

A Thermodynamic Model of Compressible Porous Materials with the Balance Equation of Porosity

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Summary

The paper is devoted to the construction of a thermodynamic continuous model of porous media with changing porosity. It is shown that these changes are described by a balance equation. The flux in this equation is connected with a relative motion of components and the source describes a spontaneous relaxation of microstructure. Deformations of the skeleton can be arbitrary and a consistent Lagrangian description of motion of all components is applied.

1. Introduction

The subject of the continuum theories of compressible porous materials belongs to the class of theories of multicomponent systems. Simultaneously porous materials are characterized by a geometrical microstructure of almost macroscopic dimensions. This means that the continuous description must be applied to such systems with a certain caution. We assume that the microstructure of systems under considerations admits the simplest continuous description by a single scalar field called the **porosity**. This quantity is equivalent to a volume fraction of skeleton which is equal to the unity minus the porosity. Physically the porosity considered in this paper is identical with an effective porosity referring solely to these pores of material which are interconnected by channels.

Another microstructural variable which may have an equally important bearing in the macroscopic description (e.g. for heterogeneous chemical reactions) is the microscopic surface of pores and channels. We return to this subject in a forthcoming paper.

The main aim of this paper is to develop thermodynamic foundations of the continuous model of two-component porous material in which the porosity is a non-trivial thermodynamic field. This is different from many other papers on this subject in which the porosity was considered to be driven by other fields of the model. For instance, M. A. BIOT [1] proposed an equation for porosity in which the flux was identified with the relative velocity and the source (relaxation) was absent.

The non-triviality of porosity field means that the porosity possesses its own field equation and it may relax spontaneously to the thermodynamic equilibrium. This yields an additional dissipation. Such an equation is derived in the section 3 of the paper from

semimicroscopic geometrical considerations. The considerations are semimicroscopic because the image analysis used in this derivation is based on the continuity assumption for the microstructure. This means in turn that the microstructure cannot be so fine that the molecular effects would influence the considerations. The balance equation of porosity has been already anticipated in my earlier papers and it has been shown that many measurable effects connected, for instance, with the propagation of sound waves are indeed predicted through this equation. Most important of them is the attenuation of acceleration waves. Similar results follow from the model of R.M.BOWEN who proposed an evolution equation for porosity [2] i.e. a balance equation without flux. Let us mention that an extension of the status of porosity to a field can be achieved by another type of balance equation following from the so-called balance of equilibrated forces introduced by M. A. GOODMAN and S. C. COWIN [3]. In contrast to a rather unclear energetical justification of the latter approach the balance equation of porosity of the present work is motivated solely by semimicroscopic changes of geometry of the skeleton.

In the section 4 we present the fields of the new model describing isothermal processes in two-component porous materials. It is assumed that the skeleton is elastic for porosity equal to zero and the fluid is ideal for porosity equal to one. In the formulation of field equations we rely on the Lagrangian description which has been introduced in my paper [4].

Sections 5,6 and 7 contain a thermodynamic analysis of constitutive relations. It is shown that the balance equation of porosity, in contrast to the Bowen's evolution equation, yields essential couplings between components.

Section 8 has a special character. It contains a very simple example illustrating the opinion that continuum models of porous materials such as the one presented in the paper do not give usually any direct information on the real „true“ semimicroscopic quantities. Therefore the questions frequently asked what in the continuous model describes, for instance, the true pore water pressure are meaningless. In this section as well as in section 9 we indicate possibilities of the reasonable identification of fields of continuous models and some connections with real macroscopic measurements. However the latter problem is not discussed in any details in the present work. From the mathematical point of view these inverse problems are usually ill-posed.

2. Fields in the multicomponent continuous models of porous materials

Continuous models of porous materials must account for semimicroscopic geometrical structure of components (*immiscible mixtures*) and different kinematics of components (*diffusion*). Particularly the coupling of those problems yields many questions and misunderstandings. The two most important points which must be cleared are the number and character of additional macroscopic fields describing the semimicroscopic geometry and the interpretation of these fields in terms of real microstructural quantities. Some hints may be expected from statistical models. These are not yet developed to such an extent as in the case of statistical theories of gases but they are sufficient to indicate the structure of the macroscopic model. An excellent presentation of such a procedure for the porous materials can be found in the classical book of J. BEAR [5] (in particular, sections 4.5.-4.8.).

In this work we proceed along the classical line of the argument adopted already by C. TRUESDELL [6] in the theory of mixtures of miscible components. Two essential differences arise due to the presence of a compact solid skeleton as one of the components. On the one hand side this yields the necessity to describe the deformation of solid component in the same way as it is done in the continuum mechanics of solids - by means of a **deformation tensor** and not only by changes of the mass density. On the other hand the microscopic geometrical properties induced by the skeleton should be reflected by an appropriate macroscopic field. In the simplest case of isotropic microstructure it is assumed that this is the scalar field of **porosity**.

The above remarks indicate that the skeleton of the porous material has a special character among components. It forms a *deformable confinement* for the motion of fluid components. In the case of large deformations it is customary to use then the Lagrangian description for this component. All other components are assumed to be fluids and their description is similar to this of the classical mixture theory of miscible components. By means of a simple transformation we can also use the common Lagrangian description for all components, induced by the motion of the skeleton. This has been proposed for a two-component system in my work [4,7] and extended on the multicomponent systems in [9,10].

We choose the configuration $\mathcal{B} \subset \mathcal{R}^3$ of the skeleton at an initial instant of time t_0 as the reference configuration for the Lagrangian description. Then the motion of skeleton is described by the following function

$$\mathbf{x} = \boldsymbol{\chi}^s(\mathbf{X}, t), \quad \mathbf{x} \in \boldsymbol{\chi}^s(\mathcal{B}, t) \subset \mathcal{R}^3, \quad \mathbf{X} \in \mathcal{B}, \quad t \in \mathcal{T} \equiv \langle t_0, t_f \rangle, \quad (2.1)$$

where the point \mathbf{x} is the current position of the material point \mathbf{X} of skeleton. The instant of time t_f can lie in infinity. The function of motion $\boldsymbol{\chi}^s$ is assumed to admit the existence of the derivatives

$$\mathbf{x}'^s = \frac{\partial \boldsymbol{\chi}^s}{\partial t}(\mathbf{X}, t), \quad \mathbf{F}^s = \text{Grad} \boldsymbol{\chi}^s(\mathbf{X}, t), \quad J^s \equiv \det \mathbf{F}^s > 0, \quad (2.2)$$

which define the velocity field \mathbf{x}'^s and the deformation gradient \mathbf{F}^s of skeleton. Obviously $\mathbf{F}^s = \mathbf{1}$ in the reference configuration \mathcal{B} .

The above description of motion of skeleton is, certainly, macroscopic - as the whole continuous model of the porous materials. This means that we cannot transform directly these relations onto the semimicroscopic level of observation. For example macroscopic changes of the volume of skeleton, described by the changes of the determinant J^s may appear in the case of lack of any volume changes of the real skeleton, and *vice versa*, the changes of the true volume of skeleton on the semimicroscopic level may be compensated by the changes of porosity and macroscopic changes of J^s may not appear at all. These differences between the micro- and macrodescriptions yield, of course, the additional couplings between the components. For instance the compensation of changes of the microscopic volume by the changes of porosity must induce some processes in fluid components.

Further in this work we consider a two-component model of the porous material. However, in order to clear the notion of the skeleton as the confinement of motion of fluid components we consider in this section the kinematics of an arbitrary number of these components, i.e. we assume the porous material to consist of A fluid components. In the classical theory of mixtures the description of their motions is given by the Eulerian velocity fields

$$\mathbf{v}^\alpha = \mathbf{v}^\alpha(\mathbf{x}, t), \quad \alpha = 1, \dots, A, \quad \mathbf{x} \in \chi^S(\mathcal{Z}, t). \quad (2.3)$$

We transform these fields into the material description of the skeleton

$$\mathbf{x}'^\alpha = \mathbf{v}^\alpha(\chi^S(\mathbf{X}, t), t) = \mathbf{x}'^\alpha(\mathbf{X}, t), \quad \alpha = 1, \dots, A. \quad (2.4)$$

These fields can serve already the purpose of consistent description of all components on the common domain \mathcal{Z} . However the formulation of partial balance equations requires the transformation of these fields onto the reference configuration of skeleton. This problem has been discussed in my papers [4,7,8]. The pull-back transformation by means of the deformation gradient of skeleton yields the following velocities of images of fluid components across the reference domain \mathcal{Z}

$$\mathbf{X}'^\alpha(\mathbf{X}, t) = \mathbf{F}^{S-1}(\mathbf{x}'^\alpha - \mathbf{x}'^S), \quad \alpha = 1, \dots, A. \quad (2.5)$$

We call these fields the **Lagrangian velocity fields** of fluid components. The Lagrangian velocities replace the usual relative velocities of mixture theory. They describe the relative motion of fluid components with respect to the skeleton in contrast to those in the classical mixture theory of miscible components in which the relative motion is described with respect to local centers of gravity whose velocity field is called the barycentric velocity (see: [11]).

We are now in position to define the **thermomechanical process** of our model. It is the set of mappings

$$\forall \mathbf{X} \in \mathcal{Z}, t \in \mathcal{T}: (\mathbf{X}, t) \mapsto (\rho^1, \dots, \rho^A, n, \chi^S(\mathbf{X}, t), \mathbf{X}'^1, \dots, \mathbf{X}'^A, \theta^S, \theta^1, \dots, \theta^A), \quad (2.6)$$

provided it is a solution of an appropriate initial-boundary value problem for the set of field equations. In the above expression the quantities ρ^α , $\alpha=1, \dots, A$, describe the macroscopic partial mass densities of fluid components related to the unit reference volume and $\theta^S, \theta^1, \dots, \theta^A$ are partial temperatures of components. The partial mass density of skeleton ρ^S does not appear among those quantities because it is a constant under the assumption of the uniformity of skeleton and in processes without mass exchange between components.

