$ for each fluid phase α . Thus, while some of the fluids occupying the rock's interstices may exhibit significant compressibilities, the velocities of interest are sufficiently small that the effects of compressibility on the flow field are negligible. Since fluid densities must be positive, the assumption that fluid motions are isochoric reduces the mass balance for fluids (2) to

$$\partial \phi^{\alpha} / \partial t + \nabla \cdot (\phi^{\alpha} \mathbf{v}^{\alpha}) = 0, \qquad \alpha = \mathbf{W}, \mathbf{N}, \mathbf{A}.$$

Second, assume that the rock matrix moves rigidly, so that the Jacobian of its motion is just the identity tensor

$$\nabla_{\mathbf{x}\mathbf{R}}\mathbf{x} = \mathbf{F}^{\mathbf{R}} = \mathbf{1}.$$

This assumption allows us to affix a rigid Cartesian coordinate system to the rock phase, forcing $v^{R} = 0$. Also, since a rigid motion must be isochoric, the rock mass balance becomes

$$\frac{\frac{R}{D\phi^{R}}}{Dt} = \frac{\partial\phi^{R}}{\partial t} = 0.$$

Now define the porosity of the rock matrix to be $\phi = 1 - \phi^{R}$, that is, the total volume fraction available to fluids. Since each fluid phase occupies a fraction of the voids, let us call $S^{\alpha} = \phi^{\alpha}/\phi$ the saturation of fluid α , where $\alpha = W$, N, A.

As constitutive laws for fluid stresses, let us postulate that each fluid phase $\alpha = W$, N, A possesses a stress tensor

$$\mathbf{t}^{\alpha} = -p^{\alpha}\mathbf{1} + \sum_{\beta} \lambda^{\alpha\beta} \operatorname{trace} \left(\mathbf{d}^{\beta}\right)\mathbf{1} + \sum_{\beta} 2\mu^{\alpha\beta}\mathbf{d}^{\beta}.$$
 (7)

Here the dummy index β ranges over all fluid phases. The tensor $\mathbf{d}^{\alpha} = \frac{1}{2} [\nabla \mathbf{v}^{\alpha} + (\nabla \mathbf{v}^{\alpha})^{\mathrm{T}}]$ is the deformation rate in fluid phase α ; $\lambda^{\alpha\alpha} = \lambda^{\alpha}$ and $\mu^{\alpha\alpha} = \mu^{\alpha}$ are the coefficients of viscosity of the fluid α , and $\lambda^{\alpha\beta}$, $\mu^{\alpha\beta}$, $\alpha \neq \beta$, are coefficients of interphase traction between fluids α and β . The parameter p^{α} is the mechanical pressure in fluid phase α . Equation (7) is essentially a generalized version of Newton's law of viscosity.

For the Helmholtz free energies A^{α} we shall adopt a constitutive law of the form

$$A^{\alpha} = A^{\alpha}(\{\phi^{\beta}\}, \rho^{\alpha}, T),$$

where the notation $\{\phi^{\beta}\}$ signifies the set of volume fractions as β ranges over all phases. The temperature dependence indicated in this law, although formally appropriate, will turn out to be trivial, since the mixture is isothermal.

Let us assume that the body forces are entirely attributable to gravity, so that $\phi^{\alpha}\rho^{\alpha}\mathbf{b}^{\alpha} = \rho^{\alpha}g\nabla Z$, where g is the acceleration of the gravitational field, assumed uniform, and Z denotes depth below some datum. If we locate the origin of our Cartesian coordinate system (x_1, x_2, x_3) at this datum, then $Z = -x_3$. Furthermore, we shall restrict attention to multiphase mixtures in which heat sources are absent $(h^{\alpha} = 0)$ and heat fluxes are negligible $(\mathbf{q}^{\alpha} = 0)$.

