

## Evolution of a fibre-reinforced growing mixture

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**Summary.** — We consider a biological tissue that can be macroscopically modelled as a biphasic mixture composed of a fluid and a solid phase. The former is a multi-constituent fluid, and the latter consists of a deformable porous medium comprising matrix and fibre-like inclusions. Both phases are assumed to be composed of several constituents, and are allowed to experience exchange interactions. In response to these interactions, the solid phase may either grow or be absorbed. We assume that each of these behaviours leads to the development of material inhomogeneities. Material inhomogeneities are treated by enforcing Kröner's multiplicative decomposition of the solid-phase deformation gradient tensor, and introducing an *inhomogeneity velocity "gradient"*. Through Onsager's principle, it is proven that inhomogeneity velocity "gradient" is related to the Mandel stress tensor of the solid phase, and chemical potentials of fluid constituents. This relation is used in order to show that, in response to growth (or adsorption), development of material inhomogeneities may trigger fibre reorientation in the solid phase by inducing the evolution in time of its texture tensor.

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### 1. – Introduction

The macroscopic mechanical behaviour of biological tissues is influenced by the presence of inclusions. For example, in the case of articular cartilage, the mechanical

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properties and geometric distribution of collagen fibres enhance the tissue resistance to external loads, and determine the tissue material symmetry.

If inclusions evolve in time, the response of the tissue to external stimuli adapts to the current organisation of the inclusions. This rearrangement of the internal structure, known as *remodelling* [1], implies that tissue material symmetries evolve in time. For this reason, an accurate characterisation of the tissue mechanical behaviour should be able to predict how inclusions evolve, and how their evolution is related to the quantities that determine the mechanical state of the tissue.

In the literature, several studies can be found where the influence of the inclusions on the tissue overall mechanical behaviour is considered. In the case of monophasic continua<sup>(1)</sup>, the role played by inclusions is investigated both in *small strain approximation* and in large deformations. Tools of investigation are different depending on the phenomenon that has to be described. In linear elasticity, models are usually based on the theory of composite materials with spheroidal inclusions (cf., for example, [2-4]). According to this approach, the effective strain in the inclusions,  $\tilde{\boldsymbol{\varepsilon}}$ , is expressed through the relation  $\tilde{\boldsymbol{\varepsilon}} = \mathbb{A} : \boldsymbol{\varepsilon}$ , where  $\boldsymbol{\varepsilon}$  is the strain field “felt” by the composite as a whole, and  $\mathbb{A}$  is a fourth-order tensor called *strain intensification tensor*. Tensor  $\mathbb{A}$  is expressed through the fourth-order *Eshelby tensor*, and, for spheroidal inclusions, depends on the inclusion aspect ratio and the elastic properties of both the matrix and inclusions. For large deformations, the presence of fibre-like inclusions is usually accounted for by enlisting the *texture tensor*,  $\boldsymbol{\Xi}$ , among the arguments of the body strain energy function. This approach has been followed, for example, in modelling arterial walls by [5-7].

Inclusions may also exhibit statistical orientation. In this case, it is possible to introduce a probability density distribution, which describes the probability of finding a fibre-like inclusion aligned along a given direction in space (this approach was used, for example, in [8-10] for modelling articular cartilage).

The texture tensor,  $\boldsymbol{\Xi}$ , is defined by  $\boldsymbol{\Xi} = \boldsymbol{\xi} \otimes \boldsymbol{\xi}$ , where  $\boldsymbol{\xi}$  is a unit vector describing the local alignment of a fibre-like inclusion along a prescribed direction of space. According to this description, and denoting by  $\mathbf{X}$  the material point of the body at which an inclusion is attached, the inclusions are locally rectified<sup>(2)</sup> in a neighbourhood of  $\mathbf{X} \in \mathcal{B}_R$ ,  $\mathcal{B}_R$  being the reference configuration of the body.

As the body deforms, the unit vector changes direction in time according to the following evolution law (cf., for example, [11]):

$$(1.1) \quad D_t \boldsymbol{\xi} = \mathbf{L} \boldsymbol{\xi} - [\boldsymbol{\xi} \cdot (\mathbf{D} \boldsymbol{\xi})] \boldsymbol{\xi},$$

where  $D_t \boldsymbol{\xi}$  is the convective time derivative of unit vector  $\boldsymbol{\xi}$  with respect to the body velocity,  $\mathbf{v}$ ,  $\mathbf{L} = \nabla \mathbf{v}$  is the velocity gradient tensor, and  $\mathbf{D} = \text{Sym}(\mathbf{L})$  is the symmetric part of  $\mathbf{L}$ . According to eq. (1.1) (which contains neither phenomenological parameters, nor material properties of the inclusion), the reorientation of the inclusion follows the deformation process  $\mathbf{L} = (D_t \mathbf{F}) \mathbf{F}^{-1}$ , where  $\mathbf{F}$  is the deformation gradient tensor of the body as a whole. This picture, valid as long as the unit vector is considered to be

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<sup>(1)</sup> Here, we use this terminology in order to distinguish continua consisting of a solid matrix and inclusions (which may be also referred to as solid constituents or sub-phases), from mixtures composed of both fluid and solid phases.

<sup>(2)</sup> This means that, in a given neighbourhood of  $\mathbf{X} \in \mathcal{B}_R$ , each fibre is assumed to be replaced by a straight fibre having the same elastic properties as the original fibre.

a mere descriptor of the inclusion orientation at point  $\mathbf{X} \in \mathcal{B}_R$ , ceases to hold if vector  $\boldsymbol{\xi}$  represents the direction of an inclusion embedded in the body. Indeed, in the latter case, the evolution of the inclusion should also take into account the intrinsic stiffness of the inclusion itself. This is due to the fact that, in the presence of an inclusion, the overall response of the medium (comprised of matrix and embedded inclusion) to external stimuli is statically undetermined, for the inclusion tends to bear a portion of stress which, within the small strain approximation, is proportional to its stiffness relative to that of the matrix.

Following the theory presented in [2-4], Federico *et al.* [12] proposed two approaches to this problem. The first approach, valid in *linear elasticity*, consists of introducing a velocity gradient tensor for the inclusion, the symmetric part of which is obtained by differentiating the effective strain in the inclusion,  $\tilde{\boldsymbol{\varepsilon}}$ , with respect to time. The second approach, instead, assumes that the effect of the inclusion on the symmetric part of the body velocity gradient,  $\mathbf{D}$ , can be given an expression similar to  $\tilde{\boldsymbol{\varepsilon}} = \mathbb{A} : \boldsymbol{\varepsilon}$ .

When growth is considered, the change of internal structure of the tissue is also driven by the adaptive re-distribution of its mass density as new material is added (or subtracted) to the pre-existing one. In the case of a materially uniform continuum body (*i.e.* a body whose points, made of the same material, can be brought to attain the same state simultaneously) described by a first-order constitutive theory, Epstein and Maugin [13] pointed out that the process of growth is essentially governed by temporal changes of mass density, and distortions of material-point neighbourhoods in the reference configuration of the body. Distortions, and related residual stresses, arise because of the possible loss of geometric compatibility of material-point neighbourhoods as growth takes place. Growth is thus viewed as a process capable of developing body material inhomogeneities. In analogy with plasticity, this is accounted for by introducing the multiplicative decomposition of the deformation gradient tensor,  $\mathbf{F} = \mathbf{F}^e \mathbf{K}^{-1}$ , where  $\mathbf{F}^e$  describes the *true* elastic deformation of the body, while  $\mathbf{K}^{-1}$  keeps track of the evolution of material inhomogeneities. Tensor  $\mathbf{K}^{-1}$  is called “*transplant operator*”, and maps the tangent space of the body reference configuration into the tangent space of an elastically released configuration, which is said to be the “*reference crystal*” [13]. In this context, the quantity enclosing necessary information about the evolution of material inhomogeneities is defined by  $\bar{\mathbf{L}}_K := \mathbf{K}^{-1} \mathbf{D}_t \mathbf{K}$ , and is called *inhomogeneity velocity gradient*. An equivalent formulation can be given by introducing the *growth tensor*  $\mathbf{F}^{\text{an}} \equiv \mathbf{K}^{-1}$  (here, the index “an” stands for “*an-elastic*”). Growth tensor accounts for the anelastic deformation ascribable to growth, and maps the tangent space of the body reference configuration into the tangent space of an elastically released intermediate configuration, called *natural configuration*. In this case, the *growth velocity gradient*,  $\mathbf{L}^{\text{an}} := (\mathbf{D}_t \mathbf{F}^{\text{an}})(\mathbf{F}^{\text{an}})^{-1}$ , is used in place of  $\bar{\mathbf{L}}_K$ .

In order to describe growth and remodelling, Imatani and Maugin [14], and Maugin and Imatani [15] modified eq. (1.1) by using the inhomogeneity velocity gradient tensor,  $\bar{\mathbf{L}}_K$ , and analysed the evolution of material anisotropy according to two different cases: a) the unit vector describing material anisotropy is embedded in the body (in particular, in the reference crystal); b) the unit vector is regarded as a *float*. In case a), the evolution of the unit vector embedded in the reference crystal,  $\boldsymbol{\lambda}^N$ , was found to be

$$(1.2) \quad \mathbf{D}_t \boldsymbol{\lambda}^N = [\boldsymbol{\lambda}^N \cdot (\bar{\mathbf{L}}_K \boldsymbol{\lambda}^N)] \boldsymbol{\lambda}^N - \bar{\mathbf{L}}_K \boldsymbol{\lambda}^N,$$

where  $\bar{\mathbf{L}}_K$  is assumed to depend on texture tensor,  $\boldsymbol{\Xi}^N = \boldsymbol{\lambda}^N \otimes \boldsymbol{\lambda}^N$ , and the elastic Mandel stress tensor. In case b), tensor  $\bar{\mathbf{L}}_K$  was replaced by the elastic Mandel stress tensor on

the basis of the fact that any other second order tensor fulfilling frame indifference in the reference crystal can be used instead of  $\bar{\mathbf{L}}_K$  in eq. (1.2) [14, 15].

In our paper, we would like to approach the problem of growth and remodelling in the context of mixture theory. For our purposes, we consider a mixture consisting of a fluid and a solid phase, and we assume that the solid phase is composed by a matrix and fibre-like inclusions. In the following, we refer to matrix and fibre-like inclusions as to solid sub-phases. We remark that, since fibre-like inclusions are regarded as a phase, their dimension is not seen in the model, while their orientation is accounted for by the solid-phase texture tensor,  $\Xi_S$ . According to this description, the solid phase of the mixture studied in this paper describes a homogenised system consisting of matrix and fibres. However, a possible way for accounting the dimension as well as the orientation of fibres is given, for example, in [16], where the effective field method is applied (cf., for example, [17]).