Field equations for the fields (2.6) follow from the partial balance equations. We shall not discuss here their structure (see: [8,9,10]). In the Lagrangian description they have the following form

- the partial mass balance equations for the fluid components

$$\frac{\partial \rho^\alpha}{\partial t} + \text{Div}(\rho^\alpha \mathbf{X}'^\alpha) = 0, \quad \alpha = 1, \dots, A, \quad (2.7)$$

- the momentum balance equation of the skeleton

$$\rho^S \frac{\partial \mathbf{x}'^S}{\partial t} = \text{Div} \mathbf{P}^S + \rho^S \mathbf{b}^S + \hat{\mathbf{p}}^S, \quad (2.8)$$

- the momentum balance equations for the fluid components

$$\rho^\alpha \left\{ \frac{\partial \mathbf{x}'^\alpha}{\partial t} + \text{Grad} \mathbf{x}'^\alpha \mathbf{X}'^\alpha \right\} = \text{Div} \mathbf{P}^\alpha + \rho^\alpha \mathbf{b}^\alpha + \hat{\mathbf{p}}^\alpha, \quad \alpha = 1, \dots, A, \quad (2.9)$$

- the energy balance equation of the skeleton

$$\frac{\partial \rho^S \left[\varepsilon^S + \frac{1}{2} \mathbf{x}'^{S2} \right]}{\partial t} + \text{Div} \mathbf{Q}^S = \text{Div}(\mathbf{P}^{ST} \mathbf{x}'^S) + \rho^S \mathbf{b}^S \cdot \mathbf{x}'^S + \rho^S r^S + \hat{\varepsilon}^S, \quad (2.10)$$

- the energy balance equations for the fluid components

$$\begin{aligned} \frac{\partial \rho^\alpha \left[\varepsilon^\alpha + \frac{1}{2} \mathbf{x}'^{\alpha 2} \right]}{\partial t} + \text{Div} \left\{ \rho^\alpha \left[\varepsilon^\alpha + \frac{1}{2} \mathbf{x}'^{\alpha 2} \right] \mathbf{X}'^\alpha + \mathbf{Q}^\alpha \right\} = \text{Div}(\mathbf{P}^{\alpha T} \mathbf{x}'^\alpha) + \\ + \rho^\alpha \mathbf{b}^\alpha \cdot \mathbf{x}'^\alpha + \rho^\alpha r^\alpha + \hat{\varepsilon}^\alpha, \quad \alpha = 1, \dots, A. \end{aligned} \quad (2.11)$$

In the above equations we use the following notation. The tensor \mathbf{P}^S denotes the Piola-Kirchhoff partial stress tensor in skeleton with respect to the configuration \mathcal{Z} , \mathbf{b}^S is the mass force for this component and $\hat{\mathbf{p}}^S$ is the intensity of momentum source in the skeleton relative to the reference configuration. In the whole work we use inertial reference frames which means that \mathbf{b}^S contains solely the action of the external world on the skeleton. The tensors \mathbf{P}^α denote the partial Piola-Kirchhoff stress tensors of the fluid components α with respect to the configuration \mathcal{Z} , \mathbf{b}^α are the partial mass forces for these components and $\hat{\mathbf{p}}^\alpha$ are the intensities of the momentum sources for the α -components per unit volume of the reference configuration \mathcal{Z} . The scalar ε^S is the specific partial internal energy of skeleton per unit mass, \mathbf{Q}^S is the partial heat flux in the material description, r^S is the energy radiation per unit mass and $\hat{\varepsilon}^S$ is the intensity of energy source in the skeleton per unit reference volume. Finally ε^α , \mathbf{Q}^α , r^α and $\hat{\varepsilon}^\alpha$ denote the specific partial internal energy, the partial heat flux, the energy radiation per unit mass and the energy source per unit reference volume of \mathcal{Z} all for the α -component, respectively. The source terms must fulfil the conditions

$$\hat{\mathbf{p}}^S + \sum_{\alpha=1}^A \hat{\mathbf{p}}^\alpha = 0, \quad \hat{\boldsymbol{\varepsilon}}^S + \sum_{\alpha=1}^A \hat{\boldsymbol{\varepsilon}}^\alpha = 0, \quad (2.12)$$

which follow from the bulk conservation laws of momentum and energy.

As usual we have to add the constitutive relations in order to turn these equations into the field equations for quantities (2.6). This problem is discussed further in this work for a certain limited class of porous materials.

However, even if those constitutive relations were given, we are still missing one equation. This is due to the additional field - porosity - describing the microstructure of porous medium. In the next section we present a solution of this problem which is based on a heuristic argument guiding this work that the skeleton is the confinement for the motion of fluid components.

3. Geometrical properties of the microstructure

The basis for the macroscopic description of the semimicroscopic geometry is the assumption that solely the volume contribution of pores to the typical microscopic domain bears a hand in the distinction between the miscible and immiscible mixtures. This is in the spirit of confinement of the space of motion for the fluid components by the skeleton. We consider the true configurations of the real porous material to be embedded in the three-dimensional Euclidean space. In order to describe the local changes of these configurations we choose a *control domain* \mathfrak{m}_X of the microstructure (representative volume element) which we attach to the macroscopic point \mathbf{X} of the reference configuration \mathcal{B} . Let us denote the position vector of an arbitrary point of this domain by \mathbf{y} . Then the control domain in any other point $\mathbf{X}' \in \mathcal{B}$ is assumed to be defined by the following shift

$$\mathfrak{m}_{X'} = \left\{ \mathbf{y} \mid \mathbf{y} + (\boldsymbol{\chi}^S(\mathbf{X}, t) - \boldsymbol{\chi}^S(\mathbf{X}', t)) \in \mathfrak{m}_X \right\}. \quad (3.1)$$

Consequently all control domains can be considered to be isomorphic three-dimensional vector spaces.

In order to describe the microstructure in the Lagrangian way we transform the above set by means of the inverse function of motion of the skeleton. We define

$$\forall \mathbf{X} \in \mathcal{B}: \mathcal{M}_X = \left\{ \mathbf{Y} \mid \mathbf{Y} - \mathbf{X} = \varepsilon \mathbf{Z}, \varepsilon \mathbf{Z} \equiv \mathbf{y} - \boldsymbol{\chi}^S(\mathbf{X}, t), \mathbf{y} \in \mathfrak{m}_X \right\}. \quad (3.2)$$

The positive parameter ε is of the same order of magnitude as the ratio of the characteristic linear dimension of the microstructure to a characteristic macroscopic length. It is assumed to be much smaller than the unity. In such a case the lengths of vectors \mathbf{X} and \mathbf{Z} is of the same order of magnitude. This scaling allows to introduce a certain perturbation procedure which we use in the sequel. Further we shall always use the notion of the control domain in its Lagrangian description (3.2).

We show schematically the image \mathcal{M}_X of such a control domain in the Figure 1. It is the part of the space limited by the boundary $\partial \mathcal{M}_X$ of the magnifying glass.

We proceed to discuss the distribution of the real skeleton within the control domain. It is assumed that geometrical properties of real skeleton can be described in the same way as it is done in continuum mechanics.

We are interested in the volume fraction of control domain which is occupied by the skeleton. This is obviously given by the following relation

$$1 - n(\mathbf{X}, t) = \frac{1}{V_c} \int_{\mathcal{M}_X} \mathcal{H}(\mathbf{Y}, t) dV, \quad V_c \equiv \int_{\mathcal{M}_X} dV = \text{const.}, \quad \mathcal{H}(\mathbf{Y}, t) = \begin{cases} 1 & \text{for } \mathbf{Y} \in \mathcal{M}_X^S, \\ 0 & \text{for } \mathbf{Y} \notin \mathcal{M}_X^S, \end{cases} \quad (3.3)$$

where \mathcal{M}_X^S is the part of the control domain occupied by the skeleton. The scalar quantity n shall be, of course, identified with the porosity.

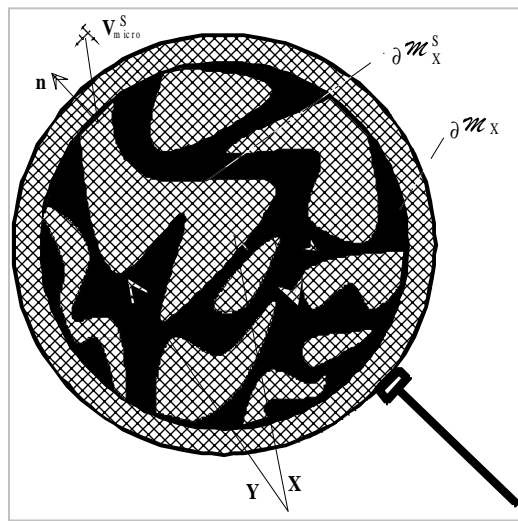


Figure 1: *The geometrical notions within the microstructure \mathcal{M}_X at the point \mathbf{X} of the porous body*

The parts with the pattern correspond to the microscopical domain \mathcal{M}_X^S of the skeleton

We proceed to investigate time changes of the porosity. The most important observation is that the time changes of the image \mathcal{M}_X^S are determined solely by the velocity of points of the boundary $\partial \mathcal{M}_X^S$. The relative velocity of these points with respect to a chosen macroscopic point \mathbf{X} , i.e. the difference of the microscopic velocity and the velocity $\mathbf{x}'^S(\mathbf{X}, t)$, is denoted by $\mathbf{V}_{\text{micro}}^S$. Let us notice that this is a true velocity of the **material points of the true skeleton** because we have made the continuity assumption for the semimicroscopic level of observation. This means that material points of the skeleton which happen to be located on the boundary $\partial \mathcal{M}_X^S$ at the initial instant of time must remain boundary points forever. Consequently if we were able to solve a full boundary value problems on the semimicroscopic level the velocity $\mathbf{V}_{\text{micro}}^S$

would be determined and so would be also changes of the image of \mathcal{M}_X^S . In a macroscopic continuum approach such problems cannot be formulated. Rather they are replaced by macroscopic constitutive assumptions which we discuss in the next section.

Bearing this in mind and denoting by \mathbf{n} the unit outward normal vector to the boundary surface $\partial\mathcal{M}_X^S$ we can write time changes of the porosity n in the following way

$$\frac{\partial n}{\partial t} \equiv -\frac{1}{V_c} \frac{\partial}{\partial t} \int_{\mathcal{M}_X} \mathcal{F}(\mathbf{Y}, t) dV = -\frac{1}{V_c} \int_{\partial\mathcal{M}_X^S} \mathbf{V}_{\text{micro}}^S \cdot \mathbf{n} dA. \quad (3.4)$$

The surface integral on the right-hand side can be approximately written as the integral over the closed surface. Namely

$$\int_{\partial\mathcal{M}_X^S} dA = \oint_{A_{\text{micro}}} dA - \int_{\mathcal{M}_X^S \cap \partial\mathcal{M}_X} dA, \quad (3.5)$$

where

$$A_{\text{micro}} \equiv \partial\mathcal{M}_X^S \cup (\mathcal{M}_X^S \cap \partial\mathcal{M}_X). \quad (3.6)$$

Certainly the surface $\mathcal{M}_X^S \cap \partial\mathcal{M}_X$ contains the points of the true skeleton which instantaneously coincide with the boundary of the control domain \mathcal{M}_X . We assume that an area of this surface, i.e. the second integral on the right-hand side of (3.5) is much smaller than the area of $\partial\mathcal{M}_X^S$. Physically it means that microscopic channels have a small diameter compared with the length within the control domain. There is also a positive contribution of a tortuosity to this assumption.