Finally, we need a constitutive relationship for the momentum exchanges $\hat{\mathbf{p}}^{\alpha}$. The assumption that is common to most theories of flow in porous media is that the exchanges of momentum between fluids and the rock dominate interfluid exchanges and have the form of Stokes drags:

$$\hat{\mathbf{p}}^{\alpha} = \phi^{\alpha} (\mathbf{\Lambda}^{\alpha})^{-1} (\mathbf{v}^{\mathbf{R}} - \mathbf{v}^{\alpha}).$$
(8)

In this equation Λ^{α} is an invertible transformation giving a tensor relationship between momentum exchanges and relative velocities, guaranteeing that $\hat{\mathbf{p}}^{\alpha}$ is objective with respect to changes of frame. Since $\mathbf{v}^{R} = \mathbf{0}$, however, eqn (8) simplifies in our frame of reference to

$$\hat{\mathbf{p}}^{\alpha} = -\phi^{\alpha} (\mathbf{\Lambda}^{\alpha})^{-1} \mathbf{v}^{\alpha}. \tag{9}$$

Physically, Λ^{α} varies with the microscopic configurations of the rock and fluid phases, that is, with both the volume fractions and pore-level geometries of the phases. In practice, however, the pore-level geometries are typically inaccessible to measurement. Therefore we consider Λ^{α} to be a function of the fractions $\{\phi^{\beta}\}$ for a given rock-fluid mixture in which the rock geometry is fixed and the interfacial tensions remain constant. Thus, in a sense, Λ^{α} is a purely phenomenological variable requiring direct measurement for each system of rock and fluids under investigation.

There are now several mechanisms for the transfer of momentum in the mixture. These include transfers through shear stresses within each fluid, interphase tractions, and

direct momentum transfer through fluid drags on the rock matrix. Let us assume that the last of these mechanisms overwhelms the others, so that shear stresses and tractions exert influences on momentum transfers that are negligible compared with the Stokes drag in eqn (9). Thus we need only consider the normal fluid stresses, approximating the constitutive law (7) by

$$\mathbf{t}^{\alpha} = -p^{\alpha}\mathbf{1}, \qquad \alpha = \mathbf{W}, \mathbf{N}, \mathbf{A}.$$

For each fluid the mechanical pressure p^{α} in this law may vary as a function of the fluid density in phase α and of the volume fractions $\{\phi^{\beta}\}$. Thus we have an equation of state $p^{\alpha} = p^{\alpha}(\rho^{\alpha}, \{\phi^{\beta}\}), \alpha = W, N, A$, where the index β ranges over all fluid phases.

The Clausius–Duhem inequality (6) imposes certain thermodynamic restrictions on the functional relationships admissible as constitutive laws. To deduce these restrictions in the general case, one must follow the methodology detailed by Coleman and Noll [14] and extended to mixtures by Ingram and Eringen [10]. This procedure involves expanding the inequality (6) in terms of the functional dependencies in the Helmholtz free energy, then reasoning about the values of certain coefficients, given that a linear combination of material derivatives having arbitrary sign must be nonnegative. The present development adopts a less general tack, examining in a similar fashion the restrictions that the Clausius–Duhem inequality imposes on the particular constitutive laws postulated above.

To begin, let us expand inequality (6) using the chain rule and the functional relationship $A^{\alpha} = A^{\alpha}(\{\phi^{\beta}\}, \rho^{\alpha}T)$:

$$-\sum_{\alpha}\phi^{\alpha}\rho^{\alpha}\left(\frac{\partial A^{\alpha}}{\partial T}\frac{\overset{\alpha}{D}T}{Dt}+\sum_{\beta}\frac{\partial A^{\alpha}}{\partial \phi^{\beta}}\frac{\overset{\alpha}{D}\phi^{\beta}}{Dt}+\frac{\partial A^{\alpha}}{\partial \rho^{\alpha}}\frac{\overset{\alpha}{D}\rho^{\alpha}}{Dt}\right)-\sum_{\alpha}\phi^{\alpha}p^{\alpha}\nabla\cdot\mathbf{v}^{\alpha}+\sum_{\alpha}\hat{\mathbf{p}}^{\alpha}\cdot\boldsymbol{v}^{\alpha}\geq0.$$
 (10)

Now in an isothermal mixture DT/Dt = 0. Also,

$$\frac{\overset{\alpha}{D}\phi^{\beta}}{Dt} = \frac{\overset{\beta}{D}\phi^{\beta}}{Dt} + (\mathbf{v}^{\alpha} - \mathbf{v}^{\beta}) \cdot \nabla \phi^{\beta}, \qquad \frac{\overset{\alpha}{D}\rho^{\alpha}}{Dt} = 0,$$

since each phase's motion is isochoric. What is more, the mass balance for phase α implies

$$\phi^{\alpha} \nabla \cdot \mathbf{v}^{\alpha} = -\frac{\overset{\alpha}{D} \phi^{\alpha}}{Dt} \,.$$