Growth is understood here as the process of mass increase (or decrease) of the solid phase due to mass exchanges of the latter with the fluid phase. This implies that the mixture is closed. Analogous approaches can be found, for example, in [18] and [19]. Although a more general framework, in which the mixture is assumed to be open, has been given (cf., for example, [20] and [16]), we considered here the case of a closed mixture for the sake of conciseness. The main hypothesis of our treatment is that solid sub-phases move with a common velocity, but are nonetheless allowed to undergo growth and remodelling independently. This means that, although the deformation of the solid-phase as a whole is described by only one deformation gradient tensor,  $\mathbf{F}_S$ , two multiplicative decompositions of  $\mathbf{F}_S$  are introduced, *i.e.*  $\mathbf{F}_S = \mathbf{F}_I^e \mathbf{F}_I^{\text{an}} = \mathbf{F}_M^e \mathbf{F}_M^{\text{an}}$ , where indices  $I$  and  $M$  denote fibre-like inclusions, and matrix, respectively, and tensors  $\mathbf{F}_I^{\text{an}}$  and  $\mathbf{F}_M^{\text{an}}$  account for the anelastic deformation in each solid sub-phase as the process of growth takes place (with respect to the work by Epstein and Maugin [13], each of the two tensors  $\mathbf{F}_I^{\text{an}}$  and  $\mathbf{F}_M^{\text{an}}$  can be viewed as the inverse of the corresponding transplant operator). In our picture, the unit vector describing the local orientation of fibre-like inclusions is denoted by  $\xi_S$ , and the *kinematic* evolution of the corresponding texture tensor,  $\Xi_S = \xi_S \otimes \xi_S$ , reads<sup>(3)</sup>

$$(1.3) \quad \frac{D_S \Xi_S}{Dt} = \Xi_S \mathbf{L}_S^T + \mathbf{L}_S \Xi_S - 2(\mathbf{L}_S : \Xi_S) \Xi_S,$$

where we use the solid-phase velocity gradient tensor,  $\mathbf{L}_S = (D_S \mathbf{F}_S / Dt) \mathbf{F}_S^{-1}$ , because of the assumption that fibre-like inclusions and matrix move with the same common velocity, *i.e.* the solid-phase velocity,  $\mathbf{v}_S$ . Furthermore, we find a modified form for the evolution equation (1.2), in which the growth velocity gradient of the fibre-like inclusion sub-phase is given by an Onsager relation, based on the exploitation of entropy production inequality of the mixture as a whole. This Onsager's relation expresses a direct connection between the growth velocity gradient, and the solid-phase Mandel stress tensor. We also

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<sup>(3)</sup> This equation is obtained by attaching the label “ $S$ ” to every quantity featuring in eq. (1.1), dyadic multiplication by  $\xi_S$  (once from the right, and once from the left), and adding the two resulting expressions. The symbol

$$\frac{D_S}{Dt} = \frac{\partial}{\partial t} + \mathbf{v}_S \cdot \nabla$$

is the convective derivative with respect to the solid-phase velocity,  $\mathbf{v}_S$ .

introduce a “*formal*” Mandel stress tensor associated with the fluid-phase, and we show that this tensor plays a role in Onsager’s relation determining the growth velocity gradient of the fibre-like inclusion sub-phase.

The determination of a thermodynamically consistent evolution law for the texture tensor was put forward, for example, by [21, 19]. For the thermodynamic study performed in our paper, we referred to [22].

## 2. – Model description

We consider a class of biological tissues that can be macroscopically modelled as mixtures composed of a fluid and a solid phase. The former is a multi-constituent fluid experiencing single-phase flow, and the latter is a deformable medium consisting of a porous matrix and fibre-like inclusions. We refer to matrix and fibre-like inclusions as to solid sub-phases. In the following, we denote the fluid phase by  $\mathcal{F}_F$ , and the solid phase by  $\mathcal{F}_S = \mathcal{F}_M \cup \mathcal{F}_I$ , where  $\mathcal{F}_M$  and  $\mathcal{F}_I$  represent the matrix, and inclusion sub-phase, respectively.

Following the picture proposed by Bennethum *et al.* [23], we assume that the fluid phase,  $\mathcal{F}_F$ , and the sub-phases  $\mathcal{F}_M$  and  $\mathcal{F}_I$  are mixtures on their own, which comprise the same number of constituents. Constituents 0-th and 1-st confer the sub-phases  $\mathcal{F}_M$  and  $\mathcal{F}_I$  the mechanical properties of a solid, respectively. The  $N$ -th constituent is identified with water, and all other constituents ( $\gamma = 2, \dots, N-1$ ) represent, for example, nutrients, byproducts of cellular metabolic reactions, chemical agents, and molecular species. The  $\alpha$ -th constituent in phase  $\mathcal{F}_j$  (with  $\alpha \in \{0, \dots, N\}$  and  $j \in \{F\} \cup \{M, I\}$ ) is denoted by  $\mathcal{C}_{\alpha j}$ . If a given constituent is present in one phase but absent in the other two phases, its mass exchange term is set identically equal to zero, and it is *formally* regarded as present with zero concentration.

We require that the mixture satisfies the *saturation condition*. According to this condition, the sum of solid- and fluid-phase volume fractions, denoted by  $\phi_F$  and  $\phi_S$ , respectively, is constrained to equal unity at all times and all points of the mixture, *i.e.*  $\phi_F + \phi_S = 1$ . We remark that, due to the presence of matrix and inclusions, the volume fraction of the solid phase as a whole,  $\phi_S$ , is actually defined by the sum of the volume fractions of the solid sub-phases  $\mathcal{F}_M$  and  $\mathcal{F}_I$ , *i.e.*  $\phi_S := \phi_M + \phi_I$ . Following [24], we define the mass density of fluid-phase,  $\mathcal{F}_j$  (with  $j \in \{F\} \cup \{M, I\}$ ), by the sum

$$(2.1) \quad \rho_j := \sum_{\alpha=0}^N \rho_{\alpha j},$$

where  $\rho_{\alpha j}$  (with  $\alpha \in \{0, \dots, N\}$ ) is the mass density of constituent  $\mathcal{C}_{\alpha j}$  in the phase  $\mathcal{F}_j$ . The mass fractions of constituents  $\mathcal{C}_{\alpha j}$  are defined by the ratios  $C_{\alpha j} := \rho_{\alpha j}/\rho_j$ , and are thus linearly dependent through the constraint  $\sum_{\alpha=0}^N C_{\alpha j} = 1$ . The mass density of the solid phase is given by  $\phi_S \rho_S = \phi_M \rho_M + \phi_I \rho_I$ , whereas densities  $\rho_M$  and  $\rho_I$  are defined as in eq. (2.1).

**2.1. General form of balance laws.** – With respect to each constituent  $\mathcal{C}_{\alpha j}$  in phase  $\mathcal{F}_j$  (with  $\alpha \in \{0, \dots, N\}$  and  $j \in \{F\} \cup \{M, I\}$ ), the macroscopic balance laws can be written in the following general form:

$$(2.2) \quad \frac{\partial(\phi_j \rho_j C_{\alpha j} \psi_{\alpha j})}{\partial t} + \nabla \cdot (\phi_j \rho_j C_{\alpha j} \psi_{\alpha j} \mathbf{v}_{\alpha j}) + \nabla \cdot \Phi_{\alpha j} - \phi_j \rho_j C_{\alpha j} \mathcal{F}_{\alpha j} = \phi_j \rho_j C_{\alpha j} \mathcal{G}_{\alpha j}.$$

TABLE I. – *Thermodynamic quantities and related fluxes to be substituted in eq. (2.2).*

Quantity	$\psi_{\alpha j}$	$\Phi_{\alpha j}$
Mass	1	$\mathbf{0}$
Momentum	$\mathbf{v}_{\alpha j}$	$-\boldsymbol{\sigma}_{\alpha j}$
Energy	$E_{\alpha j} + \frac{1}{2}\mathbf{v}_{\alpha j}^2$	$-(\boldsymbol{\sigma}_{\alpha j} \cdot \mathbf{v}_{\alpha j} + \mathbf{q}_{\alpha j})$
Entropy	$S_{\alpha j}$	$-\Theta^{-1}\mathbf{q}_{\alpha j}$

Here,  $\mathbf{v}_{\alpha j}$  is the velocity, while  $\Phi_{\alpha j}$ ,  $\mathcal{F}_{\alpha j}$ , and  $\mathcal{G}_{\alpha j}$  denote *flux*, net production (or decay), and source (or sink) of the generic thermodynamic quantity  $\psi_{\alpha j}$  associated with constituent  $\mathcal{C}_{\alpha j}$ . Since eq. (2.2) is a compact way of writing balance of mass, momentum, energy, and entropy, quantity  $\psi_{\alpha j}$  may represent either a scalar or a vector field. In the case of balance of momentum,  $\psi_{\alpha j}$  is identified with  $\mathbf{v}_{\alpha j}$ , quantities  $\mathcal{F}_{\alpha j}$  and  $\mathcal{G}_{\alpha j}$  are vector fields, flux  $\Phi_{\alpha j}$  is a second-order tensor, and the product  $\psi_{\alpha j}\mathbf{v}_{\alpha j}$  on the LHS of eq. (2.2) is understood as the dyadic product  $\psi_{\alpha j}\mathbf{v}_{\alpha j} = \mathbf{v}_{\alpha j} \otimes \mathbf{v}_{\alpha j}$ . Balance laws are obtained in specific form by substituting the quantities in tables I and II into eq. (2.2). In tables I and II,  $E_{\alpha j}$  is the internal energy density,  $\boldsymbol{\sigma}_{\alpha j}$  is the Cauchy stress tensor,  $\mathbf{q}_{\alpha j}$  is the heat flux vector,  $\Theta$  is the absolute temperature,  $S_{\alpha j}$  is the entropy density,  $\mathbf{g}$  is the gravity acceleration vector, and  $R_{\alpha j}$ ,  $\mathbf{T}_{\alpha j}$ ,  $Q_{\alpha j}$ , and  $\eta_{\alpha j}$  are sources (or sinks) of mass, momentum, energy, and entropy due to exchange interactions among constituents. Finally, in table II,  $Q_{\alpha j}^*$  and  $\eta_{\alpha j}^*$  represent a source of internal energy, and entropy related to remodelling and growth. Although here a rather general formalism has been used, these last two quantities are referred only to the solid phase. Therefore, it is understood that  $Q_{\alpha F}^* = 0$ , and  $\eta_{\alpha F}^* = 0$  for all fluid-phase constituents  $\mathcal{C}_{\alpha F}$  ( $\alpha \in \{0, \dots, N\}$ ). To our knowledge, similar terms were introduced, for example, by Menzel [6].