Simultaneously the velocity of points on the surface $\mathcal{M}_X^S \cap \partial\mathcal{M}_X$ is of the same order of magnitude as $\mathbf{V}_{\text{micro}}^S$ because this is also the velocity of material points of the true skeleton. Hence

$$\begin{aligned} \frac{\partial n}{\partial t} &\approx -\frac{1}{V_c} \oint_{A_{\text{micro}}} \mathbf{V}_{\text{micro}}^S \cdot \mathbf{n} dA = -\frac{1}{V_c} \int_{\mathcal{M}_X^S} \text{div}_Y \mathbf{V}_{\text{micro}}^S dV = \\ &= -\frac{1}{V_c} \int_{\mathcal{M}_X} \mathcal{F}(\mathbf{Y}, t) \text{div}_Y \mathbf{V}_{\text{micro}}^S dV. \end{aligned} \quad (3.7)$$

The divergence in the above formula is calculated with respect to the variable \mathbf{Y} . We can write the right-hand side in the following form

$$\begin{aligned} -\frac{1}{V_c} \int_{\mathcal{M}_X} \mathcal{F}(\mathbf{Y}, t) \text{div}_Y \mathbf{V}_{\text{micro}}^S dV &= -\frac{1}{V_c} \int_{\mathcal{M}_X} \text{div}_Y [\mathcal{F}(\mathbf{Y}, t) \mathbf{V}_{\text{micro}}^S] dV + \\ &+ \frac{1}{V_c} \int_{\mathcal{M}_X} \mathbf{V}_{\text{micro}}^S \cdot \text{grad}_Y \mathcal{F}(\mathbf{Y}, t) dV. \end{aligned} \quad (3.8)$$

The gradient in (3.8) describes, of course, the Dirac δ -distribution with the support identical with $\partial \mathcal{M}_X^S$. It reduces to $\mathbf{n}\delta(z)$ in the local coordinates with the variable z measuring the distance from this surface. Consequently

$$\frac{\partial \mathbf{n}}{\partial t} \approx -\frac{1}{V_c} \int_{\mathcal{M}_X} \operatorname{div}_Y [\mathcal{N}(\mathbf{Y}, t) \mathbf{V}_{\text{micro}}^S] dV + \frac{1}{V_c} \oint_{A_{\text{micro}}} \mathbf{V}_{\text{micro}}^S \cdot \mathbf{n} dA. \quad (3.9)$$

Substitution in (3.7) yields finally

$$\frac{\partial \mathbf{n}}{\partial t} = -\frac{1}{2} \frac{1}{V_c} \int_{\mathcal{M}_X} \operatorname{div}_Y [\mathcal{N}(\mathbf{Y}, t) \mathbf{V}_{\text{micro}}^S] dV. \quad (3.10)$$

The velocity field in the square brackets is defined on the whole control domain - in contrast to the field $\mathbf{V}_{\text{micro}}^S$. It depends on the location of the point \mathbf{Z} within the control domain as well as on the choice of the point $\mathbf{X} \in \mathcal{E}$. This dependence can be used to approximate semimicroscopic properties because the microstructure introduces the small parameter ε . We proceed to describe this procedure.

It is clear from the above considerations that the existence of microstructure yields two different types of spatial changes of an arbitrary microscopic function of the variable \mathbf{Y} . One type is connected with changes of the variable \mathbf{Y} whose order of magnitude related to the characteristic macroscopic length is much larger than the parameter ε . These are macroscopic changes. Another type is connected with the changes of the variable \mathbf{Y} whose order of magnitude estimated in the same way is much smaller than the parameter ε . These are microstructural changes. Consequently we can consider two different *scales of spatial changes*. For this reason we change the interpretation of variables \mathbf{X} and \mathbf{Z} . Namely we consider them as the two **independent** variables. The function of these two variables which we have to consider is a vector function $\mathbf{V}(\mathbf{X}, \mathbf{Z}, t)$. This function is called the **extension** of the microscopic velocity iff

$$\mathbf{V}(\mathbf{X} = \mathbf{Y}, \mathbf{Z} = \mathbf{Y}/\varepsilon, t) = \mathcal{N}(\mathbf{Y}, t) \mathbf{V}_{\text{micro}}^S(\mathbf{Y}, t). \quad (3.11)$$

This type of **multiscaling** is well-known in the kinetic theory of gases where it is used for different time scales. Some details of the general procedure can be found in the paper of G. SANDRI [12]. For the multiphase flows it was also recently used by ZHANGXIN CHEN [13]. In this paper one can find as well a very extensive presentation of literature on related topics within theories of multicomponent systems.

Now the extension \mathbf{V} is assumed to be approximated by a truncated regular perturbation series

$$\mathbf{V}(\mathbf{X}, \mathbf{Z}, t) = \mathbf{V}_0(\mathbf{X}, \mathbf{Z}, t) + \varepsilon \mathbf{V}_1(\mathbf{X}, \mathbf{Z}, t), \quad (3.12)$$

which is assumed to satisfy the extension of the equation (3.10). Namely we have for $\mathbf{X}=\mathbf{Y}$ and $\mathbf{Z}=\mathbf{Y}/\varepsilon$

$$\operatorname{div}_Y \mathbf{V} = \operatorname{Div}(\mathbf{V}_0 + \varepsilon \mathbf{V}_1) + \frac{1}{\varepsilon} \operatorname{div}_Z(\mathbf{V}_0 + \varepsilon \mathbf{V}_1), \quad (3.13)$$

where div_Y is the divergence with respect to the variable \mathbf{Y} , Div is the divergence with respect to the variable \mathbf{X} and div_Z is the divergence with respect to the variable \mathbf{Z} . The Equation (3.10) becomes

$$\frac{\partial n}{\partial t} \approx -\frac{1}{2} \frac{1}{V_c} \operatorname{Div} \int_{\mathcal{M}_x} (\mathbf{V}_0 + \varepsilon \mathbf{V}_1) dV_Z - \frac{1}{\varepsilon} \frac{1}{2} \frac{1}{V_c} \int_{\mathcal{M}_x} \operatorname{div}_Z(\mathbf{V}_0 + \varepsilon \mathbf{V}_1) dV_Z. \quad (3.14)$$

The volume integration over the microstructure \mathcal{M}_x is, certainly, the integration with respect to the „fast“ (*fine grained*) variable \mathbf{Z} which is indicated by dV_Z .

The application of the perturbation procedure to (3.14) yields for two subsequent powers of ε

$$\begin{aligned} \varepsilon^{-1}: \quad & \int_{\mathcal{M}_x} \operatorname{div}_Z \mathbf{V}_0 dV_Z = 0, \\ \varepsilon^0: \quad & \frac{\partial n}{\partial t} = -\frac{1}{2} \frac{1}{V_c} \operatorname{Div} \int_{\mathcal{M}_x} \mathbf{V}_0 dV_Z - \frac{1}{2} \frac{1}{V_c} \int_{\mathcal{M}_x} \operatorname{div}_Z \mathbf{V}_1 dV_Z. \end{aligned} \quad (3.15)$$

Higher order terms are not reliable any more due to the form of the truncation (3.12).

The first relation (3.15) limits the dependence of \mathbf{V}_0 on the microscopic variable \mathbf{Z} .

Let us introduce the notation

$$\begin{aligned} \mathbf{J}(\mathbf{X}, t) &\equiv \frac{1}{2} \frac{1}{V_c} \int_{\mathcal{M}_x} \mathbf{V}_0 dV_Z, \\ \hat{n}(\mathbf{X}, t) &\equiv -\frac{1}{2} \frac{1}{V_c} \int_{\mathcal{M}_x} \operatorname{div}_Z \mathbf{V}_1 dV_Z. \end{aligned} \quad (3.16)$$

Then the equation (3.15) has the form

$$\frac{\partial n}{\partial t} + \operatorname{Div} \mathbf{J} = \hat{n}. \quad (3.17)$$

This is the **balance equation of porosity** which supplements our set of macroscopic field equations.

The relations (3.16) yield a simple semimicroscopic interpretation of the flux \mathbf{J} and the source \hat{n} . It is obvious that the flux \mathbf{J} is primarily connected with the bulk micromotion of microstructure through the control domain \mathcal{M}_x , i.e. it describes the in- and out-flow of the material of true skeleton to and from the domain of observation connected with the macroscopic point $\mathbf{X} \in \mathcal{E}$. Simultaneously the source \hat{n} is produced by the micromotion within the control domain, i.e. it is primarily connected with the *relaxation processes* within the pores.

Let us finally notice that the above motivation of the balance equation of porosity (3.17) is solely based on the **image analysis**. It bears no information on a constitutive

character of true components such as the distribution of true mass densities. It is not even required that the pores carry any fluid components at all. The latter is connected with the assumption that it is the solid component - the true skeleton which geometrically determines the changes of porosity.

However, the dependence on the material properties is hidden in the macroscopic constitutive form of \mathbf{J} and \hat{n} and it is solely the structure of the equation (3.17) which has the above presented geometrical motivation.

4. Constitutive assumptions for the two-component model - isothermal processes

We are now in the position to formulate the complete set of field equations for the fields (2.6). In order to expose the most important features of the new equation for porosity we shall investigate solely the simplest *two-component* model of the porous material. We assume as well all processes to be *isothermal*. Consequently the list of fields (2.6) reduces to the following one

$$\forall \mathbf{X} \in \mathcal{Z}, t \in \mathcal{T}: (\mathbf{X}, t) \mapsto (\rho^F, n, \boldsymbol{\chi}^S(\mathbf{X}, t), \mathbf{X}'^F), \quad (4.1)$$

where ρ^F denotes the mass density of the single fluid component and \mathbf{X}'^F is the Lagrangian velocity of this component.

The balance equations (2.7-11), (3.17) reduce to the following set

$$\begin{aligned} \frac{\partial \rho^F}{\partial t} + \text{Div}(\rho^F \mathbf{X}'^F) &= 0, & \frac{\partial n}{\partial t} + \text{Div} \mathbf{J} &= \hat{n}, \\ \rho^S \frac{\partial \mathbf{x}'^S}{\partial t} &= \text{Div} \mathbf{P}^S + \rho^S \mathbf{b}^S + \hat{\mathbf{p}}, & & (4.2) \\ \rho^F \left\{ \frac{\partial \mathbf{x}'^F}{\partial t} + \text{Grad}_{\mathbf{x}'^F} \mathbf{X}'^F \right\} &= \text{Div} \mathbf{P}^F + \rho^F \mathbf{b}^F - \hat{\mathbf{p}}. \end{aligned}$$

In all quantities for the fluid components in the relations (2.7), (2.9) and (2.10) the running index α has been replaced by the index F for the single fluid component. The energy balance equations do not appear at all because the processes are assumed to be isothermal. The momentum source appears without any index because we have made use of the condition (2.12)₁ for the case of two components.