So, (10) becomes

$$-\sum_{\alpha} \left[\phi^{\alpha} \rho^{\alpha} \left(\frac{\partial A^{\alpha}}{\partial \phi^{\alpha}} - p^{\alpha} \right) \frac{\partial \phi^{\alpha}}{Dt} - \hat{\mathbf{p}}^{\alpha} \cdot \mathbf{v} \right] - \sum_{\alpha} \sum_{\alpha \neq \beta} \left[\phi^{\alpha} \rho^{\alpha} \frac{\partial A^{\alpha}}{\partial \phi^{\beta}} \frac{\partial \phi^{\beta}}{Dt} + \frac{\partial A^{\alpha}}{\partial \phi^{\beta}} (\mathbf{v}^{\alpha} - \mathbf{v}^{\beta}) \cdot \nabla \phi^{\beta} \right] \ge 0.$$

Since this inequality must hold for arbitrary variations in the volume fractions ϕ^{β} , we must conclude that

 $\partial A^{\alpha}/\partial \phi^{\beta} = 0, \qquad \alpha \neq \beta,$

and

$$\partial A^{\alpha} / \partial \phi^{\alpha} = p^{\alpha}. \tag{11}$$

This last equation identifies the mechanical pressure of fluid phase α as the pore pressure of that phase. Thus, our constitutive relationship for the Helmholtz free energy reduces to $A^{\alpha} = A^{\alpha}(\phi^{\alpha}, \rho^{\alpha})$, ignoring dependence on temperature in the isothermal mixture.

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Equation (11) allows the pressures in different fluids to differ. Thus, between any two fluids α and β , we can define the difference

$$p_{c\alpha\beta} = p^{\alpha} - p^{\beta} = \frac{\partial A^{\alpha}}{\partial \phi^{\alpha}} (\phi^{\alpha}, \rho^{\alpha}) - \frac{\partial A^{\alpha}}{\partial \phi^{\beta}} (\phi^{\beta}, \rho^{\beta}).$$

This quantity is the capillary pressure between the two phases. From the definition it appears that $p_{c\alpha\beta}$ depends on the volume fractions and densities of each phase; however, if the fluids flow isochorically, the density dependence becomes trivial. Also, since the rock matrix is rigid we can factor the porosity ϕ out of the volume fractions and thus consider the capillary pressure to be a function of fluid saturations:

$$p_{\alpha\alpha\beta} = p_{\alpha\alpha\beta}(S^{\alpha}, S^{\beta}). \tag{12}$$

This functional relationship stands in accord with the usual theories of multiphase flow in porous media. Observe that, in a system where three fluids W, N, A are present, only two capillary pressure functions can be independent.

FLOW EQUATIONS

We are now in a position to combine the balance laws and constitutive equations to derive the equations governing the behavior of multiphase flows in variably saturated porous media. To begin with, for each fluid phase α we have a mass balance

$$\frac{\partial}{\partial t} \left(\phi S^{\alpha} \rho^{\alpha} \right) + \nabla \cdot \left(\phi S^{\alpha} \rho^{\alpha} \mathbf{v}^{\alpha} \right) = 0.$$
(13)

For the rock, rigidity of the matrix and our choice of a coordinate frame in which $\mathbf{v}^{R} = 0$ reduce the mass balance to the equation

$$\partial \phi / \partial t = 0$$

There is also a momentum balance for each phase. We shall not concern ourselves with the rock momentum balance, however, since the rock is rigid and stationary in our frame. For each fluid phase α , though, our constitutive assumptions for \mathbf{t}^{α} , \mathbf{b}^{α} and $\hat{\mathbf{p}}^{\alpha}$ convert eqn (3) to

$$\phi^{\alpha}\rho^{\alpha}\left(\frac{\partial \mathbf{v}^{\alpha}}{\partial t}+\mathbf{v}^{\alpha}\cdot\nabla\mathbf{v}^{\alpha}\right)+\nabla p^{\alpha}-\rho^{\alpha}g\nabla Z=-\phi^{\alpha}(\mathbf{\Lambda}^{\alpha})^{-1}\cdot\mathbf{v}^{\alpha}.$$

As is common in porous-media theories, let us assume that the inertial terms in parentheses on the left of this equation contribute negligibly to the fluid motions, being dominated by the effects of normal stresses, gravity and momentum loss through interactions with the rock matrix. There follows the velocity field equation

$$-\frac{1}{\phi^{\alpha}}\Lambda^{\alpha}\cdot(\nabla p^{\alpha}-\rho^{\alpha}g\nabla Z)=\mathbf{v}^{\alpha}.$$

