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Immiscible two-phase fluid flows in deformable porous media

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Abstract

Macroscopic differential equations of mass and momentum balance for two immiscible fluids in a deformable porous medium are derived in an Eulerian framework using the continuum theory of mixtures. After inclusion of constitutive relationships, the resulting momentum balance equations feature terms characterizing the coupling among the fluid phases and the solid matrix caused by their relative accelerations. These terms, which imply a number of interesting phenomena, do not appear in current hydrologic models of subsurface multiphase flow. Our equations of momentum balance are shown to reduce to the Berryman–Thigpen–Chen model of bulk elastic wave propagation through unsaturated porous media after simplification (e.g., isothermal conditions, neglect of gravity, etc.) and under the assumption of constant volume fractions and material densities. When specialized to the case of a porous medium containing a single fluid and an elastic solid, our momentum balance equations reduce to the well-known Biot model of poroelasticity. We also show that mass balance alone is sufficient to derive the Biot model stress–strain relations, provided that a closure condition for porosity change suggested by de la Cruz and Spanos is invoked. Finally, a relation between elastic parameters and inertial coupling coefficients is derived that permits the partial differential equations of the Biot model to be decoupled into a telegraph equation and a wave equation whose respective dependent variables are two different linear combinations of the dilatations of the solid and the fluid.

1. Introduction

The mathematical description of multiphase fluid transport through deformable porous media is a problem of great practical importance to subsurface hydrology, including within its purview diverse applications to underground waste containment, enhanced recovery of petroleum, aquifer remediation, and seismic phenomena in geological formations and unconsolidated earth materials. At the heart of this description lies a physical understanding of linear momentum balance based on the thermomechanical behavior of an elastic solid framework permeated by compressible viscous fluids [1,2]. This behavior, in turn, depends on the thermomechanical properties of the individual phases and on the coupling that exists among them because of mutual interactions [2]. In order to characterize this intricate porous framework-fluid system, a variety of authors [1–12] exploiting methods in continuum physics has contributed to the development of a theory whose complexity thus far has prevented its full assimilation into numerical simulations and field applications.

The continuum theory of mixtures [2,6] provides a rigorous theoretical approach for describing multiphase systems from a strictly macroscopic viewpoint. Raats and Klute [13,14] pioneered this approach for modeling water movement in unsaturated soil, whereas Hilfer [8] and Hilfer and Besserer [11] recently have utilized it to model concurrent water and oil movement through reservoirs containing both connected and dead-end pore space. Zienkiewicz et al. [15], Lewis and Schrefler [16], and Schrefler and Scotta [17] have implemented numerical models of water movement through unsaturated deforming porous media that build on the continuum theory of mixtures while retaining a close formal resemblance to phenomenological models of poroelasticity [18–20]. Coussy et al. [6] made a detailed comparison between these latter models (which stem from some pioneering studies by Biot [21] on the coupling between a solid framework and the fluid permeating it, as caused by changes in applied stress or pore pressure) and continuum mixture theory. Contrasting the natural preference for a Lagrangian approach in poroelasticity models with that for an Eulerian approach in mixture theory, they

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Nomenclature

A elastic coefficient
A_{ff} coefficient related to inertial drag in a one-fluid system
A_{11}, A_{12}, A_{21}, A_{22} coefficients related to inertial drag
$\underline{A}_\alpha$ Helmholtz potential of the phase α
$\overline{\overline{A}}_{\xi\beta}$ isotropic tensors related to inertial drag
$\vec{a}_\alpha$ acceleration of the phase α
$\vec{a}_{\xi\mathrm{s}}$ acceleration of the phase ξ relative to the solid phase
b viscous drag coefficient
C_{D} diffusivity
D ratio of the dynamic and elastic constants, used for decoupling
$\underline{d}_\xi$ volumetric strain rate in the phase ξ
$\overline{\overline{d}}_\alpha$ rate of deformation tensor in the phase α
$e = \vec{\nabla}\cdot\vec{u}_{\mathrm{s}}$ dilatation of the solid phase
$\overline{\overline{e}}$ strain tensor for the solid phase
F linear transformation matrix
G shear modulus of the porous medium
$\vec{g}$ gravitational acceleration
J_α Jacobian determinant for the phase α
K linear transformation matrix
K_{b} bulk modulus of the porous medium
K_{f} bulk modulus of the fluid phase
K_{s} bulk modulus of the solid phase
$\underline{L}$ linear transformation matrix
$\overline{\overline{L}}_\alpha$ velocity gradient tensor of the phase α
$\vec{M}_\alpha$ interphase exchange of momentum for the phase α
$\overline{\overline{O}}(t)$ proper orthogonal transformation
P elastic coefficient [$P = A + 2G$]
p_{f} pressure in the fluid phase
p_{s} mean principal dilatational stress in the solid phase
p_ξ pressure in the phase ξ
$\underline{Q}$ elastic coefficient
$\underline{Q}_\alpha$ interphase exchange of energy for the phase α
$\vec{q}_\alpha$ heating flux vector of the phase α
R elastic coefficient
R_{ff} coefficient related to viscous drag in a one-fluid system
$\underline{R}_{11}$, R_{12}, R_{21}, R_{22} coefficients related to viscous drag
$\overline{\overline{R}}_{\xi\beta}$ isotropic tensors related to viscous drag
S_α entropy of the phase α
T_α temperature of the phase α
$T_{\alpha\mathrm{s}}$ temperature of the phase α relative to the solid phase
$\underline{t}$ time
$\overline{\overline{t}}_\alpha$ stress tensor of the phase α
$\vec{u}_{\mathrm{f}}$ displacement vector of the fluid phase
$\vec{u}_\alpha$ displacement vector of the phase α
V total volume
V_{c} reference velocity
V_α volume occupied by the phase α
$\vec{v}_\alpha$ velocity of the phase α
$\vec{v}_{\mathrm{f}}$ velocity of the fluid phase
$\vec{v}_{\alpha\mathrm{s}}$ velocity of the phase α relative to the solid phase
$\vec{w}$ total relative fluid displacement vector
$\vec{w}_\xi$ average displacement vector of the phase ξ relative to the solid phase
$\vec{X}_\alpha$ material position vector
$\vec{x}_\alpha$ spatial position vector
Z linear transformation matrix

Greek symbols
Γ_α rate of entropy production per unit mass of the phase α
Γ entropy production rate per unit mass of the system
$\overline{\overline{\Pi}}_{\xi\alpha}$ isotropic tensors pertinent to heat transfer
$\underline{\delta}_{\mathrm{f}}$, δ_{s} de la Cruz and Spanos elastic coefficients
$\overline{\overline{\delta}}$ unit tensor
$\varepsilon = \vec{\nabla}\cdot\vec{u}_{\mathrm{f}}$ dilatation of the fluid phase
$\underline{\zeta}$ linearized increment of fluid content
$\vec{\zeta}$ normal coordinate
θ_{f} volume fraction of the fluid phase
θ_α volume fraction of the phase α
λ_ξ constitutive coefficient associated with macroscopic viscosity of the phase ξ
μ_ξ constitutive coefficient associated with macroscopic viscosity of the phase ξ
$\vec{\xi}$ normal coordinate
π interfacial pressure
ρ_{f} material mass density of the fluid phase (mass per unit volume of the fluid phase)
ρ_α material mass density of the phase α (mass per unit volume of the phase α)
$\bar{\rho}_{\mathrm{T}} = \bar{\rho}_1 + \bar{\rho}_2 + \bar{\rho}_{\mathrm{s}}$ partial mass density of the system (total mass per unit total volume)
$\bar{\rho}_\alpha$ partial mass density of the phase α (mass per unit total volume) [$\bar{\rho}_\alpha = \rho_\alpha\theta_\alpha$]
ρ_{11}, ρ_{12}, ρ_{22} inertial coupling coefficients in the Biot model [$\bar{\rho}_{\mathrm{s}} = \rho_{11} + \rho_{12}$, $\bar{\rho}_{\mathrm{f}} = \rho_{12} + \rho_{22}$]
ϕ porosity
$\Delta\phi$ porosity change
$\underline{\vec{\chi}_\alpha}$ ( ) vector-valued mapping function from $\vec{X}_\alpha$ to $\vec{x}_\alpha$
$\overline{\overline{\varpi}}_\alpha$ spin tensor in the phase α

demonstrated equivalence between the two with respect to physical balance equations and the entropy inequality. Long ago, Brutsaert [22,23] illustrated the relevance of poroelasticity to subsurface flow phenomena, following Biot [18], who had put forward a now celebrated [20] model of poroelastic behavior, postulating *inter alia* that inertial drag acts on the solid matrix when it is accelerated relative to the fluid. Despite these long-standing insights, inertial drag has typically not been represented in subsurface multiphase flow models [1,8–15,17,22–25].

On the other hand, the Biot [18,19] model of poroelasticity has been extended to deformable porous media containing two immiscible fluids by Berryman et al. [26] and Santos et al. [27] using a Lagrangian approach, and by Tuncay and Corapcioglu [28] using an Eulerian approach. Inertial drag was considered by Berryman et al. [26] and Santos et al. [27], but not by Tuncay and Corapcioglu [28]. The present paper presents an attempt to include this effect systematically in the Eulerian approach more commonly utilized to describe subsurface multiphase flows in theoretical hydrology [24,29]. Our model is based in the continuum theory of mixtures [2], which is applied to derive balance equations for mass and linear momentum in a porous medium bearing two immiscible fluids. Constitutive relationships needed to make the balance equations tractable are then derived based on objectivity and linearity, along with the entropy inequality, which are well-known constraints used in continuum mechanics [2]. These coupled equations govern the linear behavior of a porous medium containing an elastic solid and two immiscible viscous fluids. After some simplifications, our partial differential equations reduce directly to the model of Berryman et al. [26]. Finally, the Biot model [18] of poroelasticity is considered as a special case of our equations when applied to a porous medium containing a single fluid and an elastic solid. It is demonstrated that mass balance alone is adequate to derive the Biot model linear stress–strain equations [18] if a closure relation for porosity change [4,5] also is invoked. We show that, for dilatational motions, decoupling the Biot model equations is possible by using a similarity transformation while invoking a specific constraint relating the elastic parameters and inertial coupling coefficients in the model. After decoupling, two different linear combinations of the dilatations of the solid and the fluid are found to satisfy respectively a telegraph equation and a wave equation.