As usual we have to close the set (4.2) by means of the constitutive relations. Under the condition of the *material frame indifference* (material objectivity) these relations must be chosen for the following set of the **constitutive quantities**

$$\mathcal{Z} = \mathcal{Z}(\mathbf{J}, \hat{n}, \mathbf{F}^{S-1} \mathbf{P}^S, \mathbf{F}^{S-1} \mathbf{P}^S, \mathbf{F}^{ST} \hat{\mathbf{p}}). \quad (4.3)$$

Hence we have to formulate constitutive laws not only for the usual quantities of partial stress tensors and for the source of momentum (diffusion force) but for the flux of porosity \mathbf{J} and for the source of porosity \hat{n} as well. We have mentioned this constitutive problem deriving the balance equation for porosity in section 3.

In this paper we consider the class of materials for which the above constitutive quantities are functions of the following **constitutive variables**

$$\mathcal{C} = \{\rho^F, n, \mathbf{C}^S, \mathbf{X}'^F\}, \quad \mathbf{C}^S \equiv \mathbf{F}^{ST}\mathbf{F}^S, \quad (4.4)$$

where \mathbf{C}^S denotes, of course, the right Cauchy-Green deformation tensor of the skeleton.

The **constitutive relations** are then assumed to have the form

$$\mathfrak{z} = \mathfrak{z}(\mathcal{C}), \quad \mathcal{C} \in \mathfrak{R}^{11}, \quad \mathfrak{z} \in \mathfrak{R}^{19}, \quad (4.5)$$

and all these functions are assumed to be twice continuously differentiable with respect to all arguments.

If these relations were given the set (4.2) of two scalar and two vector balance equations would be the set of field equations for two scalar and two vector fields (4.1). This is usually not the case and for this reason we check the thermodynamic admissibility of formally chosen constitutive relations (4.5). This is the subject of the next three sections of this work.

The skeleton of the porous material described by the constitutive variables (4.4) is said to be **elastic** and the fluid of this material is said to be **ideal** (inviscid). This, certainly, does not mean that processes in such a porous material are reversible. As we see further there are two sources of dissipation for the material described by the above constitutive relations - the diffusion with its force of relative motion $\hat{\mathbf{p}}$ and the pore relaxation with the source \hat{n} . The terminology is solely indicating that if the porosity was identically equal to zero the skeleton would behave as a one component nonlinear elastic solid and if the porosity was identically equal to one the fluid component would behave as a one component ideal fluid.

The field equation for porosity which follows from the balance equation (4.2)₂ by means of the substitution of constitutive relations differs from the evolution equation proposed by R.M.BOWEN [2], as we have already mentioned, by to the presence of the flux \mathbf{J} . Both approaches yield spontaneous pore relaxation processes which have been indicated in section 3. We shall discuss some of their properties further in this work. Consequently both models, the present one and this of Bowen, give the porosity the status of the non-trivial thermodynamic variable. However the Bowen's model yields the behaviour typical for internal variables - namely they cannot be controlled by boundary (external) conditions. This is not the case in the present model. In some particular cases these additional boundary conditions may not be needed. This is, for instance, the case for processes yielding small deviations from the thermodynamic equilibrium. However, in the general case such an additional control appears in this new model. This point has been discussed in a general framework by G. A. MAUGIN and W. MUSCHIK [14]. In section 5.2. of this work they point out that the control of internal variables through "external forces" may be limited to some small boundary layers. Beyond these regions, the additional field may behave as a "true" internal variable. We refer to this paper for further details concerning the location of porosity field equation among possible ways of describing the deviation from the thermodynamic equilibrium by means of internal variables. In terms of the paper [14] the porosity of the present model would fulfil the evolution-diffusion equation. It should be pointed out that the

presence of the flux of porosity \mathbf{J} (“diffusivity” of the internal variable) yields in any case, boundary conditions or not, the essential coupling effects between components. Further in this work we discuss the latter problem (coupling) but not the former (boundary conditions).

Let us also mention that the balance equation of porosity yields an important contribution to interactions appearing in steady state processes. This would not be the case within the models of Biot and Bowen. The problem is extensively discussed for a cylindrical filter in the paper of B. ALBERS and K. WILMANSKI [15].

We shall skip here also the problem of the remaining boundary conditions needed for the construction of solutions of the above set of field equations. Some aspects of this problem have been discussed in my work [4] and in the paper of W. KEMPA [16].

5. Thermodynamical admissibility

We derive the thermodynamic restrictions for the constitutive relations (4.5) in the way which since some years is already standard for models within continuum thermodynamics. Foundations and examples of this procedure can be found in the book of I.MÜLLER [17] (see also: K. WILMANSKI [18], where porous materials are discussed).

For isothermal processes in two-component porous materials in the Lagrangian description (e.g. see: [5,11]) it can be formulated as follows. Any solution of field equations must satisfy identically the following inequality

$$\begin{aligned} & \rho^S \frac{\partial \Psi^S}{\partial t} + \rho^F \left(\frac{\partial \Psi^F}{\partial t} + \mathbf{X}'^F \cdot \text{Grad } \Psi^F \right) - \\ & - \mathbf{P}^S \cdot \frac{\partial \mathbf{F}^S}{\partial t} - \mathbf{P}^F \cdot \text{Grad } \mathbf{x}'^F - \mathbf{F}^{\text{ST}} \hat{\mathbf{p}} \cdot \mathbf{X}'^F \leq 0, \end{aligned} \quad (5.1)$$

where Ψ^S , Ψ^F denote partial Helmholtz free energies of the skeleton and of the fluid component, respectively. These are assumed to be constitutive quantities, i.e.

$$\Psi^S = \Psi^S(\boldsymbol{\epsilon}), \quad \Psi^F = \Psi^F(\boldsymbol{\epsilon}). \quad (5.2)$$

The inequality (5.1) follows easily from the entropy inequality and the energy conservation law under the condition of constant temperature. It is the main part of the second law of thermodynamics. In the sequel we consider some other conditions imposed by this law on the constitutive relations.

The limitation of the inequality to solutions of the field equations can be eliminated by the method of Lagrange multipliers. We skip here the technical details of this method and present the final results.

Bearing this in mind we substitute the constitutive relations (5.2) in (5.1) and eliminate constraints of the mass balance (4.1)₁ and the balance of porosity (4.1)₂ by means of the Lagrange multipliers Λ^p and Λ^n , respectively. Momentum balance equations are assumed to make no contribution due to the presence of arbitrary external body forces. It can be shown that this additional assumption yields sufficient conditions for thermodynamic compatibility. It can be avoided on the cost of simplicity yielding undesired generality.

Making use of the chain rule of differentiation we arrive at the following **thermodynamic admissibility conditions**

$$\begin{aligned} \Lambda^\rho &= \rho^S \frac{\partial \Psi^S}{\partial \rho^F} + \rho^F \frac{\partial \Psi^F}{\partial \rho^F}, & \Lambda^n &= \rho^S \frac{\partial \Psi^S}{\partial n} + \rho^F \frac{\partial \Psi^F}{\partial n}, \\ \rho^S \frac{\partial \Psi^S}{\partial \rho^F} \mathbf{X}'^F + \Lambda^n \frac{\partial \mathbf{J}}{\partial \rho^F} &= 0, & \rho^F \frac{\partial \Psi^F}{\partial n} \mathbf{X}'^F - \Lambda^n \frac{\partial \mathbf{J}}{\partial n} &= 0, \\ \rho^S \frac{\partial \Psi^S}{\partial \mathbf{X}'^F} + \rho^F \frac{\partial \Psi^F}{\partial \mathbf{X}'^F} &= 0, \end{aligned} \quad (5.3)$$

and

$$\begin{aligned} \mathbf{P}^S + \mathbf{P}^F &= 2\mathbf{F}^S \left(\rho^S \frac{\partial \Psi^S}{\partial \mathbf{C}^S} + \rho^F \frac{\partial \Psi^F}{\partial \mathbf{C}^S} \right), \\ \mathbf{F}^{ST} \mathbf{P}^F &= -\rho^F \Lambda^\rho \mathbf{1} + \rho^F \frac{\partial \Psi^F}{\partial \mathbf{X}'^F} \otimes \mathbf{X}'^F - \Lambda^n \left(\frac{\partial \mathbf{J}}{\partial \mathbf{X}'^F} \right)^T, \\ \text{sym}^{23} \left\{ \mathbf{P}^F \otimes \mathbf{X}'^F - 2\mathbf{F}^S \left[\rho^F \frac{\partial \Psi^F}{\partial \mathbf{C}^S} \otimes \mathbf{X}'^F - \Lambda^n \left(\frac{\partial \mathbf{J}}{\partial \mathbf{C}^S} \right)^{T^{13}} \right] \right\} &= 0. \end{aligned} \quad (5.4)$$

There remains the *residual inequality* which describes the **dissipation** density \mathcal{D}

$$\mathcal{D} \equiv \mathbf{F}^{ST} \hat{\mathbf{p}} \cdot \mathbf{X}'^F - \Lambda^n \hat{n} \geq 0. \quad (5.5)$$

We return to the discussion of the identities (5.3) and (5.4) in the next section. However we comment on the inequality (5.5).

The structure of dissipation density \mathcal{D} shows that the irreversibility of processes described by the present model is due to two mechanisms - the diffusion and the pore relaxation. The former is absent if there is no relative motion of components: $\mathbf{X}'^F = 0$. The latter source of dissipation vanishes with the vanishing source of porosity: $\hat{n} = 0$. In such a case the balance equation of porosity (4.2)₂ becomes the conservation law - the changes of porosity are driven by other fields of the model and one can introduce a constitutive relation for porosity as it has been done in my work [4].

A thermodynamic state in which both sources of dissipation vanish simultaneously is called the **state of thermodynamical equilibrium**. Clearly the dissipation density \mathcal{D} may not only be equal to zero in this state. However, according to the inequality (5.5), it reaches a **minimum** in this state. This means that we have to impose additional conditions on the constitutive functions appearing in (5.5) which are called the **thermodynamic stability conditions**. We shall discuss them in the section 7 of this work.