From a phenomenological point of view, the mobility tensor Λ^{α} accounts for the geometry of the rock matrix, the configurations of rock-fluid interfaces, and the flow properties of the fluids. Treating these influences as separable factors leads us to write $\Lambda^{\alpha} = \mathbf{k} k_{r\alpha} / \mu^{\alpha}$, where **k** is the permeability of the rock, having dimensions $[L^2]$; $k_{r\alpha}$ is the dimensionless relative permeability modeling the effects that other fluids have in blocking the flow of phase α , and μ^{α} is the dynamic viscosity of fluid α , having dimensions $[ML^{-1}T^{-1}]$. Thus the velocity field equation becomes

$$\mathbf{v}^{\alpha} = -\frac{\mathbf{k}k_{r\alpha}}{\phi S^{\alpha}\mu^{\alpha}} (\nabla p^{\alpha} - \rho^{\alpha}g\nabla Z), \tag{14}$$

which is the familiar form of Darcy's law for multiphase flows.

We need not consider the angular momentum balance explicitly as a governing equation for fluids, since the constitutive law $\mathbf{t}^{\alpha} = -p^{\alpha}\mathbf{1}$ guarantees symmetry of the fluid stresses automatically. Furthermore, we shall not make explicit use of the energy balance, since we assume that the mixture is isothermal with no heat fluxes or heat sources. In a strict sense this neglect is unwarranted, since even when the rock matrix is rigid the loss of fluid momenta to the rock must be accompanied by concomitant heating of the matrix (and hence the mixture as a whole) via dissipation through the vibrational modes in the solid. By neglecting the energy balance we are therefore excluding from further consideration porous-media flows in which this dissipative heating is significant.

Finally, in addition to the balance laws we have independent functional relationships $p_{c\alpha\beta} = p_{c\alpha\beta}(S^{\alpha}, S^{\beta})$ for two of the three capillary pressures and the saturation constraint $S^{W} + S^{N} + S^{A} = 1$.

Substituting the fluid velocity field equation (14) into the mass balance equation (13) yields a flow equation

$$\frac{\partial}{\partial t} \left(\phi S^{\alpha} \rho^{\alpha} \right) = \nabla \cdot \left[\frac{\rho^{\alpha} k_{r\alpha} \mathbf{k}}{\mu^{\alpha}} \cdot \left(\nabla p^{\alpha} - \rho^{\alpha} g \nabla Z \right) \right].$$

An equation of this form applies to each fluid phase, $\alpha = W$, N, A. However, in the variably saturated zones of many soils, the effects of air flow on the dynamics of the system are limited to the influence of the air saturation S^A on relative permeabilities and capillary pressures. Since the details of air movement are likely to hold little interest compared to the movements of the liquid phases, let us therefore neglect the flow equation in the case $\alpha = A$.

For the aqueous and nonaqueous liquids the assumption that density variations are collinear with pressure variations—a weaker assumption than we have made in stipulating that the flow is isochoric—allows us to define a hydraulic head H^{α} in phase α (Hubbert [15]) as

$$H^{\alpha} = \frac{1}{g} \int_{p^{\alpha}(0)}^{p^{\alpha}(x_{3})} \frac{\mathrm{d}p^{\alpha}}{\rho^{\alpha}} + \int_{0}^{x_{3}} \mathrm{d}x_{3}.$$

Thus $\nabla H^{\alpha} = (\rho^{\alpha}g)^{-1}\nabla p^{\alpha} - \nabla Z$, and the flow equation for a liquid phase ($\alpha = W, N$) becomes

$$\frac{\partial}{\partial t} \left(\phi S^{\alpha} \rho^{\alpha} \right) = \nabla \cdot \left(\rho^{\alpha} k_{r \alpha} \mathbf{K}^{\alpha} \cdot \nabla H^{\alpha} \right). \tag{15}$$

Here $\mathbf{K}^{\alpha} = \rho^{\alpha} g \mathbf{k} / \mu^{\alpha}$ is the hydraulic conductivity of phase α , defined by analogy with the classical single-liquid case.