2. Macroscopic equations of mass and momentum balance with constitutive relationships

2.1. Preliminaries

The configurations of sets of particles B_α of the phases α ($\alpha = 1, 2, \ldots, N$) in a porous medium at time t occupy simultaneously the same region R bounded by a closed surface ∂R in three-dimensional Euclidian space. Following Truesdell [2], we define $\vec{x}_\alpha$ to be position vectors of material points in B_α at time t and $\vec{X}_\alpha$ to be position vectors of material points in a reference configuration of B_α, customarily taken to be that at time $t = 0$. The *motion* of B_α is the vector-valued smooth mapping

$$\vec{x}_\alpha = \vec{\chi}_\alpha(\vec{X}_\alpha, t), \quad 0 < t < \infty, \tag{1}$$

$$\vec{X}_\alpha = \vec{\chi}_\alpha^{-1}(\vec{x}_\alpha, t), \tag{2}$$

where a superscript -1 denotes the inverse function and, by convention,

$$\vec{X}_\alpha = \vec{\chi}_\alpha^{-1}(\vec{X}_\alpha, 0). \tag{3}$$

The invertible mapping $\vec{\chi}_\alpha$ is differentiable with respect to $\vec{X}_\alpha$ and t as many times as desired. The necessary and sufficient condition for the property of invertibility of the mapping is that the Jacobian determinant J_α of the transformation from $\vec{X}_\alpha$ to $\vec{x}_\alpha$ is not zero:

$$J_\alpha = \det\left(\frac{\partial\vec{\chi}_\alpha}{\partial\vec{X}_\alpha}\right) \neq 0. \tag{4}$$

The velocity $\vec{v}_\alpha$ at time t is

$$\vec{v}_\alpha = \frac{\partial\vec{\chi}_\alpha(\vec{X}_\alpha, t)}{\partial t}. \tag{5}$$

The acceleration $\vec{a}_\alpha$ at time t is

$$\vec{a}_\alpha = \frac{\partial^2\vec{\chi}_\alpha(\vec{X}_\alpha, t)}{\partial t^2}. \tag{6}$$

The velocity gradient tensor $\overline{\overline{L}}_\alpha \equiv \vec{\nabla}\vec{v}_\alpha$ can be uniquely decomposed into a symmetric part $\overline{\overline{d}}_\alpha$ and a skew-symmetric part $\overline{\overline{\varpi}}_\alpha$:

$$\overline{\overline{L}}_\alpha = \overline{\overline{d}}_\alpha + \overline{\overline{\varpi}}_\alpha. \tag{7}$$

The rate of deformation tensor (or stretching tensor) $\overline{\overline{d}}_\alpha$ at time t is

$$\overline{\overline{d}}_\alpha = \frac{1}{2}\left[\vec{\nabla}\vec{v}_\alpha + (\vec{\nabla}\vec{v}_\alpha)^{\mathrm{T}}\right], \tag{8}$$

where a superscript T refers to the transpose. The vorticity (or spin) tensor $\overline{\overline{\varpi}}_\alpha$ at time t is

$$\overline{\overline{\varpi}}_\alpha = \frac{1}{2}[\vec{\nabla}\vec{v}_\alpha - (\vec{\nabla}\vec{v}_\alpha)^{\mathrm{T}}]. \tag{9}$$

The operator of material differentiation is

$$\frac{\mathrm{D}^\alpha(\ )}{\mathrm{D}t} = \frac{\partial(\ )}{\partial t} + \vec{v}_\alpha \cdot \vec{\nabla}(\ ). \tag{10}$$

The volume fraction θ_α is the portion of the total volume occupied by phase α:

$$\sum_\alpha \theta_\alpha = 1, \quad \theta_\alpha = \frac{V_\alpha}{V} \quad (0 \leqslant \theta_\alpha \leqslant 1), \tag{11}$$

where V is the total volume of the system, V_α is the volume occupied by phase α, and saturation of the pore space has been assumed. Finally, the partial density $\overline{\rho}_\alpha$ (mass per unit total volume) is equal to the material density ρ_α (mass per unit volume of phase α) multiplied by the volume fraction θ_α, i.e., $\overline{\rho}_\alpha = \rho_\alpha\theta_\alpha$. All three quantities are assumed to be smooth fields [2].

2.2. Balance of mass and momentum

We commence with the balance equations for mass and momentum taken from the continuum theory of mixtures [2] and specialized to the case of immiscible phases with no chemical reactions and no interphase mass transfer. These balance equations are the same as the macroscale phase conservation equations that appear in current multiphase flow theories [7,9,13,14,24] and numerical simulations [15,17] when similarly specialized.

Balance of mass

$$\frac{D^\alpha(\rho_\alpha\theta_\alpha)}{\mathrm{D}t} + \rho_\alpha\theta_\alpha\vec{\nabla}\cdot\vec{v}_\alpha = 0. \tag{12}$$

Balance of momentum

$$\rho_\alpha\theta_\alpha\frac{D^\alpha\vec{v}_\alpha}{Dt} - \vec{\nabla}\cdot(\bar{\bar{t}}_\alpha) - \rho_\alpha\theta_\alpha\vec{g} - \rho_\alpha\theta_\alpha\vec{M}_\alpha = \vec{0}. \tag{13}$$

Across each phase interface, momentum must be conserved:

$$\sum_\alpha \rho_\alpha\theta_\alpha\vec{M}_\alpha = \vec{0}, \tag{14}$$

where hereafter α signifies the three immiscible phases: fluid phase 1 (1), fluid phase 2 (2), and the solid phase (s). In Eqs. (12)–(14), $\bar{\bar{t}}_\alpha = (\bar{\bar{t}}_\alpha)^{\mathrm{T}}$ is the stress tensor in phase α, $\vec{g}$ is gravitational acceleration, and $\vec{M}_\alpha$ in the fourth term of Eq. (13) is the interphase exchange of momentum for phase α. In Eq. (12), mass is balanced by the rate of change of mass within phase α (first term) and advective mass flux across the boundaries of phase α (second term). In Eq. (13), the first term represents the rate of change of momentum within phase α. The second term specifies surface tractions, i.e., forces per unit volume acting on the boundaries of phase α. The third term is the supply of momentum from a gravitational field.

2.3. Constitutive equations

The mass and momentum balance equations are insufficient in number to provide an analytical or numerical solution for all dependent variables; therefore, constitutive relationships must be constructed to supplement the balance equations [2]. These relationships, which characterize how individual phases in a multiphase system behave and how they interact with the other phases, must agree with experiment. Beyond this obvious requirement, some significant stipulations guide the construction of constitutive relationships [2,30]. *Local action* and *phase separation* stipulate that the behavior of any phase at any $\vec{x}_\alpha$ depends only on vicinal thermokinetic processes (i.e., nearby values of $\chi_\alpha(\vec{X}_\alpha, t)$, $\theta_\alpha(\vec{x}, t)$, and the temperature $T_\alpha(\vec{x}, t)$), and that the field variables describing a phase α (e.g., the Helmholtz potential $A_\alpha(\vec{x}, t)$) depend only on the independent constitutive variables pertaining to that phase [2]. This latter stipulation, phase separation, explicitly recognizes the integrity and physical segregation of phases into connected subregions bounded by interfaces [31]. *Frame-indifference* or objectivity stipulates that independent constitutive variables must obey the transformation relations [32]:

$$S' = S, \tag{15.1}$$

$$\vec{V}' = \overline{\overline{O}}(t)\cdot\vec{V}, \tag{15.2}$$

$$\overline{\overline{T}}' = \overline{\overline{O}}(t)\cdot\overline{\overline{T}}\cdot[\overline{\overline{O}}(t)]^{\mathrm{T}}, \tag{15.3}$$

where the prime indicates the scalar (S), vector ($\vec{V}$), and second-order tensor ($\overline{\overline{T}}$) in a different material frame, and $\overline{\overline{O}}(t)$ designates a time-dependent proper ($\det[\overline{\overline{O}}(t)] = 1$) orthogonal transformation. For example, the partial density is an objective scalar, but velocity is not an objective vector. Relative velocity and relative acceleration, however, possess objectivity [30,32]. In addition, dependent constitutive variables must be isotropic functions of the independent constitutive variables, which means that, if $\vec{m}$ and $\overline{\overline{F}}$ be vector-valued and tensor-valued functions, respectively, of the objective scalars, vectors, and tensors, $\{S_k\}$, $\{\vec{V}_k\}$, and $\{\overline{\overline{T}}_k\}$, their mathematical representations must have the forms [30,33]:

$$\vec{m} = \sum_k a_k\vec{V}_k + \sum_i\sum_j(b_{ij}\vec{V}_i\cdot\overline{\overline{T}}_j + c_{ij}\overline{\overline{T}}_j\cdot\vec{V}_i), \tag{16.1}$$

$$\begin{aligned}\overline{\overline{F}} = {} & \sum_k e_k\overline{\overline{T}}_k + \sum_i\sum_j f_{ij}\vec{V}_i\vec{V}_j \\ & + \sum_i\sum_j\sum_k\sum_l(g_{ijkl}\vec{V}_i\cdot\overline{\overline{T}}_k\vec{V}_j\cdot\overline{\overline{T}}_l \\ & + h_{ijkl}\overline{\overline{T}}_k\cdot\vec{V}_i\vec{V}_j\cdot\overline{\overline{T}}_l + m_{ijkl}\vec{V}_i\cdot\overline{\overline{T}}_k\overline{\overline{T}}_l\cdot\vec{V}_j \\ & + n_{ijkl}\overline{\overline{T}}_k\cdot\vec{V}_i\overline{\overline{T}}_l\cdot\vec{V}_j),\end{aligned} \tag{16.2}$$

where the coefficients a_k, b_{ij}, c_{ij}, e_k, f_{ij}, g_{ijkl}, h_{ijkl}, m_{ijkl}, and n_{ijkl} are scalar-valued functions of the objective scalars $\{S_k\}$ and of the joint invariants [33] which can be constructed with the $\{\vec{V}_k\}$ and $\{\overline{\overline{T}}_k\}$.