6. Isotropy

We proceed to investigate the thermodynamic admissibility conditions (5.3) and (5.4) under an additional assumption of **isotropy**. This means that constitutive quantities are invariant with respect to an arbitrary orthogonal transformation in reference configuration. Consequently we have for scalar constitutive quantities

$$\begin{aligned}\forall \mathbf{O} \in \mathcal{Orth}: \quad \Psi^S(\rho^F, n, \mathbf{C}^S, \mathbf{X}'^F) &= \Psi^S(\rho^F, n, \mathbf{O}\mathbf{C}^S\mathbf{O}^T, \mathbf{O}\mathbf{X}'^F), \\ \Psi^F(\rho^F, n, \mathbf{C}^S, \mathbf{X}'^F) &= \Psi^F(\rho^F, n, \mathbf{O}\mathbf{C}^S\mathbf{O}^T, \mathbf{O}\mathbf{X}'^F), \\ \hat{n}(\rho^F, n, \mathbf{C}^S, \mathbf{X}'^F) &= \hat{n}(\rho^F, n, \mathbf{O}\mathbf{C}^S\mathbf{O}^T, \mathbf{O}\mathbf{X}'^F),\end{aligned}\tag{6.1}$$

and for vector constitutive quantities

$$\begin{aligned}\forall \mathbf{O} \in \mathcal{Orth}: \quad \mathbf{O}\mathbf{J}(\rho^F, n, \mathbf{C}^S, \mathbf{X}'^F) &= \mathbf{J}(\rho^F, n, \mathbf{O}\mathbf{C}^S\mathbf{O}^T, \mathbf{O}\mathbf{X}'^F), \\ \mathbf{O}\mathbf{F}^{ST}\hat{\mathbf{p}}(\rho^F, n, \mathbf{C}^S, \mathbf{X}'^F) &= \mathbf{F}^{ST}\hat{\mathbf{p}}(\rho^F, n, \mathbf{O}\mathbf{C}^S\mathbf{O}^T, \mathbf{O}\mathbf{X}'^F),\end{aligned}\tag{6.2}$$

where

$$\mathcal{Orth} \equiv \left\{ \mathbf{O} - \text{second rank tensor} \mid \mathbf{O}^T = \mathbf{O}^{-1} \right\}.\tag{6.3}$$

We shall not quote here the relations for isotropic tensors of the second rank. There would be needed for partial stress tensors but these are given in terms of scalar and vector functions through thermodynamical admissibility relations.

It can be easily shown that the scalar functions Ψ^S , Ψ^F and \hat{n} satisfy the relations (6.1) if and only if they depend on their arguments solely through the following **invariants**

$$\begin{aligned}\mathcal{I}_{\text{ISO}} &\equiv \left\{ \rho^F, n, \text{I, II, III, IV, V, VI} \right\}, \\ \text{I} &= \mathbf{1} \cdot \mathbf{C}^S, \quad \text{II} = \frac{1}{2}(\text{I}^2 - \mathbf{1} \cdot \mathbf{C}^{S2}), \quad \text{III} = \det \mathbf{C}^S \equiv \text{J}^{S2}, \\ \text{IV} &= \mathbf{X}'^F \cdot \mathbf{X}'^F, \quad \text{V} = \mathbf{C}^S \cdot (\mathbf{X}'^F \otimes \mathbf{X}'^F), \quad \text{VI} = \mathbf{C}^{S2} \cdot (\mathbf{X}'^F \otimes \mathbf{X}'^F).\end{aligned}\tag{6.4}$$

On the other hand the vector functions must have the following general representation

$$\begin{aligned}\mathbf{J} &= (\Phi_0 \mathbf{1} + \Phi_1 \mathbf{C}^S + \Phi_2 \mathbf{C}^{S2}) \mathbf{X}'^F, \\ \mathbf{F}^{ST}\hat{\mathbf{p}} &= (\pi_0 \mathbf{1} + \pi_1 \mathbf{C}^S + \pi_2 \mathbf{C}^{S2}) \mathbf{X}'^F,\end{aligned}\tag{6.5}$$

where the coefficients are the isotropic scalar functions

$$\Phi_a = \Phi_a(\mathcal{I}_{\text{ISO}}), \quad \pi_a = \pi_a(\mathcal{I}_{\text{ISO}}), \quad a = 0, 1, 2.\tag{6.6}$$

Substitution of the above representations in the thermodynamic identities (5.3)_{3,4} yields (see: Appendix)

$$\begin{aligned} \rho^S \frac{\partial \Psi^S}{\partial \rho^F} + \left(\rho^S \frac{\partial \Psi^S}{\partial n} + \rho^F \frac{\partial \Psi^F}{\partial n} \right) \frac{\partial \Phi_0}{\partial \rho^F} &= 0, \\ \rho^F \frac{\partial \Psi^F}{\partial n} - \left(\rho^S \frac{\partial \Psi^S}{\partial n} + \rho^F \frac{\partial \Psi^F}{\partial n} \right) \frac{\partial \Phi_0}{\partial n} &= 0. \end{aligned} \quad (6.7)$$

These relations expose the rule played by the flux of porosity \mathbf{J} in the description of „static“ couplings between components. If the function Φ_0 was independent of the porosity than the partial Helmholtz free energy Ψ^F would have to be independent of the porosity as well. This would eliminate coupling of processes in the fluid component with the skeleton due to the changes of the confinement of motion of the fluid component to pores of the skeleton. Simultaneously if the function Φ_0 was independent of the mass density of the fluid component than the partial Helmholtz free energy Ψ^S would have to be independent of the mass density of the fluid. This would eliminate the coupling between volume changes of the fluid and corresponding reaction stresses in the skeleton. Solely the coupling through the diffusive forces in momentum balance equations would remain in the model.

We proceed to make use of the identities (5.4)_{2,3}. In addition to these identities we use the symmetry of the partial Cauchy stress tensor in the fluid which yields

$$\mathbf{P}^F \mathbf{F}^{\text{ST}} = \mathbf{F}^S \mathbf{P}^{\text{FT}}. \quad (6.8)$$

This is the consequence of the moment of momentum conservation for the fluid component. The similar symmetry condition for the partial Cauchy stress tensor in the skeleton follows then identically from the relation (5.4)₁.

Apart from the above condition we make the simplifying assumption concerning the identity (5.4)₃. We assume namely that it is not only the symmetrical part with respect to the last two indicies which must be zero but that the whole tensor of the third rank vanishes. There is a strong indication that it is not necessary to make this assumption in order to obtain the results which we present further in this section. However I have not been able to prove this statement.

Bearing in mind the above remarks we obtain (see: Appendix for the details)

$$\Phi_1 = 0, \quad \Phi_2 = 0 \quad \Rightarrow \quad \mathbf{J} = \Phi_0 \mathbf{X}'^F. \quad (6.9)$$

This important result simplifies immensely all thermodynamic admissibility relations.

First of all we see that the flux of porosity \mathbf{J} must be colinear with the Lagrangian (relative) velocity. This property allows to consider steady state processes in the microstructure, in particular changes of porosity, as driven by volume changes of both components [15].

Secondly the remaining identities can be jointly written in the following compact form

$$\mathbf{J}^S d\left(\frac{\Phi_0}{\mathbf{J}^S}\right) - \frac{\partial \Phi_0}{\partial V} dV = \frac{1}{\Lambda^n} \left[\rho^F d\Psi^F - \rho^F \frac{\partial \Psi^F}{\partial V} dV - \mathbf{J}^S \Lambda^p d\left(\frac{\rho^F}{\mathbf{J}^S}\right) \right]. \quad (6.10)$$

where Λ^p and Λ^n are the multipliers given by the relations (5.3)_{1,2}.

The formula (6.10) is the generalization of the classical Gibbs relation and, in contrast to the latter, contains contributions of nonequilibrium quantities, connected with diffusion and relaxation of porosity.

It is easy to see that it is solely the dependence of the flux function Φ_0 on the invariant V , i.e. on the magnitude of the Euclidean relative velocity: $V \equiv (\mathbf{x}'^F - \mathbf{x}'^S) \cdot (\mathbf{x}'^F - \mathbf{x}'^S)$, which is not determined by the partial Helmholtz free energy functions. If this dependence was known we could find the flux of porosity up to a constant. The experimental data would be needed solely to determine this constant. Such cases have been already considered (e.g. [7,9]) in the wave analysis. Incidentally, the dependence of Φ_0 on V must be at least quadratic and, consequently it would not appear in the case of processes near the thermodynamic equilibrium.

In addition the scalar constitutive functions Ψ^S , Ψ^F , Φ_0 are thermodynamically admissible if they satisfy the following conditions

$$\rho^S \frac{\partial \Psi^S}{\partial \mathcal{A}_2} + \rho^F \frac{\partial \Psi^F}{\partial \mathcal{A}_2} = 0, \quad \mathcal{A}_2 = \text{IV, V, VI}, \quad (6.11)$$

We obtain as well the following relations for the partial stress tensors

$$\begin{aligned} \mathbf{P}^S &= 2\rho^S \mathbf{F}^S \left[\frac{\partial \Psi^S}{\partial \text{I}} \mathbf{1} + \left(\text{II} \frac{\partial \Psi^S}{\partial \text{II}} + \text{III} \frac{\partial \Psi^S}{\partial \text{III}} \right) \mathbf{C}^S - \text{III} \frac{\partial \Psi^S}{\partial \text{II}} \mathbf{C}^{S2} + \right. \\ &\quad \left. + 2 \frac{\partial \Psi^S}{\partial V} \mathbf{X}'^F \otimes \mathbf{X}'^F \right] - 2 \left(\rho^S \frac{\partial \Psi^S}{\partial n} + \rho^F \frac{\partial \Psi^F}{\partial n} \right) \left[\frac{\partial \Phi_0}{\partial \text{I}} \mathbf{1} + \right. \\ &\quad \left. + \left(\text{II} \frac{\partial \Phi_0}{\partial \text{II}} + \text{III} \frac{\partial \Phi_0}{\partial \text{III}} \right) \mathbf{C}^S - \text{III} \frac{\partial \Phi_0}{\partial \text{II}} \mathbf{C}^{S2} + 2 \frac{\partial \Phi_0}{\partial V} \mathbf{X}'^F \otimes \mathbf{X}'^F \right], \quad (6.12) \\ \mathbf{P}^F &= - \left[\rho^F \left(\rho^S \frac{\partial \Psi^S}{\partial \rho^F} + \rho^F \frac{\partial \Psi^F}{\partial \rho^F} \right) + \left(\rho^S \frac{\partial \Psi^S}{\partial n} + \rho^F \frac{\partial \Psi^F}{\partial n} \right) \Phi_0 \right] \mathbf{F}^{S-T} + \\ &\quad + 2 \left[\rho^F \frac{\partial \Psi^F}{\partial V} + \left(\rho^S \frac{\partial \Psi^S}{\partial n} + \rho^F \frac{\partial \Psi^F}{\partial n} \right) \frac{\partial \Phi_0}{\partial V} \right] \mathbf{F}^S \mathbf{X}'^F \otimes \mathbf{X}'^F. \end{aligned}$$

The nonlinear dependence on the diffusion velocity yields a non-spherical structure of the partial stress tensor in the fluid. If we assume that this cannot be the case then the dependence on the fifth invariant must also vanish from the Helmholtz free energy for the fluid and, consequently, due to the relation (6.11) - from the free energy of the skeleton. In such a case we need indeed only a constant to determine Φ_0 .

The partial stress tensors described by the relations (6.12) have almost an expected structure if we ignore the above mentioned problem with the fifth invariant. The partial

stress tensor for the skeleton contains the part which is formally identical with this of the non-linear elasticity. This is the part following from the differentiation of the partial Helmholtz free energy function for skeleton. The interaction with the fluid is described by a similar differentiation of the flux Φ_0 . Essential in this contribution is the dependence of the partial free energies on porosity. Without this dependence the coupling terms in stresses would vanish.