**2.2. Source terms.** – Although recent publications present theoretical settings more general than that described in the present paper (cf., for example, [20, 25, 16]), for the sake of simplicity we assume here that constituents are allowed to undergo exchange interactions among each other, but neither chemical reactions nor intrinsic sources (or sinks) of thermodynamic quantities are considered, except for the quantities  $Q_{\alpha j}^*$  and  $\eta_{\alpha j}^*$ , which represent remodelling and growth. Therefore, in our framework, the mixture is closed with respect to all sources (or sinks) related to exchange processes but open with respect to remodelling, and the interaction of the mixture with the surrounding world is described by supplying balance equations with appropriate boundary conditions. This requirement implies that source (or sink) terms related to exchange processes satisfy constraints such that no mass, momentum and energy is produced in the mixture as

TABLE II. – *Net production (decay) and source (sink) terms to be substituted in eq. (2.2).*

Quantity	$\mathcal{F}_{\alpha j}$	$\mathcal{G}_{\alpha j}$
Mass	0	$R_{\alpha j}$
Momentum	$\mathbf{g}$	$R_{\alpha j}\mathbf{v}_{\alpha j} + \mathbf{T}_{\alpha j}$
Energy	$\mathbf{g} \cdot \mathbf{v}_{\alpha j} + h_{\alpha j}$	$R_{\alpha j}(E_{\alpha j} + \frac{1}{2}\mathbf{v}_{\alpha j}^2) + \mathbf{T}_{\alpha j} \cdot \mathbf{v}_{\alpha j} + Q_{\alpha j} + Q_{\alpha j}^*$
Entropy	$\Theta^{-1}h_{\alpha j}$	$\Gamma_{\alpha j} + R_{\alpha j}S_{\alpha j} + \eta_{\alpha j} + \eta_{\alpha j}^*$

a whole. Mathematically, this condition is expressed by imposing that the sum of a given source term over all phases is zero. In order to visualise the statement given above, we first define, for each  $\mathcal{F}_j$  (with  $j \in \{F\} \cup \{M, I\}$ ), the following overall source terms: *Mass*

$$(2.3) \quad R_j := \sum_{\alpha=0}^N C_{\alpha j} R_{\alpha j},$$

*Momentum*

$$(2.4) \quad \mathbf{T}_j := \sum_{\alpha=0}^N C_{\alpha j} (R_{\alpha j} \mathbf{u}_{\alpha j} + \mathbf{T}_{\alpha j}),$$

*Energy*

$$(2.5) \quad Q_j := \sum_{\alpha=0}^N C_{\alpha j} \left[ Q_{\alpha j} + \mathbf{T}_{\alpha j} \cdot \mathbf{u}_{\alpha j} + R_{\alpha j} \left( E_{\alpha j} - E_j + \frac{1}{2} \mathbf{u}_{\alpha j}^2 \right) \right],$$

*Entropy*

$$(2.6) \quad \eta_j := \sum_{\alpha=0}^N C_{\alpha j} [\eta_{\alpha j} + R_{\alpha j} (S_{\alpha j} - S_j)],$$

where  $\mathbf{u}_{\alpha j} := \mathbf{v}_{\alpha j} - \mathbf{v}_j$  is the diffusive velocity of constituent  $C_{\alpha j}$  in phase  $\mathcal{F}_j$  with respect to the phase average velocity  $\mathbf{v}_j := \sum_{\alpha=0}^N C_{\alpha j} \mathbf{v}_{\alpha j}$ ,  $E_j := \sum_{\alpha=0}^N C_{\alpha j} (E_{\alpha j} + \frac{1}{2} \mathbf{u}_{\alpha j}^2)$  is the internal energy density, and  $S_j := \sum_{\alpha=0}^N C_{\alpha j} S_{\alpha j}$  is the entropy density of phase  $\mathcal{F}_j$ .

By virtue of eqs. (2.3)–(2.5), the fact that the mixture is closed is expressed by requiring

$$(2.7) \quad 0 = \sum_{j \in \{F, M, I\}} \phi_j \rho_j R_j,$$

$$(2.8) \quad \mathbf{0} = \sum_{j \in \{F, M, I\}} \phi_j \rho_j (\mathbf{T}_j + R_j \mathbf{v}_j),$$

$$(2.9) \quad 0 = \sum_{j \in \{F, M, I\}} \phi_j \rho_j \left[ Q_j + \mathbf{T}_j \cdot \mathbf{v}_j + R_j \left( E_j + \frac{1}{2} \mathbf{v}_j^2 \right) \right],$$

$$(2.10) \quad 0 \leq \sum_{j \in \{F, M, I\}} \phi_j \rho_j (\eta_j + R_j S_j).$$

Quantities  $Q_{\alpha j}^*$  and  $\eta_{\alpha j}^*$  cannot feature in the closure relations given above because they are not associated with any exchange process. Equation (2.10) is satisfied as an equality when, for example, the fluid-solid interface is assumed to be ideal in the sense that *no* entropy generation in the fluid-solid interface is accounted for. Equations (2.7)–(2.10) represent averaged thermodynamic exchange interactions occurring at the interface between the fluid and the solid phase.

### 3. – Second principle of thermodynamics

In order to study the expression of entropy production according to the Coleman-Noll method, we introduce the Helmholtz free-energy density of constituent  $\mathcal{C}_{\alpha j}$  (with  $\alpha \in \{0, \dots, N\}$  and  $j \in \{F\} \cup \{M, I\}$ ), *i.e.*  $A_{\alpha j} := E_{\alpha j} - \Theta S_{\alpha j}$ . By substituting the Helmholtz free energy densities,  $A_{\alpha j}$ , into the balance of entropy (this balance law is obtained by using the quantities in tables I and II in eq. (2.2)), and under the hypothesis that the mixture undergoes only isothermal processes, the expression of entropy production for constituent  $\mathcal{C}_{\alpha j}$  can be written as

$$(3.1) \quad \Theta \phi_j \rho_j C_{\alpha j} \Gamma_{\alpha j} = -\phi_j \rho_j C_{\alpha j} \frac{D_{\alpha j} A_{\alpha j}}{Dt} - \phi_j \rho_j C_{\alpha j} S_{\alpha j} \frac{D_{\alpha j} \Theta}{Dt} + \boldsymbol{\sigma}_{\alpha j} : \nabla \mathbf{v}_{\alpha j} \\ + \phi_j \rho_j C_{\alpha j} R_{\alpha j} \Theta S_{\alpha j} + \phi_j \rho_j C_{\alpha j} Q_{\alpha j} - \phi_j \rho_j C_{\alpha j} \Theta (R_{\alpha j} S_{\alpha j} + \eta_{\alpha j}) \\ + \phi_j \rho_j C_{\alpha j} (Q_{\alpha j}^* - \Theta \eta_{\alpha j}^*),$$

where  $D_{\alpha j}/Dt$  is the *convective* derivative with respect to the motion of constituent  $\mathcal{C}_{\alpha j}$ <sup>(4)</sup>. The fourth and the sixth term on the RHS of eq. (3.1) could be combined so to obtain the quantity  $\phi_j \rho_j C_{\alpha j} (Q_{\alpha j} - \Theta \eta_{\alpha j})$ . If such a simplification were done, only the rate of entropy production due to the exchange interactions  $Q_{\alpha j}$  and  $\eta_{\alpha j}$  would feature in eq. (3.1), and the entropic contribution due to the mass transfer of constituent  $\mathcal{C}_{\alpha j}$ ,  $R_{\alpha j} S_{\alpha j}$ , would be eliminated. Although the study of the dissipation inequality should not be affected by such an alternative way of writing  $\Gamma_{\alpha j}$ , we use eq. (3.1) because it allows for keeping track of the exchange of entropy among constituents which, at the pore-level, occurs at the fluid-solid interface.

Summation of eq. (3.1) over all constituents  $\mathcal{C}_{\alpha j}$  (with  $\alpha \in \{0, \dots, N\}$ ) leads to the expression of entropy production of phase  $\mathcal{F}_j$ ,  $\Gamma_j := \sum_{\alpha=0}^N C_{\alpha j} \Gamma_{\alpha j}$  ( $j \in \{F\} \cup \{M, I\}$ ), *i.e.*

$$(3.2) \quad \Theta \phi_j \rho_j \Gamma_j = -\phi_j \rho_j \frac{D_j A_j}{Dt} - \phi_j \rho_j S_j \frac{D_j \Theta}{Dt} - \sum_{\alpha=0}^N \nabla \cdot (\phi_j \rho_j C_{\alpha j} A_{\alpha j} \mathbf{u}_{\alpha j}) \\ + \sum_{\alpha=0}^N \phi_j \rho_j C_{\alpha j} R_{\alpha j} (A_{\alpha j} - A_j) + \sum_{\alpha=0}^N \boldsymbol{\sigma}_{\alpha j} : \nabla \mathbf{u}_{\alpha j} \\ + \left[ \boldsymbol{\sigma}_j + \sum_{\alpha=0}^N \phi_j \rho_j C_{\alpha j} \mathbf{u}_{\alpha j} \otimes \mathbf{u}_{\alpha j} \right] : \nabla \mathbf{v}_j + \sum_{\alpha=0}^N \phi_j \rho_j C_{\alpha j} R_{\alpha j} \Theta S_{\alpha j} \\ + \sum_{\alpha=0}^N \phi_j \rho_j C_{\alpha j} Q_{\alpha j} - \sum_{\alpha=0}^N \phi_j \rho_j C_{\alpha j} \Theta (R_{\alpha j} S_{\alpha j} + \eta_{\alpha j}) \\ + \phi_j \rho_j (Q_j^* - \Theta \eta_j^*).$$

In eq. (3.2), the symbol  $D_j/Dt$  denotes the *convective* derivative with respect to the

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<sup>(4)</sup> In explicit form, the operator  $D_{\alpha j}/Dt$  is given by

$$\frac{D_{\alpha j}}{Dt} = \frac{\partial}{\partial t} + \mathbf{v}_{\alpha j} \cdot \nabla.$$



motion of phase  $\mathcal{F}_j$ <sup>(5)</sup>, and the notation  $Q_j^* := \sum_{\alpha=0}^N C_{\alpha j} Q_{\alpha j}^*$ , and  $\eta_j^* := \sum_{\alpha=0}^N C_{\alpha j} \eta_{\alpha j}^*$  has been introduced. Moreover,  $A_j := \sum_{\alpha=0}^N C_{\alpha j} A_{\alpha j}$ ,  $S_j := \sum_{\alpha=0}^N C_{\alpha j} S_{\alpha j}$ , and  $\boldsymbol{\sigma}_j := \sum_{\alpha=0}^N [\boldsymbol{\sigma}_{\alpha j} - \phi_j \rho_j C_{\alpha j} \mathbf{u}_{\alpha j} \otimes \mathbf{u}_{\alpha j}]$  are the Helmholtz free-energy density, entropy, and Cauchy stress tensor of phase  $\mathcal{F}_j$ , respectively.