In any natural process, constitutive equations cannot violate the second law of thermodynamics as expressed in the *entropy* (or *dissipation*) *inequality*: the net entropy production rate is never negative for a closed system [2]. For a multiphase system described by the continuum theory of mixtures, the entropy inequality can be expressed [2,24]:

$$\bar{\rho}_T \Gamma = \sum_\alpha \rho_\alpha \theta_\alpha \Gamma_\alpha$$
$$= \sum_\alpha \frac{1}{T_\alpha} \left\{ -\rho_\alpha \theta_\alpha \left[\frac{D^\alpha A_\alpha}{Dt} + S_\alpha \frac{D^\alpha T_\alpha}{Dt} \right] + \bar{\bar{t}}_\alpha : \bar{\bar{d}}_\alpha \right.$$
$$\left. + \frac{\vec{q}_\alpha}{T_\alpha} \cdot \vec{\nabla} T_\alpha \right\} - \sum_\alpha \frac{\rho_\alpha \theta_\alpha T_{\alpha s}}{T_\alpha T_s} \widehat{Q}_\alpha$$
$$- \frac{1}{T_s} \sum_\alpha \rho_\alpha \theta_\alpha \vec{M}_\alpha \cdot \vec{v}_{\alpha s} \geqslant 0, \tag{17}$$

where A_α is the Helmholtz potential of the phase α, $\widehat{Q}_\alpha$ is the interphase exchange of energy for the phase α produced by mechanical interactions and subject to $\sum_\alpha \rho_\alpha \theta_\alpha (\vec{v}_\alpha \cdot \vec{M}_\alpha + \widehat{Q}_\alpha) = 0$; S_α is the entropy of the phase α; T_α is its temperature; $\vec{q}_\alpha$ is its heating flux vector; Γ_α is the rate of entropy production per unit mass of the phase α, Γ is the entropy production rate per unit mass of the system; $\bar{\rho}_T \equiv \sum_\alpha \rho_\alpha \theta_\alpha$; $\vec{v}_{\alpha s} = \vec{v}_\alpha - \vec{v}_s$ is the velocity of the phase α relative to that of the solid phase; and $T_{\alpha s} = T_\alpha - T_s$ is a relative temperature.

Although not required for constitutive relationships, linearization of Eqs. (16.1) and (16.2) assures tractability of the governing differential equations after specification of all phenomenological coefficients. The linearized forms of Eqs. (16.1) and (16.2) are:

$$\vec{m} = \sum_k a_k \vec{V}_k, \tag{18.1}$$

$$\bar{\bar{F}} = \sum_k e_k \bar{\bar{T}}_k. \tag{18.2}$$

These truncated equations will be used in Eq. (17) to represent $\vec{M}_\alpha$ and $\bar{\bar{t}}_\alpha$ in Eq. (13).

2.4. Balance equations with constitutive relationships

A multiphase continuum mixture is termed *immiscible* if the volume fractions θ_α are required to be among its dependent constitutive variables [2]. In the present paper, the set of these variables that has been defined typically to describe porous media containing immiscible fluids [1–3,5,7–11,14–17,24,25,28] is extended to include the relative acceleration vectors, $\vec{a}_{\xi s} \equiv \vec{a}_\xi - \vec{a}_s$ $(\xi = 1, 2)$, where $\vec{a}_\xi$ is the acceleration of fluid phase ξ and $\vec{a}_s$ is that of the solid matrix. Using the chain rule to express the functional dependence of the Helmholtz potential for each phase on $\vec{a}_{1s}$ and $\vec{a}_{2s}$ along with the entropy inequality in Eq. (17), one may obtain the following additional restrictions on constitutive relationships (see Appendix A):

$$\frac{\theta_1 \rho_1}{T_1} \frac{\partial A_1}{\partial a^i_{1s}} + \frac{\theta_2 \rho_2}{T_2} \frac{\partial A_2}{\partial a^i_{1s}} + \frac{\theta_s \rho_s}{T_s} \frac{\partial A_s}{\partial a^i_{1s}} = 0, \tag{19.1}$$

$$\frac{\theta_1 \rho_1}{T_1} \frac{\partial A_1}{\partial a^i_{2s}} + \frac{\theta_2 \rho_2}{T_2} \frac{\partial A_2}{\partial a^i_{2s}} + \frac{\theta_s \rho_s}{T_s} \frac{\partial A_s}{\partial a^i_{2s}} = 0, \tag{19.2}$$

$$\frac{\theta_1 \rho_1}{T_1} \frac{\partial A_1}{\partial a^i_{1s}} v^j_{1s} + \frac{\theta_2 \rho_2}{T_2} \frac{\partial A_2}{\partial a^i_{1s}} v^j_{2s} = 0, \tag{19.3}$$

$$\frac{\theta_1 \rho_1}{T_1} \frac{\partial A_1}{\partial a^i_{2s}} v^j_{1s} + \frac{\theta_2 \rho_2}{T_2} \frac{\partial A_2}{\partial a^i_{2s}} v^j_{2s} = 0, \tag{19.4}$$

where $a^i_{\xi s}$ and $v^j_{\xi s}$ are the cartesian coordinates of $\vec{a}_{\xi s}$ and $\vec{v}_{\xi s}$, respectively. Following Gray [24], if A_1 and A_2 are isotropic scalar-valued functions of $\vec{a}_{1s}$ and $\vec{a}_{2s}$, one concludes that A_1 and A_2 must also be independent of $\vec{a}_{1s}$ and $\vec{a}_{2s}$. Thus, by Eq. (19.1), A_s is independent of $\vec{a}_{1s}$. Similarly, by Eq. (19.2), A_s does not depend on $\vec{a}_{2s}$. Therefore, development of the terms in $(D^\alpha A_\alpha)/(Dt)$ in the entropy inequality (Eq. (17)) is not altered by the inclusion of $\vec{a}_{1s}$ and $\vec{a}_{2s}$ as independent constitutive variables.

Following well-known procedures [2] as exemplified in [1,3,24], we can write down constitutive relationships for the fluid stress tensors $\bar{\bar{t}}_\xi$ and momentum-exchange vectors $\vec{M}_\xi$ $(\xi = 1, 2)$ consistent with the linearized expressions in Eqs. (18.1) and (18.2):

$$\bar{\bar{t}}_1 = -p_1 \theta_1 \bar{\bar{\delta}} + \lambda_1 d_1 \bar{\bar{\delta}} + 2\mu_1 \bar{\bar{d}}_1, \tag{20.1}$$

$$\bar{\bar{t}}_2 = -p_2 \theta_2 \bar{\bar{\delta}} + \lambda_2 d_2 \bar{\bar{\delta}} + 2\mu_2 \bar{\bar{d}}_2, \tag{20.2}$$

$$\rho_1 \theta_1 \vec{M}_1 = p_1 \vec{\nabla} \theta_1 + \bar{\bar{R}}_{11} \cdot \vec{v}_{1s} + \bar{\bar{R}}_{12} \cdot \vec{v}_{2s} + \bar{\bar{A}}_{11} \cdot \vec{a}_{1s} + \bar{\bar{A}}_{12} \cdot \vec{a}_{2s} + \sum_\alpha \bar{\bar{\Pi}}_{1\alpha} \cdot \vec{\nabla} T_\alpha, \tag{20.3}$$

$$\rho_2 \theta_2 \vec{M}_2 = p_2 \vec{\nabla} \theta_2 + \bar{\bar{R}}_{22} \cdot \vec{v}_{2s} + \bar{\bar{R}}_{21} \cdot \vec{v}_{1s} + \bar{\bar{A}}_{22} \cdot \vec{a}_{2s} + \bar{\bar{A}}_{21} \cdot \vec{a}_{1s} + \sum_\alpha \bar{\bar{\Pi}}_{2\alpha} \cdot \vec{\nabla} T_\alpha, \tag{20.4}$$

where $p_\xi = (\rho_\xi)^2 (\partial A_\xi / \partial \rho_\xi)$ is the thermodynamic pressure in the phase $\xi (\xi = 1, 2)$; d_ξ is the volumetric strain rate of the ξ-phase, equal to the trace of $\bar{\bar{d}}_\alpha$; $\bar{\bar{\delta}}$ is the unit tensor; λ_ξ and μ_ξ are scalar coefficients ($\lambda_\xi \geqslant 0$, $\lambda_\xi + (2/3)\mu_\xi \geqslant 0$ [24]) associated with *macroscopic* viscous properties of the phase ξ; $\bar{\bar{R}}_{11}$, $\bar{\bar{R}}_{12}$, $\bar{\bar{R}}_{21}$, and $\bar{\bar{R}}_{22}$ are negative semi-definite isotropic tensors [24] related to viscous drag; and $\bar{\bar{\Pi}}_{1\alpha}$ and $\bar{\bar{\Pi}}_{2\alpha}$ are negative semi-definite isotropic tensors [24] pertinent to heat transfer. The coefficients $\bar{\bar{A}}_{11}$, $\bar{\bar{A}}_{12}$, $\bar{\bar{A}}_{21}$, and $\bar{\bar{A}}_{22}$ are isotropic tensors related to inertial drag. It is still necessary to establish a constitutive relationship for the stress tensor of the solid phase, $\bar{\bar{t}}_s$. This will be discussed in Section 4.1. The first term on the right side of Eqs. (20.3) and (20.4) is the force per unit area induced by the gradient of the volume fraction; the second and third terms represent viscous drag, while inertial drag is incorporated into the fourth and fifth terms, and the last term arises from gradients of temperature.