By expanding the accumulation term on the left side of eqn (15), it is possible to cast the flow equation into a form where the principal unknown is a head. By the product rule,

$$\frac{\partial}{\partial t} \left(\phi S^{\alpha} \rho^{\alpha} \right) = \phi S^{\alpha} \frac{\partial \rho^{\alpha}}{\partial t} + \phi \rho^{\alpha} \frac{\partial S^{\alpha}}{\partial t} + \rho^{\alpha} S^{\alpha} \frac{\partial \phi}{\partial t} \,.$$

The last term on the right vanishes since the rock matrix is rigid. Also, if we allow the liquids to be compressible (although they flow isochorically), then

$$\phi S^{\alpha} \frac{\partial \rho^{\alpha}}{\partial t} = \phi S^{\alpha} \frac{d \rho^{\alpha}}{d H^{\alpha}} \frac{\partial H^{\alpha}}{\partial t} = s_{s}^{\alpha} \rho^{\alpha} S^{\alpha} \frac{\partial H^{\alpha}}{\partial t} \,,$$

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where $s_s^{\alpha} = \phi g \, d\rho^{\alpha}/dp^{\alpha}$ represents the specific storage of the liquid α in the matrix. Now for flow fields in which density gradients are very small, we may approximate our flow equation by writing

$$\phi \, \frac{\partial S^{\alpha}}{\partial t} + \, s_s^{\alpha} S^{\alpha} \, \frac{\partial H^{\alpha}}{\partial t} = \, \nabla \cdot (k_{r\alpha} \mathbf{K}^{\alpha} \cdot \nabla H^{\alpha}). \tag{16}$$

In a more general setting where $\partial \phi/\partial t \neq 0$, it is common to assume $\phi = \phi(H^{W})$. In this case $\rho^{\alpha}S^{\alpha}\partial\phi/\partial t = \rho^{\alpha}S^{\alpha}\rho^{W}gc_{W}\partial H^{W}/\partial t$, where $c_{W} = (\rho^{W}g\phi)^{-1} d\phi/dH^{W}$ quantifies the matrix compressibility. When several liquid phases are present, one formally encounters a matrix compressibility $c_{\alpha} = (\rho^{\alpha}g\phi)^{-1} d\phi/dH^{\alpha}$ with respect to each liquid. There is clear mathematical convenience—and perhaps some physical plausibility—in assuming $c_{W} = c_{N}$. However, for a rigid solid matrix this issue does not arise.

The conventional formulation of single-liquid flows in the variably saturated zone gives the flow equation in terms of pressure head and moisture capacity instead of hydraulic head and saturation (Pinder and Gray [16, Section 5.4]). To make our multiliquid equation conform with the familiar case, let us define the pressure head Φ^{α} in phase α by the equation

$$\nabla \Phi^{\alpha} = \frac{1}{\rho^{\alpha} g} \, \nabla p^{\alpha},$$

so that $H^{\alpha} = \Phi^{\alpha} - Z$. Also, define the moisture capacity for liquid phase α as $\Theta^{\alpha} = \phi S^{\alpha}$. If we observe that $\partial Z/\partial t = -\partial x_3/\partial t = 0$ and $-\nabla Z = \nabla x_3 = \mathbf{e}_3$, the unit vector in the (upward) x_3 direction, and if we call

$$C^{\alpha} = \phi \, \frac{\mathrm{d}}{\mathrm{d}\Phi^{\alpha}} \left(\Theta^{\alpha}\right) \tag{17}$$

the specific moisture capacity for fluid α , then the flow equation (17) becomes

$$\left(C^{\alpha} + \frac{\Theta^{\alpha} s_{s}^{\alpha}}{\phi}\right) \frac{\partial \Phi^{\alpha}}{\partial t} = \nabla \cdot [k_{r\alpha} \mathbf{K}^{\alpha} \cdot (\nabla \Phi^{\alpha} + \mathbf{e}_{3})].$$
(18)