Again if we ignore the dependence on the fifth invariant the partial stress tensor in the fluid reduces to an expected form of the pressure. This pressure consists of three parts. The classical one follows by the differentiation of the partial Helmholtz free energy for the fluid with respect to the mass density of the fluid. The second part describes the contribution of the skeleton through the dependence of the free energy Ψ^S on the mass density of the fluid. Finally the third part is due to the flux of porosity Φ_0 . Here again the dependence of the partial energies on the porosity is essential.

The above results do not exhaust all consequences of thermodynamic identities. We should add integrability conditions to these relations. In the purely mechanical model considered in this work those integrability conditions do not yield any considerable simplifications of constitutive relations. Therefore we skip their discussion in this paper.

7. Thermodynamical stability

We return now to the discussion of the dissipation inequality (5.5). After substitution of the isotropic representation (6.5) and the relation (5.3) for the multiplier Λ^n we have

$$\mathcal{D} = (\pi_0 IV + \pi_1 V + \pi_2 VI) - \left(\rho^S \frac{\partial \Psi^S}{\partial n} + \rho^F \frac{\partial \Psi^F}{\partial n} \right) \hat{n} \geq 0. \quad (7.1)$$

As we have already mentioned in the section 5 the state in which the dissipation \mathcal{D} reaches its minimum equal to zero is called the state of thermodynamic equilibrium. In the case of the general model it appears if

$$\mathbf{X}'^F|_E = 0, \quad \hat{n}|_E = 0, \quad (7.2)$$

where the subscript E denotes the equilibrium.

It is easy to see that the time derivative of porosity in this state must be also zero. Consequently we can introduce the notion of the **equilibrium porosity** n_E which must be solely the function of position \mathbf{X} . The deviation of current porosity n from this equilibrium porosity shall be denoted by Δ

$$n_E \equiv n|_E = n_E(\mathbf{X}) \quad \Rightarrow \quad \Delta|_E = 0, \quad \Delta \equiv n - n_E. \quad (7.3)$$

The new variable Δ replaces the porosity as a constitutive variable. The porosity n_E appears then solely as the parameter in constitutive functions. By means of this new variable we can replace the condition for the source of porosity in equilibrium by the following one

$$\mathbf{X}'^F|_E = 0, \Delta|_E = 0 \Rightarrow \mathcal{D}|_E \equiv \mathcal{D}(n_E; \rho^F, \Delta = 0, \text{I, II, III, IV} = 0, \text{V} = 0, \text{VI} = 0) = 0. \quad (7.4)$$

According to the inequality (7.1) the dissipation reaches in this state its minimum value. Consequently under smoothness assumptions of this work the conditions for this state have the form

$$\frac{\partial \mathcal{D}}{\partial \mathbf{X}'^F}|_E = 0, \quad \frac{\partial \mathcal{D}}{\partial \Delta}|_E = 0, \quad \begin{pmatrix} \frac{\partial^2 \mathcal{D}}{\partial \mathbf{X}'^F \partial \mathbf{X}'^F} & \frac{\partial^2 \mathcal{D}}{\partial \mathbf{X}'^F \partial \Delta} \\ \frac{\partial^2 \mathcal{D}}{\partial \Delta \partial \mathbf{X}'^F} & \frac{\partial^2 \mathcal{D}}{\partial \Delta^2} \end{pmatrix}_E - \text{positive definite.} \quad (7.5)$$

Such an equilibrium state is called **absolutely stable**.

Let us mention that some restricted models which follow from the one presented in the paper may lead to some other types of the thermodynamic equilibrium state. The most important example is the model based on the assumptions that the pore relaxation processes are absent and that the diffusion force is negligible, i.e.

$$\hat{n} \equiv 0, \quad \pi_0 \equiv 0, \quad \pi_1 \equiv 0, \quad \pi_2 \equiv 0. \quad (7.6)$$

Such models describe, for instance, spongrubbers filled with the air under atmospheric pressure as well as some other *systems with „empty“ pores*. Under the conditions (7.6) all processes in such systems are reversible which means that all states are equilibrium states. Such an equilibrium is called **neutrally stable**. The conditions (7.6) concern solely the quantities appearing in the residual inequality. Consequently all other constitutive considerations of this work remain valid for these restricted models. It is easy to see that the changes of porosity are unequal zero. According to the balance equation of porosity they are driven by the relative motion of components and primarily by the deformation of the skeleton reflected in the function Φ_0 . We shall not consider such models any further in this work.

Now we proceed to investigate the conditions (7.5). The relation (7.2) and the first two conditions (7.5) yield immediately the following equivalent conditions

$$\begin{aligned} \hat{n}|_E = 0 &\Rightarrow \exists \tau(n_E; \rho^F, \Delta, \text{I, II, III, IV, V, VI}): \hat{n} = -\frac{\Delta}{\tau}, \\ \left(\rho^S \frac{\partial \Psi^S}{\partial n} + \rho^F \frac{\partial \Psi^F}{\partial n} \right)|_E = 0 &\Rightarrow \left(\rho^S \frac{\partial \Psi^S}{\partial \Delta} + \rho^F \frac{\partial \Psi^F}{\partial \Delta} \right)|_E = 0, \end{aligned} \quad (7.7)$$

which yields in turn

$$\begin{aligned}
\rho^S \Psi^S &= \rho^S \Psi_0^S(n_E; \rho^F, I, II, III, IV, V, VI) + \\
&\quad + \frac{1}{2} \rho^S \Psi_2^S(n_E; \rho^F, \Delta, I, II, III, IV, V, VI) \Delta^2, \\
\rho^F \Psi^F &= \rho^F \Psi_0^F(n_E; \rho^F, I, II, III, IV, V, VI) + \\
&\quad + \frac{1}{2} \rho^F \Psi_2^F(n_E; \rho^F, \Delta, I, II, III, IV, V, VI) \Delta^2.
\end{aligned} \tag{7.8}$$

The last condition (7.5) is equivalent to the inequalities

$$\pi_0|_E > 0, \pi_1|_E > 0, \pi_2|_E > 0, \tau|_E (\rho^S \Psi_2^S + \rho^F \Psi_2^F)|_E > 0. \tag{7.9}$$

By means of these results we can construct simplified models describing processes which yield small deviations from the thermodynamical equilibrium. Such a model is presented, for instance, in the work [9]. For example, it can be shown that the last inequality (7.9) yields the positive relaxation time of porosity τ in such a model. It is worth mentioning that the thermodynamic admissibility conditions do not specify the sources of the model any further. We need experimental data to make the constitutive relations for the quantities π_0, π_1, π_2 and τ explicit.

Let as mention finally that the second law of thermodynamics is usually supplemented with still another stability condition. It is called the **convexity** condition of free energies and yield such results as the positivity of the specific heats, hyperbolicity of the field equations, the positivity of non-equilibrium contributions Ψ_2^S, Ψ_2^F and consequently the positivity of relaxation time τ , etc. We skip this problem in this work.

8. On identification problems

We have already mentioned identification problems arising in the relation between the macroscopic and semimicroscopic description. These problems are connected with experimental techniques which deliver data both on the macroscopic level as well as on the semimicroscopic level of observation. For instance dynamical sonar experiments in which speeds of propagation and attenuation coefficients for sound waves are recorded describe the smeared-out macroscopic properties of the porous material provided wave lengths are sufficiently large. On the other hand the examination of pores with probes delivers the semimicroscopic data on a real pore water pressure, pore water temperature etc.

Consequently if we want to incorporate both sorts of measurements in the description by means of continuous models we need to answer the question on rules of identification for quantities appearing in such models. It seems to me that a general answer to such questions cannot be given for principal reasons.

In order to appreciate better these questions we consider an almost trivial example of connection between the partial pressure p^F appearing in the macroscopic model and the pore water pressure p_w measured in the static *in situ* experiments on soils. We consider

the pressure p^F to be solely the function of the partial mass density ρ^F and such that the macroscopic compressibility κ^F is constant

$$\frac{1}{\kappa^F} \equiv \rho^F \frac{d p^F}{d \rho^F} = \text{const.} \Rightarrow p^F = p_0^F + \frac{1}{\kappa^F} \ln \left(\frac{\rho^F}{\rho_0^F} \right), \quad (8.1)$$

where $p_0^F = p^F(\rho_0^F)$ and ρ_0^F is an arbitrary reference value of the partial mass density.

We consider solely small deviations of mass density from this reference value, i.e.

$$\left| \frac{\rho^F}{\rho_0^F} - 1 \right| \ll 1 \Rightarrow \frac{\rho^F - \rho_0^F}{\rho_0^F} \approx \kappa^F (p^F - p_0^F). \quad (8.2)$$

In the static case considered in the above mentioned experiments of soil mechanics the partial momentum balance (4.2)₄ has the form

$$-\text{grad } p^F + \rho^F \mathbf{b}^F = 0, \quad \mathbf{b}^F = g \mathbf{e}_3, \quad (8.3)$$

where g is the earth acceleration and we have chosen the Cartesian coordinates with the \mathbf{e}_3 -basis vector in the direction of this acceleration.

Easy integration with respect to the z -coordinate in \mathbf{e}_3 -direction yields then the classical Torricelli solution

$$p^F \approx p_0^F + \rho_0^F g z. \quad (8.4)$$

If we assume that the partial mass density ρ_0^F and the true mass density ρ_0^{FR} are connected in the usual way through the porosity n then we have

$$\rho_0^F = n \rho_0^{\text{FR}} \Rightarrow p_w = p_{w0} + \rho_0^{\text{FR}} g z \quad \text{with} \quad p^F \approx n p_w. \quad (8.5)$$

This is the relation appearing in soil mechanics. Obviously such an identification cannot be required in the general case. The partial pressure p^F - in contrast to the pore water pressure p_w - depends on the constitutive variables describing the skeleton as well as on the relative motion of components. Consequently we cannot even expect that such universal identification relations exist! In the best case we may be able to carry through an identification analysis of a particular solution of boundary value problem - as we did in the above example.

It should be also born in mind that, in spite of the possibility of their continuous description, the true components belong to the microscopic back-ground of the macroscopic models of porous materials. If we were able to construct the full transition from this microscopic level to the macroscopic level by means of some averages - as it has been done in the section 3 of this paper for some geometrical properties - we would most likely have the relation, for instance, for the macroscopic partial pressure in the fluid in terms of the pore water pressure and some other microscopic variables. However, similarly to all other theories based on averaging procedures, there would be no way to reproduce, even only some, microscopic properties by means of macroscopic

properties because - trivially - we loose most of the microscopic information in the averaging. This obvious remark seems to be overlooked by many people working on applications of the theory of porous materials to soil mechanics.

Simultaneously any sophisticated identification of fields measured in macroscopic experiments is not needed at all because they correspond directly to fields appearing in the model. This shows that the macroscopic models of porous materials are sound from the point of view of the macroscopic physics to the same extent as any other macroscopic description in continuum thermodynamics.