We note in passing that the saturation constraint on the volume fractions in Eq. (11) imposes an *indeterminacy* on the momentum-exchange vectors $\vec{M}_\alpha$ because of the closure condition in Eq. (14) [2]. In particular, the gradient of the left side of Eq. (11) is equal to $\vec{0}$ and, therefore, the quantity $\pi \sum_\alpha \vec{\nabla}\theta_\alpha$ can be subtracted from the left side of Eq. (14) for any scalar multiplier π. It follows that the coefficient of $\vec{\nabla}\theta_\xi$ $(\xi = 1, 2)$ in Eqs. (20.3) and (20.4) is determined by the entropy inequality only to within an arbitrary scalar datum. Only with *ad hoc* constitutive assumptions can a specific physical interpretation of π be developed, which usually is depicted as an ''interfacial pressure'' needed to sustain the saturation constraint on the volume fractions [2].

With the introduction of constitutive relationships from Eqs. (20.1)–(20.4), our equations of mass and momentum balance for a deformable porous medium containing two immiscible fluids are:

Conservation of mass

$$\frac{\mathrm{D}^\alpha(\rho_\alpha\theta_\alpha)}{\mathrm{D}t} + \rho_\alpha\theta_\alpha\vec{\nabla}\cdot\vec{v}_\alpha = 0, \quad (\alpha = s, 1, 2). \tag{21.1}$$

Conservation of momentum

(1) fluid phases

$$\rho_\xi\theta_\xi\frac{\mathrm{D}^\xi\vec{v}_\xi}{\mathrm{D}t} + \theta_\xi\vec{\nabla}p_\xi - \vec{\nabla}\cdot(\lambda_\xi d_\xi\overline{\overline{\delta}} + 2\mu_\xi\overline{\overline{d}}_\xi) - \rho_\xi\theta_\xi\vec{g} - \sum_{\beta=1}^{2}\overline{\overline{R}}_{\xi\beta}\cdot\vec{v}_{\beta\mathrm{s}} - \sum_{\beta=1}^{2}\overline{\overline{A}}_{\xi\beta}\cdot\vec{a}_{\beta\mathrm{s}} - \sum_{\alpha}^{s,1,2}\overline{\overline{\Pi}}_{\xi\alpha}\cdot\vec{\nabla}T_\alpha = \vec{0} \quad (\xi = 1, 2). \tag{21.2}$$

(2) solid phase

$$\rho_\mathrm{s}\theta_\mathrm{s}\frac{\mathrm{D}^\mathrm{s}\vec{v}_\mathrm{s}}{\mathrm{D}t} - \vec{\nabla}\cdot\bar{\bar{t}}_\mathrm{s} - \rho_\mathrm{s}\theta_\mathrm{s}\vec{g} + \sum_{\xi=1}^{2}p_\xi\vec{\nabla}\theta_\xi + \sum_{\xi=1}^{2}\sum_{\beta=1}^{2}\overline{\overline{R}}_{\xi\beta}\cdot\vec{v}_{\beta\mathrm{s}} + \sum_{\xi=1}^{2}\sum_{\beta=1}^{2}\overline{\overline{A}}_{\xi\beta}\cdot\vec{a}_{\beta\mathrm{s}} + \sum_{\xi=1}^{2}\sum_{\alpha}^{s,1,2}\overline{\overline{\Pi}}_{\xi\alpha}\cdot\vec{\nabla}T_\alpha = \vec{0}. \tag{21.3}$$

3. The Berryman–Thigpen–Chen model

3.1. Preliminaries

Elastic wave phenomena in porous media containing compressible viscous fluids are of considerable interest to diverse applications such as the extraction of DNAPLs [34], the measurement of dynamic permeability [35,36], the detection of acoustic phenomena in geological formations [36–39], and the enhancement of oil production [40]. Although there is an abundance of past research [3–5,12,20–22,26–28,36] devoted to explaining the underlying physical mechanisms involved in elastic wave motion through both saturated and unsaturated porous media, the systematic formulation of a model for multiphase fluids that includes inertial coupling has been accomplished only by Berryman et al. [26] based on a variational approach initiated by Drumheller and Bedford [41] and extended to a Lagrangian framework by Berryman and Thigpen [38]. This approach is complementary and, in special cases, equivalent to a formulation using the continuum theory of mixtures, but it differs from the latter significantly by its necessity for blurring the distinction between balance equations and constitutive relationships [2].

It is possible to derive the elastic wave propagation model of Berryman et al. [26] as a special case of Eqs. (21.1)–(21.3) after making some simplifying assumptions consistent with their model:

(i) the fluid phases are macroscopically inviscid ($\lambda_\xi = \mu_\xi = 0$);
(ii) temperature gradients and gravitational effects are neglected;
(iii) the tensors $\overline{\overline{R}}_{\xi\beta}$ and $\overline{\overline{A}}_{\xi\beta}$ are diagonal, with the same principal axes;
(iv) the nonlinear term $\vec{v}_\alpha\cdot\vec{\nabla}\vec{v}_\alpha$ in the material derivative is neglected ($\mathrm{D}^\alpha\vec{v}_\alpha/\mathrm{D}t = \partial^2\vec{u}_\alpha/\partial t^2$, where $\vec{u}_\alpha$ is a displacement vector for the phase α);
(v) *changes* in capillary pressure are neglected ($\vec{\nabla}p_1 = \vec{\nabla}p_2 \equiv \vec{\nabla}p_\mathrm{f}$);
(vi) cross-coupling caused by inertial drag is symmetric ($A_{12} = A_{21}$), while that caused by viscous drag is neglected ($R_{12} = R_{21} = 0$).

As a consequence of these assumptions, Eqs. (21.2) and (21.3) become:

Conservation of momentum

(1) fluid phases

$$\rho_\xi\theta_\xi\frac{\partial^2\vec{u}_\xi}{\partial t^2} + \theta_\xi\vec{\nabla}p_\mathrm{f} - R_{\xi\xi}\left(\frac{\partial\vec{u}_\xi}{\partial t} - \frac{\partial\vec{u}_\mathrm{s}}{\partial t}\right) - \sum_{\beta=1}^{2}A_{\xi\beta}\left(\frac{\partial^2\vec{u}_\beta}{\partial t^2} - \frac{\partial^2\vec{u}_\mathrm{s}}{\partial t^2}\right) = \vec{0} \quad (\xi = 1, 2). \tag{21.2'}$$

(2) solid phase

$$\rho_\mathrm{s}\theta_\mathrm{s}\frac{\partial^2\vec{u}_\mathrm{s}}{\partial t^2} - \vec{\nabla}\cdot\bar{\bar{t}}_\mathrm{s} + \sum_{\xi=1}^{2}p_\mathrm{f}\vec{\nabla}\theta_\xi + \sum_{\xi=1}^{2}R_{\xi\xi}\left(\frac{\partial\vec{u}_\xi}{\partial t} - \frac{\partial\vec{u}_\mathrm{s}}{\partial t}\right) + \sum_{\xi=1}^{2}\sum_{\beta=1}^{2}A_{\xi\beta}\left(\frac{\partial^2\vec{u}_\beta}{\partial t^2} - \frac{\partial^2\vec{u}_\mathrm{s}}{\partial t^2}\right) = \vec{0}. \tag{21.3'}$$

3.2. Model equations

The coupled partial differential equations for elastic wave propagation and attenuation in unsaturated porous media derived by Berryman et al. [26] were established under the assumption that both the volume fraction and material density of each phase are always equal to their values in a reference configuration, denoted by a superscript 0. Following [26], we then define $\vec{w}_\xi$ as the *average displacement vector* of a fluid relative to the solid phase:

$$\vec{w}_\xi = \theta_\xi^0(\vec{u}_\xi - \vec{u}_s) \quad (\xi = 1, 2). \tag{22}$$

This vector is a measure of the flow volume per unit area of each fluid phase relative to the solid. The *total relative fluid displacement vector* is:

$$\vec{w} = \vec{w}_1 + \vec{w}_2 = \theta_1^0(\vec{u}_1 - \vec{u}_s) + \theta_2^0(\vec{u}_2 - \vec{u}_s). \tag{23}$$

A dimensionless variable ζ, defined as the negative divergence of the total relative fluid displacement vector [26]:

$$\zeta = -\vec{\nabla} \cdot \vec{w}, \tag{24}$$

is the *linearized increment of fluid content* for unsaturated porous media. It represents the fractional volume of fluids flowing in or out of a given volume element attached to the solid frame in response to an applied stress [4,19,26].

To derive the coupled partial differential equations in the Berryman et al. [26] model, we divide the momentum balance Eq. (21.2′) applied to fluid phase 1 by θ_1^0 and rearrange the pressure term:

$$\rho_1^0 \frac{\partial^2 \vec{u}_1}{\partial t^2} - \frac{R_{11}}{\theta_1^0}\left(\frac{\partial \vec{u}_1}{\partial t} - \frac{\partial \vec{u}_s}{\partial t}\right) - \frac{A_{11}}{\theta_1^0}\left(\frac{\partial^2 \vec{u}_1}{\partial t^2} - \frac{\partial^2 \vec{u}_s}{\partial t^2}\right) - \frac{A_{12}}{\theta_1^0}\left(\frac{\partial^2 \vec{u}_2}{\partial t^2} - \frac{\partial^2 \vec{u}_s}{\partial t^2}\right) = -\vec{\nabla} p_f. \tag{25}$$

Reorganizing Eq. (25), we have

$$\rho_1^0 \frac{\partial^2 \vec{u}_s}{\partial t^2} + \left[\frac{\rho_1}{\theta_1^0} - \frac{A_{11}}{(\theta_1^0)^2}\right] \frac{\partial^2}{\partial t^2}\theta_1^0\left(\vec{u}_1 - \vec{u}_s\right) - \frac{R_{11}}{(\theta_1^0)^2}\frac{\partial}{\partial t}\theta_1^0\left(\vec{u}_1 - \vec{u}_s\right) - \frac{A_{12}}{\theta_1^0\theta_2^0}\frac{\partial^2}{\partial t^2}\theta_2^0\left(\vec{u}_2 - \vec{u}_s\right) = -\vec{\nabla} p_f. \tag{26}$$