**3.1. Reduced entropy inequality.** – The expression of entropy production for the mixture as a whole is obtained by summing eq. (3.2) over all phases  $\mathcal{F}_j$  (with  $j \in \{F\} \cup \{I, M\}$ ). In order to do that, we first need to compute the sum  $\sum_{j \in \{F, I, M\}} \sum_{\alpha=0}^N \phi_j \rho_j C_{\alpha j} Q_{\alpha j}$ . For the sake of simplicity, we hypothesise here that: i) in the solid sub-phases  $\mathcal{F}_I$  and  $\mathcal{F}_M$ , constituents  $C_{\alpha I}$  and  $C_{\alpha M}$  (with  $\alpha \in \{0, \dots, N\}$ ) have *no* diffusive velocity (*i.e.*  $\mathbf{u}_{\alpha I} \equiv \mathbf{0}$ , and  $\mathbf{u}_{\alpha M} \equiv \mathbf{0}$ ,  $\forall \alpha \in \{0, \dots, N\}$ ), and ii) that sub-phases  $\mathcal{F}_I$  and  $\mathcal{F}_M$  move with the same velocity, *i.e.*  $\mathbf{v}_I \equiv \mathbf{v}_M \equiv \mathbf{v}_S$ , where  $\mathbf{v}_S$  is thus the velocity of the solid-phase as a whole. By using these assumptions, definitions (2.3)–(2.5), and restrictions (2.7)–(2.9), the sum  $\sum_{j \in \{F, I, M\}} \sum_{\alpha=0}^N \phi_j \rho_j C_{\alpha j} Q_{\alpha j}$  reads

$$(3.3) \quad \begin{aligned} & \sum_{\alpha=0}^N \phi_I \rho_I C_{\alpha I} Q_{\alpha I} + \sum_{\alpha=0}^N \phi_M \rho_M C_{\alpha M} Q_{\alpha M} + \sum_{\alpha=0}^N \phi_F \rho_F C_{\alpha F} Q_{\alpha F} = \\ & -\phi_F \rho_F \mathbf{T}_F \cdot \mathbf{w}_{FS} - \phi_F \rho_F R_F \frac{1}{2} \mathbf{w}_{FS}^2 - \sum_{\alpha=0}^N \phi_F \rho_F C_{\alpha F} \mathbf{T}_{\alpha F} \cdot \mathbf{u}_{\alpha F} \\ & - \sum_{\alpha=0}^N \phi_I \rho_I C_{\alpha I} R_{\alpha I} E_{\alpha I} - \sum_{\alpha=0}^N \phi_M \rho_M C_{\alpha M} R_{\alpha M} E_{\alpha M} \\ & - \sum_{\alpha=0}^N \phi_F \rho_F C_{\alpha F} R_{\alpha F} \left[ E_{\alpha F} + \frac{1}{2} \mathbf{u}_{\alpha F}^2 \right]. \end{aligned}$$

We notice that the sum over all phases  $\mathcal{F}_j$  (with  $j \in \{F\} \cup \{I, M\}$ ) of the second last term on the RHS of eq. (3.2) gives the averaged form of the overall entropy production due to exchange processes occurring at the fluid-solid interface. This entropy production is zero if eq. (2.10) is satisfied as an equality [23]. We assume that this is the case, and, by making use of eq. (3.3), hypotheses (i) and (ii), and eqs. (2.3)–(2.5) and (2.7)–(2.9), we define a *reduced* form of overall entropy production,  $\Gamma_{\text{red}}$ , such that

$$(3.4) \quad \begin{aligned} \Theta \rho \Gamma_{\text{red}} = & -\phi_S \rho_S \frac{D_S A_S}{Dt} - \phi_F \rho_F \frac{D_F A_F}{Dt} - \phi_S \rho_S S_S \frac{D_S \Theta}{Dt} - \phi_F \rho_F S_F \frac{D_F \Theta}{Dt} \\ & + \sum_{\alpha=0}^N [\boldsymbol{\sigma}_{\alpha F} - \phi_F \rho_F C_{\alpha F} A_{\alpha F} \mathbf{I}] : \nabla \mathbf{u}_{\alpha F} \end{aligned}$$

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<sup>(5)</sup> In explicit form, the operator  $D_j/Dt$  is given by

$$\frac{D_j}{Dt} := \frac{\partial}{\partial t} + \mathbf{v}_j \cdot \nabla.$$

$$\begin{aligned}
& - \sum_{\alpha=0}^N [\nabla(\phi_F \rho_F C_{\alpha F} A_{\alpha F}) + \phi_F \rho_F C_{\alpha F} \mathbf{T}_{\alpha F}] \cdot \mathbf{u}_{\alpha F} \\
& + \left[ \boldsymbol{\sigma}_F + \sum_{\alpha=0}^N \phi_F \rho_F C_{\alpha F} \mathbf{u}_{\alpha F} \otimes \mathbf{u}_{\alpha F} \right] : \nabla \mathbf{v}_F + \boldsymbol{\sigma}_S : \nabla \mathbf{v}_S \\
& - \phi_F \rho_F \mathbf{T}_F \cdot \mathbf{w}_{FS} - \phi_S \rho_S R_S \left[ A_S - A_F - \frac{1}{2} \mathbf{w}_{FS}^2 \right] + \phi_S \rho_S A_S^* \\
& - \sum_{\alpha=0}^N \phi_F \rho_F C_{\alpha F} R_{\alpha F} \frac{1}{2} \mathbf{u}_{\alpha F}^2 \geq 0,
\end{aligned}$$

where  $\rho := \sum_{j \in \{F, I, M\}} \phi_j \rho_j$  is the mass density of the mixture,  $\phi_S \rho_S A_S := \sum_{j \in \{I, M\}} \phi_j \rho_j A_j$  and  $\phi_S \rho_S S_S := \sum_{j \in \{I, M\}} \phi_j \rho_j S_j$  are the Helmholtz free-energy density and entropy density of the solid-phase  $\mathcal{F}_S$  (*i.e.*  $\mathcal{F}_S = \mathcal{F}_I \cup \mathcal{F}_M$ ), respectively,  $\mathbf{w}_{FS} := \mathbf{v}_F - \mathbf{v}_S$  is the fluid-phase *filtration velocity*, and  $\phi_S \rho_S A_S^* := \sum_{k=I, M} \phi_k \rho_k (Q_k^* - \Theta \eta_k^*)$  is the rate of dissipation due to remodelling and growth.

**3.2. Lagrange multiplier method.** – If the mixture is subject to constraints, these constraints should be accounted for when the expression of entropy production (in our case, eq. (3.4)) is exploited. A possible way of doing that consists in the application of the *Lagrange multiplier technique*: each constraint is multiplied by an appropriate Lagrange multiplier, and the resulting expression is combined with the expression of entropy production, in order to obtain a modified form of the entropy inequality. The reader is referred to the works by Liu [26], Liu and Müller [27], and Müller and Ruggeri [28] for details. The procedure used in our paper is based on the papers [23] and [16]. For our purposes, we adopt as constraints the balance of mass of constituents  $\mathcal{C}_{\beta j}$  (with  $\beta \in \{0, \dots, N-1\}$  and  $j \in \{F\} \cup \{I, M\}$ ), the balance of mass of all phases  $\mathcal{F}_j$  (with  $j \in \{F\} \cup \{I, M\}$ ) (*cf.*, for example, [19]), and the requirement that the weighted sum of all diffusive velocities of fluid constituents  $\mathcal{C}_{\alpha F}$  ( $\alpha \in \{0, \dots, N\}$ ) is null, *i.e.*  $\sum_{\alpha=0}^N C_{\alpha F} \mathbf{u}_{\alpha F} = \mathbf{0}$ . The modified expression of entropy production reads

$$\begin{aligned}
(3.5) \quad & \Theta \rho \widetilde{\Gamma}_{\text{red}} = \Theta \rho \Gamma_{\text{red}} \\
& + \pi \left\{ \frac{\phi_I}{\rho_I} \frac{D_S \rho_I}{Dt} + \frac{\phi_M}{\rho_M} \frac{D_S \rho_M}{Dt} + \frac{\phi_F}{\rho_F} \frac{D_S \rho_F}{Dt} + \mathbf{w}_{FS} \cdot \nabla \phi_F + \frac{\phi_F}{\rho_F} \mathbf{w}_{FS} \cdot \nabla \rho_F \right. \\
& \left. + \phi_S \nabla \cdot \mathbf{v}_S + \phi_F \nabla \cdot \mathbf{v}_F - \phi_I R_I - \phi_M R_M - \phi_F R_F \right\} \\
& + \sum_{k \in \{I, M\}} \sum_{\beta=0}^{N-1} \lambda_{\beta k} \left\{ \phi_k \rho_k \frac{D_S C_{\beta k}}{Dt} - \phi_k \rho_k C_{\beta k} (R_{\beta k} - R_k) \right\} \\
& + \sum_{\beta=0}^{N-1} \lambda_{\beta F} \left\{ \phi_F \rho_F \frac{D_F C_{\beta F}}{Dt} + \nabla \cdot (\phi_F \rho_F C_{\beta F} \mathbf{u}_{\beta F}) - \phi_F \rho_F C_{\beta F} (R_{\beta F} - R_F) \right\} \\
& + \phi_F \boldsymbol{\Lambda}_F : \sum_{\alpha=0}^N \nabla (\rho_F C_{\alpha F} \mathbf{u}_{\alpha F}) \geq 0.
\end{aligned}$$

The quantities  $\pi$ ,  $\lambda_{\beta k}$  (where  $\beta \in \{0, \dots, N-1\}$  and  $k \in \{I, M\}$ ), and  $\Lambda_F$  are Lagrange multipliers. Their determination follows from the study of eq. (3.5) according to Coleman-Noll method.

We remark that, because of the definition of the solid-phase volume fraction,  $\phi_S := \phi_I + \phi_M$ , and the saturation condition,  $\phi_F + \phi_S = 1$ , the summation of the mass balance laws of all phases implies that no convective derivative of volume fraction,  $\phi_S$  (or  $\phi_F$ ), can feature in eq. (3.5). Another consequence of the approach followed in the present paper is that, since the summation of the mass balance laws of all phases leads to a single balance of mass for the mixture as a whole, the enforcement of the resulting balance law as a constraint for  $\Theta\rho\Gamma$  requires the introduction of a single Lagrange multiplier, *i.e.*  $\pi$ . Although this approach differs from the approach followed in [16], it is probably more appropriate for the development of the following theory.