9. Concluding remarks

The most important part of the model presented in this work is the closure of the set of field equations for the compressible porous materials by means of the balance equation of porosity. We have shown in the section 3 through the image analysis of the microstructure that it is justified to use the full balance equation and not only the evolution equation for the changes of porosity as it has been done earlier. Moreover we have shown that the thermodynamic admissibility and the isotropy yield an almost explicit relation for the flux in such a balance equation. Due to the relation (6.10) the partial Helmholtz free energies determine this flux up to the dependence on the magnitude of the relative velocity.

In order to see the rule played by the equation for porosity let us inspect its consequences in a stationary process. According to (6.9) and (7.7) we have

$$\mathbf{n} = \mathbf{n}|_E - \tau \text{Div}(\Phi_0 \mathbf{X}'^F), \quad (9.1)$$

or, in Eulerian coordinates,

$$\mathbf{n} = \mathbf{n}|_E - \tau \mathbf{J}^S \text{div}[\mathbf{J}^{S-1} \Phi_0 (\mathbf{v}^F - \mathbf{v}^S)], \quad (9.2)$$

where \mathbf{v}^F and \mathbf{v}^S are the velocities of components in Eulerian description. It is obvious that changes of porosity become important for sufficiently large relaxation times τ . This is the case under large deformations of soft materials such as biological tissues or clays (see: B. ALBERS, K. WILMANSKI [15]). For moderate velocity gradients these changes can be usually neglected for such hard materials as rocks and ceramics. Let us notice that changes of porosity in stationary processes are driven primarily by volume changes of both phases.

The model proposed here is also the simplest version of all possible extensions of the mixture theory on multicomponent systems of immiscible components. The porosity is the only additional microstructural variable needed for the description of processes in such systems. This is, certainly, much simpler than the earlier approaches based on the notion of volume fractions and the saturation condition. In addition the experimental data for the sound propagation seem to support this simpler approach. In the case of the theories with volume fractions for systems consisting of more than two components one should expect more than two longitudinal waves. This has never been observed. The experiments expose solely the classical longitudinal and transversal waves and the slow Biot's wave as indeed predicted by the present theory (e.g. [7,8]).

The above mentioned relative simplicity of the present model as well as the consistent Lagrangian description of the multicomponent medium yield the real possibilities of numerical analysis of some boundary value problems. The weak formulation of the linearized version of the model and some numerical results are presented in the paper of W. KEMPA [16]. The fully non-linear model has been as yet not analyzed from this point of view.

Appendix

We present here the derivation of thermodynamic identities following from the thermodynamic relations (5.3) and (5.4) together with the isotropy assumptions of section 6.

In the further analysis we refer often to the spectral representation of vectors and tensors. Most important of them is the deformation tensor \mathbf{C}^S . We have

$$\mathbf{C}^S = \sum_{\alpha=1}^3 \lambda^{(\alpha)} \mathbf{k}_\alpha \otimes \mathbf{k}_\alpha, \quad (\mathbf{C}^S - \lambda^{(\alpha)} \mathbf{1}) \mathbf{k}_\alpha = 0, \quad \mathbf{k}_\alpha \cdot \mathbf{k}_\beta = \delta_{\alpha\beta}, \quad (\text{A.1})$$

i.e. $\lambda^{(\alpha)}$ are the eigenvalues and \mathbf{k}_α are the normalized eigenvectors of the deformation tensor \mathbf{C}^S . It is quite obvious that due to the relations

$$\text{I} = \lambda^{(1)} + \lambda^{(2)} + \lambda^{(3)}, \quad \text{II} = \lambda^{(1)}\lambda^{(2)} + \lambda^{(1)}\lambda^{(3)} + \lambda^{(2)}\lambda^{(3)}, \quad \text{III} = \lambda^{(1)}\lambda^{(2)}\lambda^{(3)}, \quad (\text{A.2})$$

the isotropic scalar functions can be equivalently made dependent on the eigenvalues $\lambda^{(\alpha)}$ instead of the invariants I, II, III. Simultaneously these functions must be independent of the eigenvectors \mathbf{k}_α .

We proceed to exploit the thermodynamical admissibility conditions. Bearing in mind the above spectral representation of the deformation tensor we can write the relation (6.5)₁ for the flux of porosity in the following form

$$\mathbf{J} = \sum_{\alpha=1}^3 (\Phi_0 + \lambda^{(\alpha)} \Phi_1 + \lambda^{(\alpha)2} \Phi_2) \mathbf{V}^\alpha \mathbf{k}_\alpha, \quad \mathbf{X}'^F \equiv \sum_{\beta=1}^3 \mathbf{V}^\beta \mathbf{k}_\beta, \quad \mathbf{V}^\beta \equiv \mathbf{X}'^F \cdot \mathbf{k}_\beta. \quad (\text{A.3})$$

Substitution of this relation in the identities (5.3)_{3,4} yields

$$\begin{aligned} \rho^S \frac{\partial \Psi^S}{\partial \rho^F} + \Lambda^n \left(\frac{\partial \Phi_0}{\partial \rho^F} + \lambda^{(\alpha)} \frac{\partial \Phi_1}{\partial \rho^F} + \lambda^{(\alpha)2} \frac{\partial \Phi_2}{\partial \rho^F} \right) &= 0, \\ \rho^F \frac{\partial \Psi^F}{\partial n} - \Lambda^n \left(\frac{\partial \Phi_0}{\partial n} + \lambda^{(\alpha)} \frac{\partial \Phi_1}{\partial n} + \lambda^{(\alpha)2} \frac{\partial \Phi_2}{\partial n} \right) &= 0, \quad \text{for } \alpha = 1, 2, 3. \end{aligned} \quad (\text{A.4})$$

We have used here the fact that the Lagrangian velocity \mathbf{X}'^F is not identically zero as well as the linear independence of the eigenvectors.

The linear combinations of the relations (A.4)₁ yield

$$\begin{aligned}
(\lambda^{(2)} - \lambda^{(1)}) \frac{\partial \Phi_1}{\partial \rho^F} + (\lambda^{(2)2} - \lambda^{(1)2}) \frac{\partial \Phi_2}{\partial \rho^F} &= 0, \\
(\lambda^{(3)} - \lambda^{(1)}) \frac{\partial \Phi_1}{\partial \rho^F} + (\lambda^{(3)2} - \lambda^{(1)2}) \frac{\partial \Phi_2}{\partial \rho^F} &= 0,
\end{aligned} \tag{A.5}$$

i.e. bearing in mind that (A.5) must hold for arbitrary eigenvalues

$$\frac{\partial \Phi_1}{\partial \rho^F} = 0, \quad \frac{\partial \Phi_2}{\partial \rho^F} = 0. \tag{A.6}$$

In the similar way we obtain from (A.4)₂

$$\frac{\partial \Phi_1}{\partial n} = 0, \quad \frac{\partial \Phi_2}{\partial n} = 0. \tag{A.7}$$

Bearing in mind the relation (6.8) we obtain by means of the identity (5.4)₂

$$\begin{aligned}
&\frac{1}{2} \text{skew} \mathbf{P}^F \mathbf{F}^{ST} \equiv \\
&\equiv \mathbf{F}^{S-T} \left\{ \left(\rho^F \frac{\partial \Psi^F}{\partial IV} - \Lambda^n \frac{\partial \Phi_0}{\partial IV} + \Lambda^n \text{II} \frac{\partial \Phi_2}{\partial IV} + \Lambda^n \text{III} \frac{\partial \Phi_2}{\partial V} \right) \text{skew}(\mathbf{X}'^F \otimes \mathbf{C}^S \mathbf{X}'^F) + \right. \\
&+ \Lambda^n \left(-\frac{\partial \Phi_1}{\partial IV} - \text{I} \frac{\partial \Phi_2}{\partial IV} + \text{III} \frac{\partial \Phi_2}{\partial VI} \right) \text{skew}(\mathbf{X}'^F \otimes \mathbf{C}^{S2} \mathbf{X}'^F) + \left(\rho^F \frac{\partial \Psi^F}{\partial VI} - \Lambda^n \frac{\partial \Phi_0}{\partial VI} + \right. \\
&\left. \left. + \Lambda^n \frac{\partial \Phi_1}{\partial V} + \Lambda^n \text{I} \frac{\partial \Phi_2}{\partial V} + \Lambda^n \text{II} \frac{\partial \Phi_2}{\partial VI} \right) \text{skew}(\mathbf{C}^{S2} \mathbf{X}'^F \otimes \mathbf{C}^S \mathbf{X}'^F) \right\} \mathbf{F}^{S-1} = 0.
\end{aligned} \tag{A.8}$$

The skew-symmetric tensors in (A.8) can be represented by three linearly independent vectors. Consequently their coefficients must vanish and we obtain

$$\begin{aligned}
\rho^F \frac{\partial \Psi^F}{\partial IV} - \Lambda^n \frac{\partial \Phi_0}{\partial IV} + \Lambda^n \text{II} \frac{\partial \Phi_2}{\partial IV} &= -\Lambda^n \text{III} \frac{\partial \Phi_2}{\partial V}, \\
\frac{\partial \Phi_1}{\partial IV} + \text{I} \frac{\partial \Phi_2}{\partial IV} &= \text{III} \frac{\partial \Phi_2}{\partial VI}, \\
\rho^F \frac{\partial \Psi^F}{\partial VI} - \Lambda^n \frac{\partial \Phi_0}{\partial VI} + \Lambda^n \text{II} \frac{\partial \Phi_2}{\partial VI} &= -\Lambda^n \left(\frac{\partial \Phi_1}{\partial V} + \text{I} \frac{\partial \Phi_2}{\partial V} \right).
\end{aligned} \tag{A.9}$$