After introduction of $\vec{w}_\xi$ from Eqs. (22), (26) becomes:

$$\rho_1^0 \frac{\partial^2 \vec{u}_s}{\partial t^2} + \left[\frac{\rho_1^0}{\theta_1^0} - \frac{A_{11}}{(\theta_1^0)^2}\right] \frac{\partial^2 \vec{w}_1}{\partial t^2} - \frac{R_{11}}{(\theta_1^0)^2}\frac{\partial \vec{w}_1}{\partial t} - \frac{A_{12}}{\theta_1^0\theta_2^0}\frac{\partial^2 \vec{w}_2}{\partial t^2} = -\vec{\nabla} p_f. \tag{27}$$

In like manner, Eq. (21.2′) applied to fluid phase 2 can be written:

$$\rho_2^0 \frac{\partial^2 \vec{u}_s}{\partial t^2} + \left[\frac{\rho_2^0}{\theta_2^0} - \frac{A_{22}}{(\theta_2^0)^2}\right] \frac{\partial^2 \vec{w}_2}{\partial t^2} - \frac{R_{22}}{(\theta_2^0)^2}\frac{\partial \vec{w}_2}{\partial t} - \frac{A_{21}}{\theta_1^0\theta_2^0}\frac{\partial^2 \vec{w}_1}{\partial t^2} = -\vec{\nabla} p_f. \tag{28}$$

The combination of Eqs. (21.2′) applied to fluid phases 1 and 2 with Eq. (21.3′) for the solid phase yields the equation:

$$\overline{\rho}_T^0 \frac{\partial^2 \vec{u}_s}{\partial t^2} + \rho_1^0 \frac{\partial^2 \vec{w}_1}{\partial t^2} + \rho_2^0 \frac{\partial^2 \vec{w}_2}{\partial t^2} = \vec{\nabla} \cdot (\bar{\bar{t}}_s - \phi^0 p_f \bar{\bar{\delta}}), \tag{29}$$

where $\phi^0 \equiv \theta_1^0 + \theta_2^0$ and $\overline{\rho}_T^0 \equiv \sum_\alpha \rho_\alpha^0 \theta_\alpha^0$. Eqs. (27)–(29) are identical with Eqs. (67)–(69) in Berryman et al. [26] after correction of a minor error in their Eqs. (68) and (69). (The divisor of their coefficients $\rho_{(lg)} = \rho_{(gl)}$ should be $\phi_{(g)}\phi_{(l)}$, corresponding to the divisor of the coefficients $A_{12} = A_{21}$ in Eqs. (27) and (28), $\theta_1^0\theta_2^0$.)

4. The Biot model

4.1. Stress–strain relations

The Biot model [18,19,21,43] has long provided a phenomenological basis for describing elastic wave propagation and attenuation in fluid-filled porous media [6,36]. The linear stress–strain relations in the model were developed using a variational principle based on a general form of the strain energy function for a macroscopic continuum. Supposing that, at the pore scale, the fluid phase is Newtonian and the Hooke–Cauchy law is valid for the solid phase, de la Cruz and Spanos [5,42] have used microscopic volume-averaging to derive these relations, but in doing so neglected the shear strain of the pore volume when they assumed the shear modulus of the solid phase to be the same as the shear modulus of the bulk medium [4]. Pride et al. [4] gave a more comprehensive microscopic-picture derivation of the linear stress–strain relations based on momentum balance without neglect of pore volume strain. In the present section, for a homogeneous porous medium ($\vec{\nabla}\rho_f = \vec{\nabla}\rho_s = \vec{\nabla}\phi = \vec{\nabla}\theta_s = \vec{0}$), we shall derive the Biot model linear stress–strain relations directly from macroscopic mass balance complemented by a linear closure relation for the fractional change in porosity [4,5,42].

Specialized to the case of a homogeneous porous medium saturated with a single fluid by setting subscript $1 = f$, with θ_f then identical to the porosity ϕ, and $\phi = 1 - \theta_s$, Eq. (21.1) can be expressed:

$$\frac{\partial(\rho_f \phi)}{\partial t} + \rho_f \phi \vec{\nabla} \cdot \vec{v}_f = 0, \tag{30.1}$$

$$\frac{\partial(\rho_s \theta_s)}{\partial t} + \rho_s \theta_s \vec{\nabla} \cdot \vec{v}_s = 0. \tag{30.2}$$

Following [44], the bulk modulus of the fluid, K_f, and that of the solid, K_s, are related to the fluid pore pressure, p_f, and the mean principal dilatational stress in the solid phase, p_s, by the definitions:

$$\frac{1}{\rho_f}\frac{\partial \rho_f}{\partial t} \equiv \frac{1}{K_f}\frac{\partial p_f}{\partial t}, \tag{31.1}$$

$$\frac{1}{\rho_s}\frac{\partial \rho_s}{\partial t} \equiv \frac{1}{K_s}\frac{\partial p_s}{\partial t}, \tag{31.2}$$

where $-(1-\phi)p_s \equiv \mathrm{tr}(\bar{\bar{t}}_s)/3$ [4]. With the incorporation of Eqs. (31.1) and (31.2), the mass balance Eqs. (30.1) and (30.2) take the form:

$$\frac{\partial p_s}{\partial t} = \frac{K_s}{1-\phi}\frac{\partial \phi}{\partial t} - K_s \vec{\nabla}\cdot\vec{v}_s, \tag{32.1}$$

$$\frac{\partial p_f}{\partial t} = -\frac{K_f}{\phi}\frac{\partial \phi}{\partial t} - K_f \vec{\nabla}\cdot\vec{v}_f. \tag{32.2}$$

To study the linear stress–strain relations of an elastic porous medium containing a single fluid, de la Cruz and Spanos [5] deduced the macroscopic strain energy function for a saturated poroelastic medium by performing volume-averaging on the energy equations of each phase at the pore scale. Their results revealed that the strain energy function was dependent on porosity change, as well as on the dilatations of the solid and fluid phases, whereas Biot [19] held that the strain energy function should depend solely on the dilatations of the solid and fluid phases. To be consistent with Biot [19], de la Cruz and Spanos [5] proposed a closure relation to connect porosity change to the dilatations of the solid and fluid phases. This closure relation was derived more carefully by Pride et al. [4] to express the three dependent variables in Eqs. (32.1) and (32.2), i.e., ϕ, p_f, and p_s, solely in terms of the dilatations of the solid and fluid phases. The general form of the closure relation is [4,5]:

$$\frac{\partial \phi}{\partial t} = \delta_s \vec{\nabla}\cdot\vec{v}_s - \delta_f \vec{\nabla}\cdot\vec{v}_f, \tag{33}$$

where δ_s and δ_f are dimensionless parameters.

At this juncture, it is useful to revisit the classic Biot-Willis [36,39,43,45] quasistatic experiments to evaluate the two dimensionless parameters in Eq. (33). Consider a saturated porous medium enclosed within a flexible "jacket" and subjected to an externally-applied isotropic stress, with the fluid inside the porous medium allowed to drain freely. The fluid pressure thus remains constant. The compressibility of the porous medium framework is equal to the inverse of the bulk modulus of the porous framework K_b [44]. The equations realizing these two conditions are:

$$\frac{\partial p_f}{\partial t} = 0, \tag{34.1}$$

$$\vec{\nabla}\cdot\vec{v}_s = -\frac{(1-\phi)}{K_b}\frac{\partial p_s}{\partial t}. \tag{34.2}$$

Combining Eq. (33) with Eqs. (32.1) and (32.2), one finds the relationships:

$$\frac{\partial p_s}{\partial t} = K_s\left(\frac{\delta_s}{1-\phi} - 1\right)\vec{\nabla}\cdot\vec{v}_s - \frac{K_s\delta_f}{1-\phi}\vec{\nabla}\cdot\vec{v}_f, \tag{35.1}$$

$$\frac{\partial p_f}{\partial t} = \frac{-K_f\delta_s}{\phi}\vec{\nabla}\cdot\vec{v}_s + K_f\left(\frac{\delta_f}{\phi} - 1\right)\vec{\nabla}\cdot\vec{v}_f. \tag{35.2}$$

A constraint on $\vec{\nabla}\cdot\vec{v}_f$ and $\vec{\nabla}\cdot\vec{v}_s$ can be developed from Eqs. (34.1) and (35.2):

$$\vec{\nabla}\cdot\vec{v}_f = \left(\frac{\delta_s}{\delta_f - \phi}\right)\vec{\nabla}\cdot\vec{v}_s. \tag{36}$$

After elimination of $\vec{\nabla}\cdot\vec{v}_f$ using Eqs. (36), (35.1) becomes:

$$\frac{\partial p_s}{\partial t} = K_s\left[\left(\frac{\delta_s}{1-\phi} - 1\right) - \frac{\delta_f\delta_s}{(1-\phi)(\delta_f-\phi)}\right]\vec{\nabla}\cdot\vec{v}_s. \tag{37}$$

Comparison of Eqs. (34.2) and (37) yields an equation for the ratio of K_b to K_s:

$$\frac{K_b}{K_s} = \frac{\delta_s\phi + \delta_f(1-\phi) - \phi(1-\phi)}{\delta_f - \phi}. \tag{38}$$