#### 4. – Constitutive framework

In order to close the field equations to be solved, a constitutive framework has to be introduced. This is done by selecting a set of *independent constitutive variables* (ICV), and treating the remaining unknowns as *dependent constitutive variables* (DCV). In the context of mixtures, however, this procedure alone does not usually provide conditions for volume fractions. Bennethum *et al.* [23] pointed out that this problem of closure is due to the loss of information in the averaging process that leads from the pore-scale analysis of the mixture to the macroscopic field equations. The solution to this problem can be obtained by following different approaches. In the work by Wilmański [29], a thermodynamic model of compressible porous materials is presented with a balance law for porosity, whereas in the work by Sciarra *et al.* [30], volume fraction is regarded as a microstructural parameter that is introduced so to enlarge the space of admissible deformations with respect to the classical theory of mixtures. More frequently, additional conditions on the mass density of phases are imposed. For example, in the case of biphasic mixtures, it is rather customary to assume that the solid phase (or the fluid phase) is *intrinsically incompressible*<sup>(6)</sup>. This assumption can be relaxed if the solid phase is modelled as a mixture of solid sub-phases [19]. Indeed, even though the mass density of each sub-phase is assumed to be constant, the mass density of the solid-phase as a whole does not need to be constant because volume fractions of solid sub-phases are allowed to change in space and time. In our paper, however, we do not assume the incompressibility of the fluid and the solid sub-phase. Rather, we assume that the mass densities of the fluid phase,  $\mathcal{F}_F$ , and the solid sub-phases,  $\mathcal{F}_I$  and  $\mathcal{F}_M$ , are functions of the mass fractions,  $C_{\beta j}$  (with  $\beta \in \{0, \dots, N-1\}$ , and  $j \in \{F, I, M\}$ ).

**4.1. Growth.** – Following the picture proposed by Epstein and Maugin [13], growth is here viewed as a process causing the development of material inhomogeneities in the reference configuration of a body due to the presence of mass sources, or sinks, acting inside

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<sup>(6)</sup> The  $j$ -th phase of a mixture is said to be incompressible, if the convective derivative of its mass density is zero, *i.e.*

$$\frac{D_j \rho_j}{Dt} = 0.$$

In the case of mixtures whose phases exchange mass, the assumption that the solid phase is incompressible does not necessarily imply that  $J_S = \det(\mathbf{F}_S) = 1$ , where  $\mathbf{F}_S$  is the deformation gradient tensor of the solid phase.

the body itself. Since, in the case of surface (or appositional) growth, a reference configuration for the body is not defined (cf., for example, [19] for explanation), we consider here volumetric (or interstitial) growth only. Growth is thus assumed to be responsible for inducing the time variation of the mass density of the body in its reference configuration. Material inhomogeneities<sup>(7)</sup> are related to the incompatibility of deformation arising as growth occurs (the reader is referred, for example, to the work by Rodriguez *et al.* [31] for explanation, and the connection of incompatible deformation with residual stresses). The anelastic deformation induced by growth is described through Kröner's *incompatibility method*, which consists of decomposing the deformation gradient tensor into the product of an elastic and an anelastic contribution.

In order to define a reference configuration for the mixture, we adhere to Biot's approach [32], and the framework proposed by Quiligotti [33], and Quiligotti *et al.* [34]. Accordingly, the solid phase is taken as the "control" phase to which a reference configuration is associated. In the study presented in our paper, growth (or adsorption) is described as a process concerning the solid sub-phases of the tissue,  $\mathcal{F}_I$  and  $\mathcal{F}_M$ , as a result of the mass exchange between the fluid-phase,  $\mathcal{F}_F$ , and sub-phases  $\mathcal{F}_I$  and  $\mathcal{F}_M$ . Although we have assumed that solid sub-phases  $\mathcal{F}_I$  and  $\mathcal{F}_M$  move with the common velocity  $\mathbf{v}_S$  (which is said to be the solid-phase velocity), growth may occur independently. Following [20], this is accounted for by assuming that, according to Kröner's method, the solid-phase deformation gradient tensor,  $\mathbf{F}_S$ , admits the multiplicative decomposition

$$(4.1) \quad \mathbf{F}_S = \mathbf{F}_I^e \mathbf{F}_I^{\text{an}} = \mathbf{F}_M^e \mathbf{F}_M^{\text{an}},$$

where  $\mathbf{F}_I^e$  and  $\mathbf{F}_M^e$  measure the true elastic deformation of the inclusions and matrix, respectively, while  $\mathbf{F}_I^{\text{an}}$  and  $\mathbf{F}_M^{\text{an}}$  describe the *anelastic* part of deformation related to the production of material inhomogeneities due to growth in sub-phases  $\mathcal{F}_I$  and  $\mathcal{F}_M$ , respectively. Each tensor  $\mathbf{F}_k^{\text{an}}$  (with  $k \in \{I, M\}$ ) maps the tangent space of the reference configuration of the mixture into the tangent space of an intermediate, elastically released configuration, which is referred to as to the *natural configuration* of sub-phase  $\mathcal{F}_k$  (with  $k \in \{I, M\}$ ). The quantities  $\mathbf{F}_S$ ,  $\mathbf{F}_I^{\text{an}}$ , and  $\mathbf{F}_M^{\text{an}}$  should be treated as free unknowns.

**4.2. Remodelling.** – In order to account for the presence of inclusions, and the possibility of remodelling, we introduce a *unit* vector  $\boldsymbol{\lambda}_I^N$ , which represents the alignment of fibre-like inclusions in the natural configuration of phase  $\mathcal{F}_I$ . An alternative description of anisotropy can be given by the texture tensor  $\boldsymbol{\Xi}_I^N := \boldsymbol{\lambda}_I^N \otimes \boldsymbol{\lambda}_I^N$ . Unit vectors  $\boldsymbol{\lambda}_I^N$  and  $\boldsymbol{\xi}_S$ , and texture tensors  $\boldsymbol{\Xi}_I^N$  and  $\boldsymbol{\Xi}_S$  are related to the each other through

$$(4.2) \quad \boldsymbol{\xi}_S = \frac{1}{\sqrt{\text{Tr} [\mathbf{F}_I^e (\boldsymbol{\lambda}_I^N \otimes \boldsymbol{\lambda}_I^N) (\mathbf{F}_I^e)^T]}} \mathbf{F}_I^e \boldsymbol{\lambda}_I^N,$$

$$(4.3) \quad \boldsymbol{\Xi}_S = \frac{1}{\text{Tr} [\mathbf{F}_I^e \boldsymbol{\Xi}_I^N (\mathbf{F}_I^e)^T]} \mathbf{F}_I^e \boldsymbol{\Xi}_I^N (\mathbf{F}_I^e)^T.$$

According to eqs. (4.2) and (4.3), the alignment of fibre-like inclusions in the current configuration, described by  $\boldsymbol{\xi}_S$  (or  $\boldsymbol{\Xi}_S$ ), can be obtained if the fibre arrangement in the natural configuration of phase  $\mathcal{F}_I$ , described by  $\boldsymbol{\lambda}_I^N$  (or  $\boldsymbol{\Xi}_I^N$ ), is known.

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<sup>(7)</sup> According to Noll's terminology, by inhomogeneities we mean here distortions.

When remodelling occurs, the change of tissue anisotropy is described by an evolution law for either the unit vector  $\boldsymbol{\lambda}_I^N$ , or the texture tensor  $\boldsymbol{\Xi}_I^N$ . Therefore, vector  $\boldsymbol{\lambda}_I^N$  (or tensor  $\boldsymbol{\Xi}_I^N$ ) has to be treated as a free unknown for the tissue. However, since the unit vector  $\boldsymbol{\lambda}_I^N$  is constrained by the relation  $\|\boldsymbol{\lambda}_I^N\| = 1$ , it may be convenient to select as independent free unknowns only the angles  $\vartheta$  and  $\varphi$  such that  $\boldsymbol{\lambda}_I^N = \sin(\varphi) \cos(\vartheta) \mathbf{e}_1 + \sin(\varphi) \sin(\vartheta) \mathbf{e}_2 + \cos(\varphi) \mathbf{e}_3$ , where  $\{\mathbf{e}_J\}_{J=1}^3$  are the basis unit vectors of the three-dimensional Euclidean space.

**4.3. Unknowns.** – We count the unknowns featuring in the modified expression of entropy production (cf. eq. (3.5)). In particular, we split these unknowns in three sets, *i.e.* *free unknowns*,  $\mathcal{U}_{\text{free}}$ , *dependent unknowns*,  $\mathcal{U}_{\text{dep}}$ , and Lagrange multipliers,  $\mathcal{M}$ . Therefore, we conclude that the set of free and dependent unknowns, and Lagrange multipliers are given by

$$(4.4) \quad \mathcal{U}_{\text{free}} = \{\phi_I, \phi_M, C_{\beta I}, C_{\beta M}, C_{\beta F}, \mathbf{u}_{\beta F}, \mathbf{w}_{FS}, \Theta, \varphi, \vartheta, \mathbf{F}_I^e, \mathbf{F}_M^e, \mathbf{F}_I^{\text{an}}, \mathbf{F}_M^{\text{an}}\},$$

$$(4.5) \quad \mathcal{U}_{\text{dep}} = \{A_S, A_F, S_S, S_F, \boldsymbol{\sigma}_{\beta F}, A_{\beta F}, \mathbf{T}_{\beta F}, \boldsymbol{\sigma}_F, \boldsymbol{\sigma}_S, \mathbf{T}_F, R_S, R_{\beta F}, \rho_I, \rho_M, \rho_F\},$$

$$(4.6) \quad \mathcal{M} = \{\pi, \lambda_{\beta I}, \lambda_{\beta M}, \lambda_{\beta F}, \boldsymbol{\Lambda}_F\}.$$

For the exploitation of the expression of entropy production (3.5), quantities  $\mathbf{F}_I^e$  and  $\mathbf{F}_M^e$  are replaced by the Green-Lagrange strain tensors  $\mathbf{E}_I^e := \frac{1}{2}[(\mathbf{F}_I^e)^T \mathbf{F}_I^e - \mathbf{I}_I^N]$  and  $\mathbf{E}_M^e := \frac{1}{2}[(\mathbf{F}_M^e)^T \mathbf{F}_M^e - \mathbf{I}_M^N]$  (where  $\mathbf{I}_I^N$  and  $\mathbf{I}_M^N$  are the identity tensors in the natural configuration of  $\mathcal{F}_I$  and  $\mathcal{F}_M$ , respectively), while quantities  $\mathbf{F}_I^{\text{an}}$  and  $\mathbf{F}_M^{\text{an}}$  are replaced by the *inhomogeneities velocity gradients*  $\mathbf{L}_I^{\text{an}}$  and  $\mathbf{L}_M^{\text{an}}$ , defined by

$$(4.7) \quad \mathbf{L}_I^{\text{an}} := \frac{D_S \mathbf{F}_I^{\text{an}}}{Dt} (\mathbf{F}_I^{\text{an}})^{-1}, \quad \text{and} \quad \mathbf{L}_M^{\text{an}} := \frac{D_S \mathbf{F}_M^{\text{an}}}{Dt} (\mathbf{F}_M^{\text{an}})^{-1}.$$

Quantities featuring in balance laws, but not present in the lists above, are related to the above-listed variables through either kinematic restrictions or their own definition. We remark that fluid-phase mass density is taken as dependent unknown (cf. eq. (4.5)) because we assume that it is prescribed as a constitutive function of mass fractions of fluid constituents. Furthermore, solid-phase velocity,  $\mathbf{v}_S$ , does not feature in eq. (4.4) because it has been replaced by deformation gradient tensor,  $\mathbf{F}_S$ , and then by tensors  $\mathbf{F}_I^e$ ,  $\mathbf{F}_M^e$ ,  $\mathbf{F}_I^{\text{an}}$ , and  $\mathbf{F}_M^{\text{an}}$  through eq. (4.1).