It remains to exploit the identity (5.4)₃. As we have mentioned we extend this condition and assume that the skewsymmetric part vanishes as well. Then we apply the tensor $\mathbf{P}^F \otimes \mathbf{X}'^F$ to the three linearly independent vectors \mathbf{X}'^F , $\mathbf{C}^S \mathbf{X}'^F$, $\mathbf{C}^{S2} \mathbf{X}'^F$. We obtain three relations for second order tensors which replace the original condition for the third order tensor. They have the following form

$$\begin{aligned}
\text{IV } \mathbf{F}^{\text{S-1}} \mathbf{P}^{\text{F}} &= -2 \Lambda^n \left[\Phi_1 \mathbf{X}'^{\text{F}} \otimes \mathbf{X}'^{\text{F}} + 2 \Phi_2 \text{sym}(\mathbf{C}^{\text{S}} \mathbf{X}'^{\text{F}} \otimes \mathbf{X}'^{\text{F}}) \right] + \\
&\quad + \text{IV } \mathbf{A}_1 + \text{V } \mathbf{A}_2 + \text{VI } \mathbf{A}_3, \\
\text{V } \mathbf{F}^{\text{S-1}} \mathbf{P}^{\text{F}} &= -2 \Lambda^n \left[\Phi_1 \mathbf{X}'^{\text{F}} \otimes \mathbf{C}^{\text{S}} \mathbf{X}'^{\text{F}} + \right. \\
&\quad \left. + \Phi_2 (\mathbf{C}^{\text{S}} \mathbf{X}'^{\text{F}} \otimes \mathbf{C}^{\text{S}} \mathbf{X}'^{\text{F}} + \mathbf{X}'^{\text{F}} \otimes \mathbf{C}^{\text{S}^2} \mathbf{X}'^{\text{F}}) \right] + \\
&\quad + \text{V } \mathbf{A}_1 + \text{VI } \mathbf{A}_2 + (\text{I VI} - \text{II V} + \text{III IV}) \mathbf{A}_3, \\
\text{VI } \mathbf{F}^{\text{S-1}} \mathbf{P}^{\text{F}} &= -2 \Lambda^n \left[\Phi_1 \mathbf{X}'^{\text{F}} \otimes \mathbf{C}^{\text{S}^2} \mathbf{X}'^{\text{F}} + \right. \\
&\quad \left. + \Phi_2 (\mathbf{C}^{\text{S}} \mathbf{X}'^{\text{F}} \otimes \mathbf{C}^{\text{S}^2} \mathbf{X}'^{\text{F}} + \mathbf{X}'^{\text{F}} \otimes \mathbf{C}^{\text{S}^3} \mathbf{X}'^{\text{F}}) \right] + \\
&\quad + \text{VI } \mathbf{A}_1 + (\text{I VI} - \text{II V} + \text{III IV}) \mathbf{A}_2 + \\
&\quad + (\text{I}^2 \text{ VI} - \text{I II V} + \text{I III IV} - \text{II VI} + \text{III V}) \mathbf{A}_3,
\end{aligned} \tag{A.10}$$

where

$$\begin{aligned}
\mathbf{A}_1 &\equiv 2 \left(\rho^{\text{F}} \frac{\partial \Psi^{\text{F}}}{\partial \text{I}} - \Lambda^n \frac{\partial \Phi_0}{\partial \text{I}} \right) \mathbf{1} - 2 \left(\rho^{\text{F}} \text{III} \frac{\partial \Psi^{\text{F}}}{\partial \text{II}} - \Lambda^n \text{III} \frac{\partial \Phi_0}{\partial \text{II}} \right) \mathbf{C}^{\text{S-2}} + \\
&\quad + 2 \left(\rho^{\text{F}} \text{II} \frac{\partial \Psi^{\text{F}}}{\partial \text{II}} + \rho^{\text{F}} \text{III} \frac{\partial \Psi^{\text{F}}}{\partial \text{III}} - \Lambda^n \text{II} \frac{\partial \Phi_0}{\partial \text{II}} - \Lambda^n \text{III} \frac{\partial \Phi_0}{\partial \text{III}} \right) \mathbf{C}^{\text{S-1}} + \\
&\quad + 2 \left(\rho^{\text{F}} \frac{\partial \Psi^{\text{F}}}{\partial \text{V}} - \Lambda^n \frac{\partial \Phi_0}{\partial \text{V}} \right) \mathbf{X}'^{\text{F}} \otimes \mathbf{X}'^{\text{F}} + \\
&\quad + 4 \left(\rho^{\text{F}} \frac{\partial \Psi^{\text{F}}}{\partial \text{VI}} - \Lambda^n \frac{\partial \Phi_0}{\partial \text{VI}} \right) \text{sym}(\mathbf{X}'^{\text{F}} \otimes \mathbf{C}^{\text{S}} \mathbf{X}'^{\text{F}}) = \mathbf{A}_1^{\text{T}}, \\
\mathbf{A}_2 &\equiv -2 \Lambda^n \left\{ \frac{\partial \Phi_1}{\partial \text{I}} \mathbf{1} - \text{III} \frac{\partial \Phi_1}{\partial \text{II}} \mathbf{C}^{\text{S-2}} + \left(\text{II} \frac{\partial \Phi_1}{\partial \text{II}} + \text{III} \frac{\partial \Phi_1}{\partial \text{III}} \right) \mathbf{C}^{\text{S-1}} + \right. \\
&\quad \left. + \frac{\partial \Phi_1}{\partial \text{V}} \mathbf{X}'^{\text{F}} \otimes \mathbf{X}'^{\text{F}} + 2 \frac{\partial \Phi_1}{\partial \text{VI}} \text{sym}(\mathbf{X}'^{\text{F}} \otimes \mathbf{C}^{\text{S}} \mathbf{X}'^{\text{F}}) \right\} = \mathbf{A}_2^{\text{T}}, \\
\mathbf{A}_3 &\equiv -2 \Lambda^n \left\{ \frac{\partial \Phi_2}{\partial \text{I}} \mathbf{1} - \text{III} \frac{\partial \Phi_2}{\partial \text{II}} \mathbf{C}^{\text{S-2}} + \left(\text{II} \frac{\partial \Phi_2}{\partial \text{II}} + \text{III} \frac{\partial \Phi_2}{\partial \text{III}} \right) \mathbf{C}^{\text{S-1}} + \right. \\
&\quad \left. + \frac{\partial \Phi_2}{\partial \text{V}} \mathbf{X}'^{\text{F}} \otimes \mathbf{X}'^{\text{F}} + 2 \frac{\partial \Phi_2}{\partial \text{VI}} \text{sym}(\mathbf{X}'^{\text{F}} \otimes \mathbf{C}^{\text{S}} \mathbf{X}'^{\text{F}}) \right\} = \mathbf{A}_3^{\text{T}},
\end{aligned} \tag{A.11}$$

and the following relations have been used

$$\begin{aligned}
\mathbf{C}^{\text{S}} \mathbf{X}'^{\text{F}} \cdot \mathbf{C}^{\text{S}^2} \mathbf{X}'^{\text{F}} &= \text{I VI} - \text{II V} + \text{III IV}, \\
\mathbf{C}^{\text{S}^2} \mathbf{X}'^{\text{F}} \cdot \mathbf{C}^{\text{S}^2} \mathbf{X}'^{\text{F}} &= \text{I}^2 \text{ VI} - \text{I II V} + \text{I III IV} - \text{II VI} + \text{III V}.
\end{aligned} \tag{A.12}$$

They can be easily proven by means of the Cayley-Hamilton theorem.

It is obvious that the left-hand side of the relations (A.10) is symmetric. Consequently

$$\begin{aligned} \Phi_1 \operatorname{skew}(\mathbf{X}'^F \otimes \mathbf{C}^S \mathbf{X}'^F) + \Phi_2 \operatorname{skew}(\mathbf{X}'^F \otimes \mathbf{C}^{S2} \mathbf{X}'^F) &= 0, \\ \Phi_1 \operatorname{skew}(\mathbf{X}'^F \otimes \mathbf{C}^{S2} \mathbf{X}'^F) + \Phi_2 \operatorname{skew}(\mathbf{C}^S \mathbf{X}'^F \otimes \mathbf{C}^{S2} \mathbf{X}'^F + \mathbf{X}'^F \otimes \mathbf{C}^{S3} \mathbf{X}'^F) &= 0. \end{aligned} \quad (\text{A.13})$$

Again we can introduce a vector representation for the above skew-symmetric tensors. We obtain then immediately the relations (6.9).

Now all three relations (A.10) reduce to the single relation

$$\mathbf{P}^F = \mathbf{F}^S \mathbf{A}_1. \quad (\text{A.14})$$

Consequently bearing in mind (5.4)₂ and (A.9) we obtain the following compatibility relations

$$\begin{aligned} & -(\rho^F \Lambda^p + \Lambda^n \Phi_0) \mathbf{C}^{S-1} = \\ & = 2 \left(\rho^F \frac{\partial \Psi^F}{\partial \text{I}} - \Lambda^n \frac{\partial \Phi_0}{\partial \text{I}} \right) \mathbf{1} - 2 \left(\rho^F \text{III} \frac{\partial \Psi^F}{\partial \text{II}} - \Lambda^n \text{III} \frac{\partial \Phi_0}{\partial \text{II}} \right) \mathbf{C}^{S-2} + \\ & + 2 \left(\rho^F \text{II} \frac{\partial \Psi^F}{\partial \text{II}} + \rho^F \text{III} \frac{\partial \Psi^F}{\partial \text{III}} - \Lambda^n \text{II} \frac{\partial \Phi_0}{\partial \text{II}} - \Lambda^n \text{III} \frac{\partial \Phi_0}{\partial \text{III}} \right) \mathbf{C}^{S-1} + \\ & + 4 \left(\rho^F \frac{\partial \Psi^F}{\partial \text{VI}} - \Lambda^n \frac{\partial \Phi_0}{\partial \text{VI}} \right) \operatorname{sym}(\mathbf{X}'^F \otimes \mathbf{C}^S \mathbf{X}'^F). \end{aligned} \quad (\text{A.15})$$

We use here again the spectral representation (A.1) and after easy calculations we obtain

$$\begin{aligned} \rho^F \frac{\partial \Psi^F}{\partial \text{I}} - \Lambda^n \frac{\partial \Phi_0}{\partial \text{I}} = 0, \quad \rho^F \frac{\partial \Psi^F}{\partial \text{II}} - \Lambda^n \frac{\partial \Phi_0}{\partial \text{II}} = 0, \\ \rho^F \frac{\partial \Psi^F}{\partial \text{VI}} - \Lambda^n \frac{\partial \Phi_0}{\partial \text{VI}} = 0. \end{aligned} \quad (\text{A.16})$$

In addition

$$\rho^F \Lambda^p + \Lambda^n \Phi_0 = -2 \text{III} \left(\rho^F \frac{\partial \Psi^F}{\partial \text{III}} - \Lambda^n \frac{\partial \Phi_0}{\partial \text{III}} \right). \quad (\text{A.17})$$

Bearing in mind the relations (5.3)_{1,2} defining the multipliers and the result (6.7) we obtain immediately

$$\begin{aligned} & \rho^F \left(\rho^F \frac{\partial \Psi^F}{\partial \rho^F} + 2\text{III} \frac{\partial \Psi^F}{\partial \text{III}} \right) \left(\frac{\partial \Phi_0}{\partial \rho^F} \right) = \\ & = -\rho^S \frac{\partial \Psi^S}{\partial \rho^F} \sqrt{\text{III}} \left[\rho^F \frac{\partial}{\partial \rho^F} \left(\frac{\Phi_0}{\sqrt{\text{III}}} \right) + 2\text{III} \frac{\partial}{\partial \text{III}} \left(\frac{\Phi_0}{\sqrt{\text{III}}} \right) \right]. \end{aligned} \quad (\text{A.18})$$

Now the combination of the results (A.16), (A.17) and (A.18) yields the relation (6.10).

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