Now consider a porous medium immersed in a fluid which is pressurized by an external source until the applied stress is distributed uniformly. The fluid pore pressure is then equal to the mean principal dilatational stress of the solid phase while it is the pore volume that remains constant:

$$\frac{\partial \phi}{\partial t} = 0, \tag{39.1}$$

$$p_s = p_f. \tag{39.2}$$

From Eqs. (32.1), (32.2), (33), (39.1), and (39.2), an equation for the ratio of δ_s to δ_f can be obtained:

$$\frac{\delta_f}{\delta_s} = \frac{K_f}{K_s}. \tag{40}$$

We now combine Eqs. (38) and (40) to determine δ_s and δ_f:

$$\delta_s = \frac{\left(1-\phi-\frac{K_b}{K_s}\right)\frac{\phi K_s}{K_f}}{1-\phi-\frac{K_b}{K_s}+\phi\frac{K_s}{K_f}} \equiv \frac{Q}{K_f}, \tag{41.1}$$

$$\delta_f = \frac{Q}{K_s}, \tag{41.2}$$

where Q is an elastic coefficient defined originally by Biot [18]. After introduction of δ_s and δ_f from Eqs. (41.1) and (41.2), the closure relation in Eq. (33) and the mass balance Eqs. (35.1) and (35.2) can be expressed in the form:

$$\frac{\partial \phi}{\partial t} = \frac{Q}{K_f}\vec{\nabla}\cdot\vec{v}_s - \frac{Q}{K_s}\vec{\nabla}\cdot\vec{v}_f, \tag{42.1}$$

$$-(1-\phi)\frac{\partial p_s}{\partial t} = \left(A + \frac{2}{3}G\right)\vec{\nabla}\cdot\vec{v}_s + Q\vec{\nabla}\cdot\vec{v}_f, \tag{42.2}$$

$$-\phi\frac{\partial p_f}{\partial t} = Q\vec{\nabla}\cdot\vec{v}_s + R\vec{\nabla}\cdot\vec{v}_f. \tag{42.3}$$

In Eqs. (42.1)–(42.3),

$$A \equiv \frac{(1-\phi)K_s\left(1-\phi-\frac{K_b}{K_s}\right)+\phi\left(\frac{K_s}{K_f}\right)K_b}{1-\phi-\frac{K_b}{K_s}+\phi\frac{K_s}{K_f}}-\frac{2}{3}G,$$

G being the shear modulus of the porous medium, and

$$R \equiv \frac{Q\phi}{\left(1-\phi-\frac{K_b}{K_s}\right)}$$

are elastic coefficients defined originally by Biot and Willis [43]. To be consistent with the Biot model [18], all elastic coefficients are assumed to have values equal to those in a reference configuration.

Integrating Eq. (42.1) with respect to t, one obtains:

$$\Delta\phi = \frac{Q}{K_f}\vec{\nabla}\cdot\vec{u}_s - \frac{Q}{K_s}\vec{\nabla}\cdot\vec{u}_f, \tag{43}$$

where $\Delta\phi = \int_0^t(\partial\phi/\partial t)\mathrm{d}t = \phi - \phi^0$ is the difference in porosity between the current configuration and a reference configuration. We now assume $\Delta\phi$ is a small quantity relative to the porosity in the reference configuration: $\Delta\phi/\phi^0 \ll 1$. Then, products of $\Delta\phi$ with stress changes being neglected, Eqs. (42.2) and (42.3) also can be expressed in a time-integrated form:

$$-(1-\phi^0)p_s = \left(A+\tfrac{2}{3}G\right)\vec{\nabla}\cdot\vec{u}_s + Q\vec{\nabla}\cdot\vec{u}_f, \tag{44.1}$$

$$-\phi^0 p_f = Q\vec{\nabla}\cdot\vec{u}_s + R\vec{\nabla}\cdot\vec{u}_f. \tag{44.2}$$

The stress–strain relations in Eqs. (44.1) and (44.2) are identical to those in Eqs. (48) and (49) of Pride et al. [4] if the bulk modulus of the fluid is a constant, consistent with the Biot model [18]. Differences between Eqs. (44.1) and (44.2) and the stress–strain relations derived by de la Cruz and Spanos [5], caused by neglect of pore volume strain, are discussed by Pride et al. [4]. Eqs. (44.1) and (44.2) are also the well-known Biot [18,19,43] linear stress–strain relations. If the solid phase is assumed to obey the Hooke–Cauchy law in an isotropic form at the pore scale, the stress tensor of the solid phase at the macroscopic scale is related to the mean principal dilatational stress of the solid phase [4]:

$$\bar{\bar{t}}_s = -(1-\phi^0)p_s\bar{\bar{\delta}} + 2G\bar{\bar{e}} - \tfrac{2}{3}G\vec{\nabla}\cdot\vec{u}_s\bar{\bar{\delta}}, \tag{45}$$

where $\bar{\bar{e}} \equiv (1/2)(\vec{\nabla}\vec{u}_s + \vec{\nabla}\vec{u}_s^{\mathrm{T}})$ is the strain tensor for the solid phase. Hence, given Eqs. (44.1) and (45), the stress tensor of the solid phase can be written:

$$\bar{\bar{t}}_s = 2G\bar{\bar{e}} + (A\vec{\nabla}\cdot\vec{u}_s + Q\vec{\nabla}\cdot\vec{u}_f)\bar{\bar{\delta}}. \tag{46}$$

as postulated by Biot [18,19]. We note in passing that Berryman et al. [26] adapted Eqs. (44.2) and (46) to their model simply by generalizing the term in $\vec{\nabla}\cdot\vec{u}_f$ to a two-fluid mixture using Eq. (23) (see Eq. (49) below) and by expressing K_f as the harmonic mean of the two fluid bulk moduli. The resulting model equations for p_f and $\bar{\bar{t}}_s$ then were introduced into Eqs. (27)–(29).

The Biot model expression [19] for the linearized increment of fluid content in a fluid-saturated porous medium can be expressed in terms of the porosity change, $\Delta\phi$ [4]. To derive this relation solely from mass balance, let us begin by considering the linearized form of Eqs. (32.1) and (32.2) after time-integration:

$$p_s = \frac{\Delta\phi K_s}{1-\phi^0} - K_s\vec{\nabla}\cdot\vec{u}_s, \tag{47.1}$$

$$p_f = -\frac{K_f}{\phi^0}\Delta\phi - K_f\vec{\nabla}\cdot\vec{u}_f. \tag{47.2}$$

Eqs. (47.1) and (47.2) can be regrouped as the single expression:

$$\left(\frac{\Delta\phi}{1-\phi^0} + \frac{\phi^0}{K_f}p_f - \frac{\phi^0}{K_s}p_s\right) + \phi^0\vec{\nabla}\cdot(\vec{u}_f - \vec{u}_s) = 0. \tag{48}$$

The linearized increment of fluid content analogous to Eq. (24) is defined by [19]:

$$\zeta = \phi^0\vec{\nabla}\cdot(\vec{u}_s - \vec{u}_f). \tag{49}$$

Combination of Eqs. (48) and (49) then yields the result found by Pride et al. [4] using momentum balance:

$$\zeta = \frac{\Delta\phi}{1-\phi^0} + \phi^0\left(\frac{p_f}{K_f} - \frac{p_s}{K_s}\right). \tag{50}$$

Eq. (50) emerges directly from linearized mass balance (Eqs. (32.1) and (32.2)). It conveys the idea [4] that the linearized increment of fluid content should involve the frame contraction (first term) and the difference in intrinsic volumetric strain between the solid and the fluid (second term). If K_f is constant, Eq. (50) is the same as Eq. (39) in Pride et al. [4].

4.2. Coupled momentum balance equations

The Biot model [18,19] can be derived after Eqs. (21.1) and (21.3) are specialized to homogeneous porous medium permeated by a single fluid whose linear stress–strain relations are given by Eqs. (44.2) and (46):

$$\rho_f^0\phi^0\frac{\partial^2\vec{u}_f}{\partial t^2} - R_{ff}\left(\frac{\partial\vec{u}_f}{\partial t} - \frac{\partial\vec{u}_s}{\partial t}\right) - A_{ff}\left(\frac{\partial^2\vec{u}_f}{\partial t^2} - \frac{\partial^2\vec{u}_s}{\partial t^2}\right) = \vec{\nabla}(Qe + R\varepsilon), \tag{51.1}$$

$$\rho_s^0\theta_s^0\frac{\partial^2\vec{u}_s}{\partial t^2} + R_{ff}\left(\frac{\partial\vec{u}_f}{\partial t} - \frac{\partial\vec{u}_s}{\partial t}\right) + A_{ff}\left(\frac{\partial^2\vec{u}_f}{\partial t^2} - \frac{\partial^2\vec{u}_s}{\partial t^2}\right) = G\nabla^2\vec{u}_s + \vec{\nabla}[(A+G)e + Q\varepsilon], \tag{51.2}$$

where $\varepsilon = \vec{\nabla}\cdot\vec{u}_f$ is the dilatation of the fluid phase; $e = \vec{\nabla}\cdot\vec{u}_s$ is the dilatation of the solid phase; and the coefficients A_{ff} and R_{ff} have constant values. Dilatational motions can be investigated after taking the divergence of both sides of Eqs. (51.1) and (51.2):

$$\rho_f^0\phi^0\frac{\partial^2\varepsilon}{\partial t^2} - R_{ff}\left(\frac{\partial\varepsilon}{\partial t} - \frac{\partial e}{\partial t}\right) - A_{ff}\left(\frac{\partial^2\varepsilon}{\partial t^2} - \frac{\partial^2 e}{\partial t^2}\right) = \nabla^2(Qe + R\varepsilon), \quad (52.1)$$

$$\rho_s^0\theta_s^0\frac{\partial^2 e}{\partial t^2} + R_{ff}\left(\frac{\partial\varepsilon}{\partial t} - \frac{\partial e}{\partial t}\right) + A_{ff}\left(\frac{\partial^2\varepsilon}{\partial t^2} - \frac{\partial^2 e}{\partial t^2}\right) = \nabla^2(Pe + Q\varepsilon), \quad (52.2)$$

where $P \equiv A + 2G$ [18,36]. A mass coupling parameter $\rho_{12} < 0$ was proposed by Biot [18] to account for inertial coupling between the solid and fluid phases, with a coupling parameter $b > 0$ doing the same for viscous drag. After making the substitutions:

$$A_{ff} \equiv \rho_{12}, \quad \rho_s^0\theta_s^0 \equiv \rho_{11} + \rho_{12}, \quad \rho_f^0\phi^0 \equiv \rho_{22} + \rho_{12}, \quad R_{ff} \equiv -b, \quad (53)$$