**4.4. Helmholtz free-energy densities.** – In our constitutive framework, the set of independent constitutive variables (ICV) is obtained through the union of the set of free unknowns,  $\mathcal{U}_{\text{free}}$ , and a set of variables containing the gradients of volume and mass fractions, and the *inhomogeneity velocity gradients*,  $\mathbf{L}_I^{\text{an}}$  and  $\mathbf{L}_M^{\text{an}}$ , while the set of dependent constitutive variables (DCV) is identified with the set of dependent unknowns,  $\mathcal{U}_{\text{dep}}$ , *i.e.*

$$(4.8) \quad \text{ICV} = \mathcal{U}_{\text{free}} \cup \left\{ \nabla C_{\beta F}, \nabla C_{\beta I}, \nabla C_{\beta M}, \frac{D_S \varphi}{Dt}, \frac{D_S \vartheta}{Dt} \right\},$$

$$(4.9) \quad \text{DCV} = \{A_S, A_F, S_S, S_F, \boldsymbol{\sigma}_{\beta F}, A_{\beta F}, \mathbf{T}_{\beta F}, \boldsymbol{\sigma}_F, \boldsymbol{\sigma}_S, \mathbf{T}_F, R_S, R_{\beta F}, \rho_I, \rho_M, \rho_F\},$$

where  $\beta \in \{0, \dots, N-1\}$ .

If the Axiom of equipresence were rigorously applied, the Helmholtz free-energy densities,  $A_F$  and  $A_S$  should be given as constitutive functions of all ICV's. However, it is possible to show that letting  $A_F$  and  $A_S$  depend only on a subset of ICV minimizes

the algebraic calculations necessary for our purposes without leading to thermodynamic inconsistencies. For this reason, we assume here that

$$(4.10) \quad A_F := \widehat{A}_F(\rho_F, \mathcal{C}_F, \Theta), \quad \text{and} \quad A_S := \widehat{A}_S(\rho_I, \rho_M, \mathcal{C}_I, \mathcal{C}_M, \Theta, \mathbf{F}, \mathbf{F}_I^{\text{an}}, \mathbf{F}_M^{\text{an}}, \varphi, \vartheta),$$

where  $\mathcal{C}_j \equiv \{C_{0j}, \dots, C_{(N-1)j}\}$  (with  $j \in \{F\} \cup \{I, M\}$ ). Moreover, by prescribing that the mass densities of both the solid sub-phases,  $\mathcal{F}_I$  and  $\mathcal{F}_M$ , and the fluid-phase,  $\mathcal{F}_F$ , are given by constitutive (or state) functions of the mass fractions of constituents, *i.e.*

$$(4.11) \quad \rho_F := \widehat{\rho}_F(\mathcal{C}_F), \quad \rho_k := \widehat{\rho}_k(\mathcal{C}_k), \quad k \in \{I, M\},$$

we write the constitutive expressions of  $\widehat{A}_F$  and  $\widehat{A}_S$  as

$$(4.12) \quad \widehat{A}_F(\rho_F, \mathcal{C}_F, \Theta) = \widehat{\Psi}_F(\mathcal{C}_F, \Theta),$$

$$(4.13) \quad \widehat{A}_S(\rho_I, \rho_M, \mathcal{C}_I, \mathcal{C}_M, \Theta, \mathbf{F}, \mathbf{F}_I^{\text{an}}, \mathbf{F}_M^{\text{an}}, \varphi, \vartheta) = \widehat{\Psi}_S(\mathcal{C}_I, \mathcal{C}_M, \Theta, \mathbf{F}, \mathbf{F}_I^{\text{an}}, \mathbf{F}_M^{\text{an}}, \varphi, \vartheta).$$

## 5. – Exploitation of the entropy production inequality

Substitution of eqs. (4.12) and (4.13) into (3.4), the result into (3.5), and application of Coleman-Noll procedure to the resulting expression of entropy production leads to the constitutive laws reported below.

**5.1. Lagrange Multipliers.** – Lagrange multipliers are found to be

$$(5.1) \quad \lambda_{\beta I} = \frac{\partial \widehat{\Psi}_I}{\partial C_{\beta I}} - \frac{\pi}{\widehat{\rho}_I^2} \frac{\partial \widehat{\rho}_I}{\partial C_{\beta I}}, \quad \lambda_{\beta M} = \frac{\partial \widehat{\Psi}_M}{\partial C_{\beta M}} - \frac{\pi}{\widehat{\rho}_M^2} \frac{\partial \widehat{\rho}_M}{\partial C_{\beta M}}, \quad \lambda_{\beta F} = \frac{\partial \widehat{\Psi}_F}{\partial C_{\beta F}} - \frac{\pi}{\widehat{\rho}_F^2} \frac{\partial \widehat{\rho}_F}{\partial C_{\beta F}}.$$

Quantities  $\lambda_{\beta j}$  (with  $\beta \in \{0, \dots, N-1\}$  and  $j \in \{F\} \cup \{I, M\}$ ) are identified with the *relative chemical potential* of constituent  $\mathcal{C}_{\beta j}$ , *i.e.*  $\lambda_{\beta j} \equiv m_{\beta j} := \mu_{\beta j} - \mu_{Nj}$ , where  $\mu_{\beta j}$  and  $\mu_{Nj}$  are the *absolute chemical potentials* of constituents  $\mathcal{C}_{\beta j}$  and  $\mathcal{C}_{Nj}$  in phase  $\mathcal{F}_j$ , respectively (cf., for example, [23]). We remark that, by definition,  $\lambda_{Nj} \equiv m_{Nj} \equiv 0$  for all  $\mathcal{F}_j$ ,  $j \in \{F, I, M\}$ . Finally, Lagrange multiplier  $\Lambda_F$  is found to be [23, 35]

$$(5.2) \quad \Lambda_F = A_{NF} \mathbf{I} - \frac{1}{\phi_F \rho_F C_{NF}} \boldsymbol{\sigma}_{NF},$$

where  $A_{NF}$ ,  $C_{NF}$ , and  $\boldsymbol{\sigma}_{NF}$  are Helmholtz free-energy density, mass fraction, and Cauchy stress tensor of constituent  $\mathcal{C}_{NF}$  in the fluid phase.

**5.2. Entropies and Cauchy stress tensors.** – Entropy densities of the solid and fluid phase are related to the derivatives of the corresponding Helmholtz free-energy density with respect to temperature, *i.e.*

$$(5.3) \quad S_S = -\frac{\partial \widehat{\Psi}_S}{\partial \Theta}, \quad \text{and} \quad S_F = -\frac{\partial \widehat{\Psi}_F}{\partial \Theta}.$$

Furthermore, the Cauchy stress tensors of fluid-phase constituents, fluid phase as a whole, and solid phase are given by

$$(5.4) \quad \boldsymbol{\sigma}_{\alpha F} = \phi_F \rho_F C_{\alpha F} (A_{\alpha F} - \lambda_{\alpha F}) \mathbf{I} - \phi_F \rho_F C_{\alpha F} \boldsymbol{\Lambda}_F,$$

$$(5.5) \quad \boldsymbol{\sigma}_F = -\phi_F \rho_F \lambda_F \mathbf{I} - \sum_{\alpha=0}^N \phi_F \rho_F C_{\alpha F} \mathbf{u}_F \otimes \mathbf{u}_F = -\phi_F \pi \mathbf{I} - \sum_{\alpha=0}^N \phi_F \rho_F C_{\alpha F} \mathbf{u}_F \otimes \mathbf{u}_F,$$

$$(5.6) \quad \boldsymbol{\sigma}_S = -\phi_S \pi \mathbf{I} + \boldsymbol{\sigma}_I^e + \boldsymbol{\sigma}_M^e.$$

We remark that, in order for eq. (5.4) to be consistent with eq. (5.5), the sum over all constituents in the fluid phase,  $C_{\alpha F}$  ( $\alpha \in \{0, \dots, N\}$ ), of the Cauchy stress tensors  $\boldsymbol{\sigma}_{\alpha F}$  must be equal to the inner part of the fluid-phase Cauchy stress tensor,  $\boldsymbol{\sigma}_F$ , *i.e.*

$$(5.7) \quad \sum_{\alpha=0}^N \boldsymbol{\sigma}_{\alpha F} = -\phi_F \pi.$$

By using eq. (5.2), this condition can be used in order to show that

$$(5.8) \quad \mu_{\alpha F} = A_{\alpha F} + \frac{\pi_{\alpha F}}{\rho_F C_{\alpha F}},$$

where  $\pi_{\alpha F}$  is the partial pressure of the fluid-phase constituent  $C_{\alpha F}$  ( $\alpha \in \{0, \dots, N\}$ ). Moreover, the Cauchy stress tensor of constituent  $C_{\alpha F}$  is given by  $\boldsymbol{\sigma}_{\alpha F} = -\phi_F \pi_{\alpha F} \mathbf{I}$ , and partial pressures  $\pi_{\alpha F}$  satisfy

$$(5.9) \quad \sum_{\alpha=0}^N \pi_{\alpha F} = \pi.$$

Finally, the sum of the quantities  $\boldsymbol{\sigma}_I^e$  and  $\boldsymbol{\sigma}_M^e$  represents the elastic part of solid-phase overall Cauchy stress tensor. These two tensors are defined by

$$(5.10) \quad \begin{aligned} \boldsymbol{\sigma}_I^e &:= \frac{1}{J_S} \mathbf{F}_I^e \frac{\partial \overline{W}_I}{\partial \mathbf{E}_I^e} (\mathbf{F}_I^e)^T + \phi_I \rho_I (A_S - A_I) \mathbf{I} \\ &= \phi_I \rho_I \mathbf{F}_I^e \frac{\partial A_I}{\partial \mathbf{E}_I^e} (\mathbf{F}_I^e)^T + \phi_I \rho_I (A_S - A_I) \mathbf{I}, \end{aligned}$$

$$(5.11) \quad \begin{aligned} \boldsymbol{\sigma}_M^e &:= \frac{1}{J_S} \mathbf{F}_M^e \frac{\partial \overline{W}_M}{\partial \mathbf{E}_M^e} (\mathbf{F}_M^e)^T + \phi_M \rho_M (A_S - A_M) \mathbf{I} \\ &= \phi_M \rho_M \mathbf{F}_M^e \frac{\partial A_M}{\partial \mathbf{E}_M^e} (\mathbf{F}_M^e)^T + \phi_M \rho_M (A_S - A_M) \mathbf{I}, \end{aligned}$$

where  $\overline{W}_I := J_S W_I$ ,  $\overline{W}_M := J_S W_M$ , and  $W_I := \phi_I \rho_I A_I$  (with  $j \in \{I, M\}$ ), and their sum is such that the elastic part of the Cauchy stress tensor of the solid phase as a whole is given by

$$(5.12) \quad \boldsymbol{\sigma}_S^e = \frac{1}{J_S} \mathbf{F}_I^e \frac{\partial \overline{W}_I}{\partial \mathbf{E}_I^e} (\mathbf{F}_I^e)^T + \frac{1}{J_S} \mathbf{F}_M^e \frac{\partial \overline{W}_M}{\partial \mathbf{E}_M^e} (\mathbf{F}_M^e)^T.$$