Equation (18) is a natural extension of Richards' [1] equation for the flow of a single liquid through variably saturated soils. There are several new features to the equation, owing to the peculiar physics of multiphase flows in porous media. For one, we must regard the specific moisture capacity, moisture capacity, specific storage, hydraulic conductivity, and pressure head as pertaining to one liquid or the other, so we have the variables C^{W} , C^{N} , Θ^{W} , Θ^{N} , s_{s}^{W} , s_{s}^{N} , \mathbf{K}^{W} , \mathbf{K}^{N} , Φ^{W} , and Φ^{N} . Also, in the flow equation for each phase there now appears a new parameter, $k_{r\alpha}$, the relative permeability of the medium to the liquid phase α . This new parameter will entail a set of measurements of the medium's response over a continuum of liquid saturations S^{W} and S^{N} , so we may consider $k_{rW} = k_{rW}(S^W, S^A)$ and $k_{rN} = k_{rN}(S^N, S^A)$ for fluids of unchanging composition. Finally, in addition to the three-phase saturation restriction $S^{W} + S^{N} + S^{A} = 1$, we now have two independent capillarity relationships giving, say, p_{cWA} and p_{cNA} as functions of saturation according to eqn (12). Equation (17) already assumes equivalent relationships in terms of pressure head and moisture capacity, namely, $\Theta^{\alpha} = \Theta^{\alpha}(\Phi^{\alpha})$, whose inverse (provided Θ^{α} is bijective and exhibits no hysteresis) is $\Phi^{\alpha} = \Phi^{\alpha}(\Theta^{\alpha})$. Thus, while (18) is formally similar to the classic Richards equation, the extension to multiliquid flows entails the quantification of additional physical effects.

CONCLUSIONS

The foregoing development shows that the fundamentals of mixture physics, together with some relatively simple assumptions about the behaviors of various phases present, lead to a set of governing equations for multiliquid flows in variably saturated soils. This 350

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formulation serves as a natural extension of Richards' single-liquid theory. The extended model suggests the kinds of measurements that will be necessary to characterize multiliquid flows and may thus serve as the basis for designing experiments. Indeed, there is a great need for such experiments in light of growing concern over near-surface contamination. Also needed are numerical studies to identify the behavior of systems governed by eqns (18) and to provide a basis for the practical simulations that will be demanded when our understanding of such systems improves.

The formulation of the model presented here identifies several simplifying assumptions that may not apply in all cases of interest. Noteworthy among these limitations are the rigidity of the solid matrix and the isochoricity of the fluid motions. Development of appropriate flow equations under more relaxed assumptions will therefore render a somewhat more general theory.

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NOTATION

The symbols in square brackets indicate physical dimensions. M stands for mass, L for length, T for time, and θ for temperature.

- Heimholtz free energy $[L^2T^{-2}]$ body force $[LT^{-2}]$ A
- b
- $\mathcal B$ material body
- matrix compressibility $[M^{-2}LT^2]$
- с С specific moisture capacity $[L^{-1}]$
- d deformation rate $[T^{-1}]$
- unit vector in three-space [L] internal energy $[L^2T^{-2}]$ е
- Ε
- generic function f
- F volume occupied by a phase $[L^3]$
- Jacobian of the motion [1] F
- gravitational acceleration $[LT^{-2}]$ g h
- heat source $[L^2T^{-3}]$
- Η hydraulic head [L]
- permeability $[L^2]$ k
- κ hydraulic conductivity $[LT^{-1}]$
- k_{ra} M relative permeability of phase α [1]
- mass [M]
- interphase angular momentum exchange $[ML^{-1}T^{-2}]$ М
- р
- mechanical pressure $[ML^{-1}T^{-2}]$ interphase momentum exchange $[ML^{-2}T^{-2}]$ Ŷ
- capillary pressure between fluids α and $\beta [ML^{-1}T^{-2}]$ р_{сад}
- heat flux $[MT^{-3}]$ q interphase mass exchange $[ML^{-3}T^{-1}]$ r
- S
- fluid saturation [1] specific storage $[L^{-1}]$ 55
- time [T] stress $[ML^{-1}T^{-2}]$ t t
- Τ temperature $[\theta]$
- ¥
- velocity $[LT^{-1}]$ measurable set of spatial points V
- position in spatial coordinates [L] х
- material point
- X X Z position in material coordinates [L] depth below datum [L]
- interphase energy exchange $[ML^{-1}T^{-3}]$ entropy $[L^2T^{-1}\theta^{-1}]$ moisture capacity [1] e
- $\overset{\eta}{\Theta}$
- bulk mass density $[ML^{-3}]$ ξ
- coefficient of viscosity or interphase traction $[ML^{-1}T^{-1}]$ mobility $[M^{-1}L^3T]$ λ
- Λ
- coefficient of viscosity or interphase traction $[ML^{-1}T^{-1}]$ diffusion velocity $[LT^{-1}]$ μ
- v
- $_{\Psi}^{\phi}$ volume fraction or porosity [1]
- intensive quantity

Indices

- Α air
- I inner part
- Ν nonaqueous liquid
- R rock
- W water
- phase index α
- β phase index
- 1.2.3 spatial directions



Figure 13