Eqs. (52.1) and (52.2) take the form of Eqs. (7.1) in Part I of Biot [18]:

$$\frac{\partial^2}{\partial t^2}(\rho_{12}e + \rho_{22}\varepsilon) - b\left(\frac{\partial e}{\partial t} - \frac{\partial\varepsilon}{\partial t}\right) = \nabla^2(Qe + R\varepsilon), \quad (54.1)$$

$$\frac{\partial^2}{\partial t^2}(\rho_{11}e + \rho_{12}\varepsilon) + b\left(\frac{\partial e}{\partial t} - \frac{\partial\varepsilon}{\partial t}\right) = \nabla^2(Pe + Q\varepsilon). \quad (54.2)$$

After neglecting the inertial terms on the left side, Chandler and Johnson [46] proposed normal coordinates ($\vec{\xi}$, $\vec{\zeta}$) to decouple Eqs. (54.1) and (54.2):

$$\begin{bmatrix}\vec{\xi}\\ \vec{\zeta}\end{bmatrix} = \begin{bmatrix}1 & -1\\ 1 & \frac{R+Q}{P+Q}\end{bmatrix}\begin{bmatrix}\vec{u}_s\\ \vec{u}_f\end{bmatrix}. \quad (55)$$

We shall demonstrate that the momentum balance equations also can be decoupled with inertial terms included. If the coupled differential Eqs. (54.1) and (54.2) are written in a matrix form:

$$K\nabla^2\begin{bmatrix}e\\ \varepsilon\end{bmatrix} = L\frac{\partial^2}{\partial t^2}\begin{bmatrix}e\\ \varepsilon\end{bmatrix} + bF\frac{\partial}{\partial t}\begin{bmatrix}e\\ \varepsilon\end{bmatrix}, \quad (56)$$

where

$$K = \begin{bmatrix}P & Q\\ Q & R\end{bmatrix}, \quad L = \begin{bmatrix}\rho_{11} & \rho_{12}\\ \rho_{12} & \rho_{22}\end{bmatrix}, \quad \text{and} \quad F = \begin{bmatrix}1 & -1\\ -1 & 1\end{bmatrix},$$

the inverse of the linear transformation $\begin{bmatrix}\vec{\nabla}\cdot\vec{\xi}\\ \vec{\nabla}\cdot\vec{\zeta}\end{bmatrix} = Z\begin{bmatrix}e\\ \varepsilon\end{bmatrix}$ can be used to uncouple Eq. (56):

$$\nabla^2\begin{bmatrix}\vec{\nabla}\cdot\vec{\xi}\\ \vec{\nabla}\cdot\vec{\zeta}\end{bmatrix} = (ZK^{-1}LZ^{-1})\frac{\partial^2}{\partial t^2}\begin{bmatrix}\vec{\nabla}\cdot\vec{\xi}\\ \vec{\nabla}\cdot\vec{\zeta}\end{bmatrix} + b(ZK^{-1}FZ^{-1})\frac{\partial}{\partial t}\begin{bmatrix}\vec{\nabla}\cdot\vec{\xi}\\ \vec{\nabla}\cdot\vec{\zeta}\end{bmatrix}. \quad (57)$$

Eq. (57) shows that decoupling requires *both* matrices in front of the time-derivative terms to be diagonalized simultaneously; i.e., $Z(\ )Z^{-1}$ must be a similarity transformation for *both* $K^{-1}L$ and $K^{-1}F$ to be put into diagonal form.

The linear transformation Z is assumed to take a form similar to that for the Chandler–Johnson [46] normal coordinates, but with an unknown element D:

$$Z = \begin{bmatrix}1 & -1\\ 1 & D\end{bmatrix}.$$

Then we determine D to make the similarity transformation successful for both $K^{-1}L$ and $K^{-1}F$. In Appendix B it is shown that $D = (R+Q)/(P+Q)$ for diagonal $K^{-1}F$, as found by Chandler and Johnson [46], but $(\rho_{12}+\rho_{22})/(\rho_{11}+\rho_{12}) = (R+Q)/(P+Q)$ if $K^{-1}L$ is also to be diagonal. Under these conditions, Eq. (57) decouples into the pair of equations:

$$\nabla^2(\vec{\nabla}\cdot\vec{\xi}) = \frac{\rho_{11}(R+Q) - \rho_{12}(P+Q)}{(PR-Q^2)}\frac{\partial^2}{\partial t^2}(\vec{\nabla}\cdot\vec{\xi}) + \frac{(P+2Q+R)b}{(PR-Q^2)}\frac{\partial}{\partial t}(\vec{\nabla}\cdot\vec{\xi}), \quad (58.1)$$

$$\nabla^2(\vec{\nabla}\cdot\vec{\zeta}) = \frac{(\rho_{11}+2\rho_{12}+\rho_{22})}{(P+2Q+R)}\frac{\partial^2}{\partial t^2}(\vec{\nabla}\cdot\vec{\zeta}) = \frac{(\overline{\rho}_s^0+\overline{\rho}_f^0)}{(P+2Q+R)}\frac{\partial^2}{\partial t^2}(\vec{\nabla}\cdot\vec{\zeta}). \quad (58.2)$$

We can see from Eqs. (58.1) and (58.2) that the momentum balance Eqs. (54.1) and (54.2) are decoupled into a telegraph equation and a wave equation whose respective dependent variables are two different linear combinations of the dilatations of the solid and the fluid, provided that the following relation between the dynamic and elastic constants applies:

$$\frac{\rho_{12}+\rho_{22}}{\rho_{11}+\rho_{12}} = \frac{\overline{\rho}_f^0}{\overline{\rho}_s^0} = \frac{R+Q}{P+Q}. \quad (59)$$

The "$\vec{\nabla}\cdot\vec{\zeta}$ wave", which is not subject to viscous drag, represents a dilatational wave for which there is no relative motion between the solid and fluid [18]. The speed of propagation of this wave is $((P+2Q+R)/(\overline{\rho}_s^0+\overline{\rho}_f^0))^{1/2}$, which is the Biot reference velocity, V_c [18]. The diffusivity associated with the "$\vec{\nabla}\cdot\vec{\xi}$ wave" is $(PR-Q^2)/(P+2Q+R)b$ and is identical to the diffusivity, C_D, derived by Chandler and Johnson [46] for the Biot "slow wave" in the low-frequency limit. Under the constraint in Eq. (59), V_c and C_D are not independent:

$$V_c = \sqrt{\frac{(PR-Q^2)}{C_D b(\overline{\rho}_s^0+\overline{\rho}_f^0)}}. \quad (60)$$

When viscous effects prevail and one may ignore inertial terms, Eqs. (58.1) and (58.2) reduce, respectively, to a diffusion equation and a Laplace equation, as found previously by Chandler and Johnson [46].

5. Conclusions

Equations (21.1)–(21.3) governing the flow of immiscible fluids in deformable porous media are the principal results in this paper. They generalize previous linear models based on the continuum theory of mixtures [2,3,8,11,13,14], or on microscopic volume-averaging [1,24], to include inertial coupling. However, they do not take explicit account of interfacial effects on the entropy inequality, either in respect to thermodynamic potentials [7,9,48] or the saturation constraint in Eq. (11) [2,3]. This difficult issue is a topic for further research on constitutive relationships. In their present form, Eqs. (21.1)–(21.3) still can be applied to extend current numerical modeling of wave phenomena in porous media containing two fluids [12,15–17,25,28,47].

By contrast to the equations of motion developed by Berryman et al. [26], in the momentum balance Eqs. (21.1)–(21.3), volume fractions and material densities are not constrained to their values in a reference configuration, and the effects of macroscopic viscosity and temperature gradients are included. However, under conditions (i) to (vi) above and the constraint of constant volume fractions and material densities, Eqs. (21.1)–(21.3) are transformed into coupled partial differential equations (Eqs. (27)–(29)) that are in agreement with the model of Berryman et al. [26]. Similarly, the Biot model [18,19] of momentum balance in a porous medium permeated by a single fluid can be derived from Eqs. (21.1)–(21.3).

Mass balance equations alone, however, are sufficient to derive the Biot [18,19] linear stress–strain relations, provided that a closure relation for porosity change is invoked [4,5]. The latter need only be a linear relation between porosity change and the dilatations of solid and fluid. Our results also lead to the Biot expression [19] for the linearized increment of fluid content.

Applying a similarity transformation, we have decoupled the Biot model [18,19] of linear momentum balance (Eqs. (54.1) and (54.2)) into a telegraph equation and a wave equation whose respective dependent variables are two different linear combinations of the dilatations of the solid and the fluid (or equivalently, two different linear combinations of fluid pressure and total dilatational stress). Decoupling requires a certain relation between the elastic and dynamic constitutive coefficients (Eq. (59)). Thus, a number of phenomena not yet included in current hydrologic models of subsurface immiscible fluid flow [3,8,11,13,14,22–24], e.g., pore pressure changes induced by accelerations of the solid matrix, "porosity diffusion" [49], and permeability variations caused by time-varying pore pressure, are implied by our equations of momentum balance.