Equations (5.10)-(5.11) are obtained by computing the total differential of  $\Psi_S$  and using the fact that, because of the multiplicative decomposition of  $\mathbf{F}_S$ , the hypothesis of *density-preserving growth* (which implies that  $\text{Tr}(\mathbf{L}_k^{\text{an}}) = R_k$ ), and the constraint  $\rho_k \equiv \widehat{\rho}_k(\mathcal{C}_k)$ , each volume fraction  $\phi_k$  can be written as a function of the elastic part of deformation,  $\mathbf{E}_k^e$ , and mass fraction of constituents in subphase  $\mathcal{F}_k$ , *i.e.*  $\phi_k \equiv \widehat{\phi}_k(\mathbf{E}_k^e, \mathcal{C}_k)$ , where

$$(5.13) \quad \frac{\partial \phi_k}{\partial \mathbf{E}_k^e} = -\phi_k \mathbf{C}_k^e -T, \quad \frac{\partial \phi_k}{\partial \mathcal{C}_{\beta k}} = -\frac{\phi_k}{\rho_k} \frac{\partial \widehat{\rho}_k}{\partial \mathcal{C}_{\beta k}},$$

and  $\mathbf{C}_k^e := \mathbf{F}_k^{eT} \mathbf{F}_k^e$ . Moreover, as done in [18], the Helmholtz free-energy density of sub-phase  $\mathcal{F}_k$ ,  $A_k$  ( $k \in \{I, M\}$ ), is assumed to depend on the overall deformation only through the elastic part of deformation associated with  $\mathcal{F}_k$ , *i.e.*  $\mathbf{E}_k^e$ .

**5.3. Residual entropy production inequality.** – By virtue of eqs. (5.1)–(5.12), under the assumptions of small diffusive velocities of fluid-phase constituents, small fluid-phase filtration velocity (*i.e.*  $\|\mathbf{u}_{\alpha F}\|^2 \ll 1$ , and  $\|\mathbf{w}_{FS}\|^2 \ll 1$ ), negligible inertial terms in momentum balance laws, and accounting for the kinematic constraints on the growth velocity gradients  $\mathbf{L}_I^{\text{an}}$  and  $\mathbf{L}_M^{\text{an}}$  [20], *i.e.*

$$(5.14) \quad \text{Tr}(\mathbf{L}_I^{\text{an}}) = \mathbf{L}_I^{\text{an}} : \mathbf{I}_I^N = R_I, \quad \text{and} \quad \text{Tr}(\mathbf{L}_M^{\text{an}}) = \mathbf{L}_M^{\text{an}} : \mathbf{I}_M^N = R_M,$$

the expression of the *residual* of entropy production can be written as

$$(5.15) \quad \begin{aligned} \Theta \rho \widetilde{\Gamma}_{\text{red}} = & \sum_{\alpha=0}^N \mathbf{u}_{\alpha F} \cdot \{-\phi_F \rho_F C_{\alpha F} (\nabla \mu_{\alpha F} - \mathbf{g})\} + \mathbf{w}_{FS} \cdot \{-\phi_F \rho_F \mathbf{T}_F + \pi \nabla \phi_F\} \\ & - \sum_{\beta=0}^{N-1} \phi_I \rho_I C_{\beta I} R_{\beta I} \{\lambda_{\beta I} - \lambda_{\beta F}\} - \sum_{\beta=0}^{N-1} \phi_M \rho_M C_{\beta M} R_{\beta M} \{\lambda_{\beta M} - \lambda_{\beta F}\} \\ & + \phi_I \rho_I \mathbf{L}_I^{\text{an}} : \left\{ \frac{1}{J_S \phi_I \rho_I} \mathbf{B}_I^e - \left( A_I + \frac{\pi}{\rho_I} - \sum_{\beta=0}^{N-1} C_{\beta I} \lambda_{\beta I} \right) \mathbf{I}_I^N \right. \\ & \left. + \left( A_F + \frac{\pi}{\rho_F} - \sum_{\beta=0}^{N-1} C_{\beta F} \lambda_{\beta F} \right) \mathbf{I}_I^N \right\} \\ & + \phi_M \rho_M \mathbf{L}_M^{\text{an}} : \left\{ \frac{1}{J_S \phi_M \rho_M} \mathbf{B}_M^e - \left( A_M + \frac{\pi}{\rho_M} - \sum_{\beta=0}^{N-1} C_{\beta M} \lambda_{\beta M} \right) \mathbf{I}_M^N \right. \\ & \left. + \left( A_F + \frac{\pi}{\rho_F} - \sum_{\beta=0}^{N-1} C_{\beta F} \lambda_{\beta F} \right) \mathbf{I}_M^N \right\} \\ & + \phi_S \rho_S A_S^* - \phi_S \rho_S \frac{D_S \varphi}{Dt} : \frac{\partial \widehat{\Psi}_S}{\partial \varphi} - \phi_S \rho_S \frac{D_S \vartheta}{Dt} : \frac{\partial \widehat{\Psi}_S}{\partial \vartheta} \geq 0, \end{aligned}$$



where we introduced the notation

$$(5.16) \quad \mathbf{B}_I^e := \mathbf{C}_I^e \frac{\partial \bar{W}_I}{\partial \mathbf{E}_I^e}, \quad \text{and} \quad \mathbf{B}_M^e := \mathbf{C}_M^e \frac{\partial \bar{W}_M}{\partial \mathbf{E}_M^e},$$

with  $\mathbf{C}_I^e = (\mathbf{F}_I^e)^T \mathbf{F}_I^e$ ,  $\mathbf{C}_M^e = (\mathbf{F}_M^e)^T \mathbf{F}_M^e$ , and  $J_S = \det(\mathbf{F}_S)$ .

Further conditions on the unknowns can be obtained by studying inequality (5.15) in the case of thermodynamic equilibrium, *i.e.* at the state at which all non-equilibrium variables

$$(5.17) \quad \text{Non-equilibrium variables} = \left\{ \frac{D_S \varphi}{Dt}, \frac{D_S \vartheta}{Dt}, \mathbf{u}_{\alpha F}, \mathbf{w}_{FS}, \mathbf{L}_I^{\text{an}}, \mathbf{L}_M^{\text{an}}, R_{\beta I}, R_{\beta M} \right\}$$

( $\beta \in \{0, \dots, N-1\}$ ) vanish identically. At equilibrium, entropy production is zero, *i.e.* it attains its minimum value. Therefore, it is possible to characterise equilibrium by requiring the “gradient”, and the “Hessian” of the residual entropy production with respect to non-equilibrium variables to be zero, and be positive-definite, respectively. For this purpose, inspired by [36], we assume that the quantity  $\phi_S \rho_S A_S^*$  can be written as

$$(5.18) \quad \phi_S \rho_S A_S^* = \phi_S \rho_S A_S^{**} + \phi_I \rho_I \mathbf{L}_I^{\text{an}} : \mathbf{H}_I + \phi_M \rho_M \mathbf{L}_M^{\text{an}} : \mathbf{H}_M,$$

where  $\mathbf{H}_I$  and  $\mathbf{H}_M$  are two second-order tensors related growth. Under this assumption, the equilibrium conditions are fulfilled by the following set of Onsager’s relations:

$$(5.19) \quad \mathbf{u}_{\beta F} = -\mathbf{M}_{\beta F} \nabla m_{\beta F},$$

$$(5.20) \quad \mathbf{w}_{FS} = -\mathbf{M}_F (\phi_F \rho_F \mathbf{T}_F - \pi \nabla \phi_F),$$

$$(5.21) \quad R_{\beta I} = -\mathcal{Z}_{\beta I} (m_{\beta I} - m_{\beta F}),$$

$$(5.22) \quad R_{\beta M} = -\mathcal{Z}_{\beta M} (m_{\beta M} - m_{\beta F}),$$

$$(5.23) \quad \mathbf{L}_I^{\text{an}} = \mathbb{M}_I : \left\{ \frac{1}{J_S \phi_I \rho_I} \mathbf{B}_I^e + \mathbf{H}_I - \left( A_I + \frac{\pi}{\rho_I} - \sum_{\beta=0}^{N-1} C_{\beta I} m_{\beta I} \right) \mathbf{I}_I^N \right. \\ \left. + \left( A_F + \frac{\pi}{\rho_F} - \sum_{\beta=0}^{N-1} C_{\beta F} m_{\beta F} \right) \mathbf{I}_I^N \right\},$$

$$(5.24) \quad \mathbf{L}_M^{\text{an}} = \mathbb{M}_M : \left\{ \frac{1}{J_S \phi_M \rho_M} \mathbf{B}_M^e + \mathbf{H}_M - \left( A_M + \frac{\pi}{\rho_M} - \sum_{\beta=0}^{N-1} C_{\beta M} m_{\beta M} \right) \mathbf{I}_M^N \right. \\ \left. + \left( A_F + \frac{\pi}{\rho_F} - \sum_{\beta=0}^{N-1} C_{\beta F} m_{\beta F} \right) \mathbf{I}_M^N \right\},$$

$$(5.25) \quad A_S^{**} - \frac{D_S \varphi}{Dt} : \frac{\partial \hat{\Psi}_S}{\partial \varphi} - \frac{D_S \vartheta}{Dt} : \frac{\partial \hat{\Psi}_S}{\partial \vartheta} \geq 0,$$

where we used the definitions of relative chemical potentials,  $\lambda_{\beta I} = m_{\beta I}$ ,  $\lambda_{\beta M} = m_{\beta M}$ , and  $\lambda_{\beta F} = m_{\beta F}$  (with  $\beta \in \{0, \dots, N-1\}$ ), given in eqs. (5.1). If some constituents are

not exchanged, then the index  $\beta$  in eqs. (5.21)–(5.22) ranges in the subset of constituents that are actually exchanged, *i.e.* those for which  $R_{\beta j} \neq 0$ . Coefficients  $\mathcal{Z}_{\beta I}$ ,  $\mathcal{Z}_{\beta M}$  are positive scalars,  $\mathbf{M}_{\beta F}$ ,  $\mathbf{M}_F$  are positive-definite second-order tensors, and  $\mathbb{M}_I$ ,  $\mathbb{M}_M$  are positive-definite fourth-order tensors endowed with the major symmetry (cf. Loret and Simões [20]). In general, each of these coefficients may be a function of all independent constitutive variables. We remark that eqs. (5.19)–(5.24) are obtained by inverting the near-equilibrium expansions of the coefficients of non-equilibrium variables (5.17), *i.e.* the terms between braces in eq. (5.15).