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Appendix A. Functional dependence of the material derivatives of the Helmholtz potential A_1, A_2, and A_s on $\vec{a}_{1s}$ and $\vec{a}_{2s}$

The functional dependence of the material derivatives of the Helmholtz potential A_1, A_2, and A_s on $\vec{a}_{1s}$ and $\vec{a}_{2s}$ can be expressed:

$$\frac{D^1 A_1}{Dt} = \frac{\partial A_1}{\partial a^i_{1s}} \frac{D^1 a^i_{1s}}{Dt} + \frac{\partial A_1}{\partial a^i_{2s}} \frac{D^1 a^i_{2s}}{Dt}, \tag{A.1.1}$$

$$\frac{D^2 A_2}{Dt} = \frac{\partial A_2}{\partial a^i_{1s}} \frac{D^2 a^i_{1s}}{Dt} + \frac{\partial A_2}{\partial a^i_{2s}} \frac{D^2 a^i_{2s}}{Dt}, \tag{A.1.2}$$

$$\frac{D^s A_s}{Dt} = \frac{\partial A_s}{\partial a^i_{1s}} \frac{D^s a^i_{1s}}{Dt} + \frac{\partial A_s}{\partial a^i_{2s}} \frac{D^s a^i_{2s}}{Dt}. \tag{A.1.3}$$

The operator of material differentiation with respect to the ξ-phase can be transformed with respect to the solid phase through the identity [1]:

$$\frac{D^\beta a^i_{\xi s}}{Dt} = \frac{D^s a^i_{\xi s}}{Dt} + v^j_{\beta s} \frac{\partial a^i_{\xi s}}{\partial x_j} \quad (\beta = 1,2;\ \xi = 1,2), \tag{A.2}$$

where $a^i_{\xi s}$ and $v^j_{\xi s}$ are the cartesian coordinates of $\vec{a}_{\xi s}$ and $\vec{v}_{\xi s}$, respectively; $\vec{v}_{\xi s} = \vec{v}_\xi - \vec{v}_s$ is the velocity of the ξ-phase relative to that of the solid, and $\vec{a}_{\xi s} = \vec{a}_\xi - \vec{a}_s$ is the acceleration of the ξ-phase relative to that of the solid.

In accordance with Eq. (A.2), Eqs. (A.1.1) and (A.1.2) now become:

$$\frac{D^1 A_1}{Dt} = \frac{\partial A_1}{\partial a^i_{1s}} \frac{D^s a^i_{1s}}{Dt} + \frac{\partial A_1}{\partial a^i_{1s}} v^j_{1s} \frac{\partial a^i_{1s}}{\partial x^j} + \frac{\partial A_1}{\partial a^i_{2s}} \frac{D^s a^i_{2s}}{Dt} + \frac{\partial A_1}{\partial a^i_{2s}} v^j_{1s} \frac{\partial a^i_{2s}}{\partial x^j}, \tag{A.3.1}$$

$$\frac{D^2 A_2}{Dt} = \frac{\partial A_2}{\partial a^i_{1s}} \frac{D^s a^i_{1s}}{Dt} + \frac{\partial A_2}{\partial a^i_{1s}} v^j_{2s} \frac{\partial a^i_{1s}}{\partial x^j} + \frac{\partial A_2}{\partial a^i_{2s}} \frac{D^s a^i_{2s}}{Dt} + \frac{\partial A_2}{\partial a^i_{2s}} v^j_{2s} \frac{\partial a^i_{2s}}{\partial x^j}. \tag{A.3.2}$$

Given Eq. (7), we extend the gradient of the velocity to the gradient of the relative velocity:

$$\frac{\partial v^i_{\xi s}}{\partial x^j} = d^{ij}_\xi - d^{ij}_s + \varpi^{ij}_\xi - \varpi^{ij}_s, \tag{A.4}$$

where d^{ij}_ξ, d^{ij}_s, ϖ^{ij}_ξ, and ϖ^{ij}_s are the elements of $\overline{\overline{d}}_\alpha$ and $\overline{\overline{\varpi}}_\alpha$. It follows that the gradient of the relative acceleration can be formulated as:

$$\frac{\partial a^i_{\xi s}}{\partial x^j} = \frac{D^s}{Dt}\left(\frac{\partial v^i_{\xi s}}{\partial x^j}\right) = \frac{D^s d^{ij}_\xi}{Dt} - \frac{D^s d^{ij}_s}{Dt} + \frac{D^s \varpi^{ij}_\xi}{Dt} - \frac{D^s \varpi^{ij}_s}{Dt}. \tag{A.5}$$

Substitution of Eq. (A.5) into Eqs. (A.3.1) and (A.3.2) gives the following functional dependence of A_1, A_2, and A_s on $\vec{a}_{1s}$ and $\vec{a}_{2s}$ as restricted by the entropy inequality in Eq. (17):

$$
\begin{aligned}
&-\left(\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{1s}}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{1s}}+\frac{\theta_s\rho_s}{T_s}\frac{\partial A_s}{\partial a^i_{1s}}\right)\frac{\mathrm{D}^s a^i_{1s}}{\mathrm{D}t}\\
&-\left(\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{2s}}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{2s}}+\frac{\theta_s\rho_s}{T_s}\frac{\partial A_s}{\partial a^i_{2s}}\right)\frac{\mathrm{D}^s a^i_{2s}}{\mathrm{D}t}\\
&-\left(\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{1s}}v^j_{1s}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{1s}}v^j_{2s}\right)\frac{\mathrm{D}^s d^{ij}_1}{\mathrm{D}t}\\
&-\left(\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{2s}}v^j_{1s}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{2s}}v^j_{2s}\right)\frac{\mathrm{D}^s d^{ij}_2}{\mathrm{D}t}\\
&-\left(\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{1s}}v^j_{1s}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{1s}}v^j_{2s}\right)\frac{\mathrm{D}^s \varpi^{ij}_1}{\mathrm{D}t}\\
&-\left(\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{2s}}v^j_{1s}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{2s}}v^j_{2s}\right)\frac{\mathrm{D}^s \varpi^{ij}_2}{\mathrm{D}t}\\
&+\left(\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{1s}}v^j_{1s}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{1s}}v^j_{2s}+\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{2s}}v^j_{1s}\right.\\
&\left.+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{2s}}v^j_{2s}\right)\frac{\mathrm{D}^s d^{ij}_s}{\mathrm{D}t}+\left(\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{1s}}v^j_{1s}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{1s}}v^j_{2s}\right.\\
&\left.+\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{2s}}v^j_{1s}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{2s}}v^j_{2s}\right)\frac{\mathrm{D}^s \varpi^{ij}_s}{\mathrm{D}t}\geqslant 0. \qquad (A.6)
\end{aligned}
$$

The implication of this equation is that the entropy inequality must depend on $\mathrm{D}^s a^i_{1s}/\mathrm{D}t$, $\mathrm{D}^s a^i_{2s}/\mathrm{D}t$, $\mathrm{D}^s d^{ij}_1/\mathrm{D}t$, $\mathrm{D}^s d^{ij}_2/\mathrm{D}t$, $\mathrm{D}^s \varpi^{ij}_1/\mathrm{D}t$, $\mathrm{D}^s \varpi^{ij}_2/\mathrm{D}t$, $\mathrm{D}^s d^{ij}_s/\mathrm{D}t$, and $\mathrm{D}^s \varpi^{ij}_s/\mathrm{D}t$. However, these material derivatives are not chosen as independent variables to construct constitutive equations, so the necessary and sufficient condition to meet the requirement of nonnegative entropy production rate is to force the coefficients of the material derivatives to vanish. This imposes constraints on the constitutive equations:

$$\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{1s}}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{1s}}+\frac{\theta_s\rho_s}{T_s}\frac{\partial A_s}{\partial a^i_{1s}}=0, \qquad (A.7.1)$$

$$\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{2s}}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{2s}}+\frac{\theta_s\rho_s}{T_s}\frac{\partial A_s}{\partial a^i_{2s}}=0, \qquad (A.7.2)$$

$$\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{1s}}v^j_{1s}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{1s}}v^j_{2s}=0, \qquad (A.7.3)$$

$$\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{2s}}v^j_{1s}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{2s}}v^j_{2s}=0, \qquad (A.7.4)$$

$$
\begin{aligned}
&\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{1s}}v^j_{1s}+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{1s}}v^j_{2s}+\frac{\theta_1\rho_1}{T_1}\frac{\partial A_1}{\partial a^i_{2s}}v^j_{1s}\\
&\quad+\frac{\theta_2\rho_2}{T_2}\frac{\partial A_2}{\partial a^i_{2s}}v^j_{2s}=0. \qquad (A.7.5)
\end{aligned}
$$

Eq. (A.7.5) is redundant to the combination of Eqs. (A.7.3) and (A.7.4).

Appendix B. Diagonalization of $K^{-1}F$ and $K^{-1}L$

Based on the definitions of the matrices F, K, L, Z in Section 4.2, one can derive explicit expressions for $ZK^{-1}FZ^{-1}$ and $ZK^{-1}LZ^{-1}$:

$$ZK^{-1}FZ^{-1}=(PR-Q^2)^{-1}\begin{bmatrix}P+2Q+R & 0\\ R+Q-D(P+Q) & 0\end{bmatrix}, \qquad (B.1.1)$$

$$ZK^{-1}LZ^{-1}=(PR-Q^2)^{-1}(1+D)^{-1}\begin{bmatrix}a1 & a2\\ a3 & a4\end{bmatrix}, \qquad (B.1.2)$$

where

$$a1=(R+Q)(\rho_{11}D-\rho_{12})-(P+Q)(\rho_{12}D-\rho_{22}), \qquad (B.2.1)$$

$$a2=(R+Q)(\rho_{11}+\rho_{12})-(P+Q)(\rho_{12}+\rho_{22}), \qquad (B.2.2)$$

$$a3=(R-QD)(\rho_{11}D-\rho_{12})+(PD-Q)(\rho_{12}D-\rho_{22}), \qquad (B.2.3)$$

$$a4=(R-QD)(\rho_{11}+\rho_{12})+(PD-Q)(\rho_{12}+\rho_{22}). \qquad (B.2.4)$$

In order to make $ZK^{-1}FZ^{-1}$ diagonal, it is necessary to have $R+Q-D(P+Q)=0$, i.e.,

$$D=\frac{R+Q}{P+Q}. \qquad (B.3)$$

For $ZK^{-1}LZ^{-1}$ to be diagonal, we must make $a2=0$ and $a3=0$. The condition $a2=0$ leads to:

$$\frac{\rho_{12}+\rho_{22}}{\rho_{11}+\rho_{12}}=\frac{R+Q}{P+Q}=D. \qquad (B.4)$$

This identity produces two relations: $R-QD=PD-Q$ and $\rho_{12}-\rho_{11}D=\rho_{12}D-\rho_{22}$. Combination of these two relations then guarantees $a3=0$.

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