Onsager's relation (5.19) provides diffusion-dispersion Fick's law for the fluid-phase constituent  $\mathcal{C}_{\beta F}$ . Indeed, by multiplying both sides of eq. (5.19) by  $\phi_F \rho_F C_{\beta F}$ , and defining the diffusive-dispersive mass flux constituent  $\mathcal{C}_{\beta F}$  as  $\mathbf{J}_{\beta F} = \phi_F \rho_F C_{\beta F} \mathbf{u}_{\beta F}$ , we obtain

$$(5.26) \quad \mathbf{J}_{\beta F} = -\phi_F \rho_F C_{\beta F} \mathbf{M}_{\beta F} \nabla m_{\beta F},$$

where tensor  $\mathbf{M}_{\beta F}$  is proportional to the diffusive-dispersive tensor associated with constituent  $\mathcal{C}_{\beta F}$ .

Equation (5.20) gives Darcy's law of flow for the filtration velocity of the fluid phase. This law is obtained by expressing the momentum exchange  $\phi_F \rho_F \mathbf{T}_F$  through the balance of momentum for the fluid phase, approximating the fluid-phase Cauchy stress tensor by  $\boldsymbol{\sigma}_F \approx -\phi_F \pi \mathbf{I}$  (cf. eq. (5.5)), and invoking the hypothesis of negligible inertial terms, *i.e.*

$$(5.27) \quad \phi_F \rho_F \mathbf{T}_F = \nabla(\phi_F \pi) - \phi_F \rho_F \mathbf{g}.$$

Substitution of eq. (5.27) into Onsager's relation (5.20) leads to

$$(5.28) \quad \mathbf{w}_{FS} = -\mathbf{K}_F (\nabla \pi - \rho_F \mathbf{g}),$$

where  $\mathbf{K}_F = \phi_F \mathbf{M}_F$  is said to be the fluid-phase permeability tensor.

Onsager's relations (5.21) and (5.22) imply that the source (or sink) of mass of constituent  $\mathcal{C}_{\beta k}$  (with  $k \in \{I, M\}$ ) depends on the difference between the relative chemical potential of that constituent,  $m_{\beta k}$ , and the relative chemical potential of constituent  $\mathcal{C}_{\beta F}$  in the fluid-phase,  $m_{\beta F}$ . Since, at equilibrium, the relative chemical potential of a given constituent has to be the same for any phase, the conditions  $m_{\beta I} \equiv m_{\beta F}$  and  $m_{\beta M} = m_{\beta F}$  imply that sources (or sinks)  $R_{\beta I}$  and  $R_{\beta M}$  have to vanish at equilibrium. Furthermore, since  $\phi_F \rho_F C_{\beta F} R_{\beta F} = -(\phi_I \rho_I C_{\beta I} R_{\beta I} + \phi_M \rho_M C_{\beta M} R_{\beta M})$ , also the source (or sink) of mass for constituent  $\mathcal{C}_{\beta F}$  in the fluid-phase has to be zero at equilibrium. Results (5.19)–(5.22) were obtained by Bennethum *et al.* in [23], and have been here slightly generalized to the case of a solid-phase consisting of two sub-phases (*i.e.* matrix and fibre-like inclusion sub-phases).

Equations (5.23) and (5.24) represent the growth laws for the solid sub-phases  $\mathcal{F}_I$  and  $\mathcal{F}_M$ , respectively. These relations can be rewritten by defining, for solid sub-phases  $\mathcal{F}_I$

and  $\mathcal{F}_M$ , and for the fluid-phase  $\mathcal{F}_F$ , the following Mandel-type stress tensors:

$$(5.29) \quad \mathbf{B}_I = \mathbf{B}_I^e + J_S \phi_I \rho_I \sum_{\beta=0}^{N-1} C_{\beta I} m_{\beta I} \mathbf{I}_I^N,$$

$$(5.30) \quad \mathbf{B}_M = \mathbf{B}_M^e + J_S \phi_M \rho_M \sum_{\beta=0}^{N-1} C_{\beta M} m_{\beta M} \mathbf{I}_M^N,$$

$$(5.31) \quad \mathbf{B}_{FI} = J_S \phi_I \rho_I \sum_{\beta=0}^{N-1} C_{\beta F} m_{\beta F} \mathbf{I}_I^N, \quad \text{and}$$

$$\mathbf{B}_{FM} = J_S \phi_M \rho_M \sum_{\beta=0}^{N-1} C_{\beta F} m_{\beta F} \mathbf{I}_M^N.$$

Substitution of definitions (5.29)–(5.31) into eqs. (5.23) and (5.24) yields

$$(5.32) \quad \mathbf{L}_I^{\text{an}} = \mathbb{M}_I : \left\{ \frac{1}{J_S \phi_I \rho_I} (\mathbf{B}_I - \mathbf{B}_{FI}) + \mathbf{H}_I - \left[ \left( A_I + \frac{\pi}{\rho_I} \right) - \left( A_F + \frac{\pi}{\rho_F} \right) \right] \mathbf{I}_I^N \right\},$$

$$(5.33) \quad \mathbf{L}_M^{\text{an}} = \mathbb{M}_M : \left\{ \frac{1}{J_S \phi_M \rho_M} (\mathbf{B}_M - \mathbf{B}_{FM}) + \mathbf{H}_M - \left[ \left( A_M + \frac{\pi}{\rho_M} \right) - \left( A_F + \frac{\pi}{\rho_F} \right) \right] \mathbf{I}_M^N \right\}.$$

Equations (5.32) and (5.33) are a generalisation of the results reported in [16]. Similar growth laws have been previously proposed by Lorete and Simões [20], Fusi *et al.* [37] for a fluid-solid mixture with mass exchange between constituents, Ambrosi and Guana [38] in the monophasic continuum, and by Ambrosi *et al.* [39] for a non-homogeneous monophasic body. Following [36, 40], tensors  $\mathbf{H}_I$  and  $\mathbf{H}_M$  may be taken as

$$(5.34) \quad \mathbf{H}_I = -\frac{1}{J_S \phi_I \rho_I} \mathbf{B}_I^0, \quad \text{and} \quad \mathbf{H}_M = -\frac{1}{J_S \phi_M \rho_M} \mathbf{B}_M^0,$$

where  $\mathbf{B}_I^0$  and  $\mathbf{B}_M^0$  are external “forces” that, at equilibrium, balance  $\mathbf{B}_I$  and  $\mathbf{B}_M$ , respectively.

By accounting for Onsager’s relations (5.19)–(5.22), and (5.32)–(5.33), we conclude that inequality (5.25) has to be verified in order for the entropy principle to be respected. In the next section, we investigate the consequences of this requirement.

## 6. – Evolution of fibre-like inclusions

Since the solid-phase Helmholtz free-energy density,  $\widehat{\Psi}_S$ , is assumed to be a dependent constitutive variable, the derivative of  $\widehat{\Psi}_S$  with respect to the texture tensor  $\widehat{\Xi}_I^N$  can be regarded as known. Therefore, it can be concluded that, in inequality (5.25), unknown quantities are  $D_S \widehat{\Xi}_I^N / Dt$  and  $A_S^{**}$ .

Following Chadwick [11], Imatani and Maugin [14], and Maugin and Imatani [15], a formulation of remodelling can be obtained on the basis of the evolution of the texture tensor  $\Xi_I^N$ , *i.e.*

$$(6.1) \quad \frac{D_S \Xi_I^N}{Dt} = \Xi_I^N (\mathbf{L}_I^{\text{an}})^T + \mathbf{L}_I^{\text{an}} \Xi_I^N - 2 (\mathbf{L}_I^{\text{an}} : \Xi_I^N) \Xi_I^N.$$

We remark that, by virtue of eq. (4.3), texture tensor  $\Xi_S$  is determined if tensors  $\Xi_I^N$  and  $\mathbf{F}_I^e$  are known. Therefore, if  $\Xi_I^N$  is the solution of the evolution law (6.1), texture tensor  $\Xi_S$  can be computed directly through eq. (4.3)

Imatani and Maugin [14], and Maugin and Imatani [15] make a distinction between the case in which vector  $\lambda_I^N$  is embedded in the continuum, and the case in which vector  $\lambda_I^N$  behaves as a float. Here, we consider only the first case, and we propose to use Onsager's relation (5.32) in order to express the growth velocity gradient,  $\mathbf{L}_I^{\text{an}}$ , for the fibre-like inclusion sub-phase  $\mathcal{F}_I$ . By doing that, tensor  $\mathbf{L}_I^{\text{an}}$  is a function of Mandel-type stress tensors  $\mathbf{B}_I$  and  $\mathbf{B}_{FI}$ , as well as other thermodynamic quantities. This confirms that Mandel stress is the driving force for remodelling.

As an example, following Olsson and Klarbring [40], we assume that the growth law (5.32) takes on the simplified form

$$(6.2) \quad \mathbf{L}_I^{\text{an}} = M_I \left\{ \frac{1}{J_S \phi_I \rho_I} (\mathbf{B}_I - \mathbf{B}_{FI} - \mathbf{B}_I^0) - \left[ \left( A_I + \frac{\pi}{\rho_I} \right) - \left( A_F + \frac{\pi}{\rho_F} \right) \right] \mathbf{I}_I^N \right\},$$

where  $M_I$  is a positive scalar coefficient. We notice that the growth velocity gradient  $\mathbf{L}_I^{\text{an}}$  contains hydrostatic contributions. Since hydrostatic terms cannot play any role in the evolution equation of  $\Xi_I^N$ , by introducing the notation

$$(6.3) \quad M_I^* = \frac{M_I}{J_S \phi_I \rho_I},$$

eq. (6.1) becomes

$$(6.4) \quad \frac{D_S \Xi_I^N}{Dt} = \Xi_I^N M_I^* (\mathbf{B}_I^e - \mathbf{B}_I^0)^T + M_I^* (\mathbf{B}_I^e - \mathbf{B}_I^0) \Xi_I^N - 2 [M_I^* (\mathbf{B}_I^e - \mathbf{B}_I^0) : \Xi_I^N] \Xi_I^N,$$

where only the *elastic* part of the Mandel-type stress tensor  $\mathbf{B}_I$  is the driving force for remodelling. We remark that, in the example described above, the fluid-phase Mandel-type stress tensor plays no role in remodelling due to the fact that it is a hydrostatic tensor.

This result shows that, in the simple case studied in this section, only the elastic Mandel-type stress tensor,  $\mathbf{B}_I^e$ , acts as a driving force for the reorientation of fibre-like inclusions.

There are alternative formulations of remodelling that, rather than starting from a prescribed evolution law for the texture tensor,  $\Xi_I^N$ , determine the evolution of given “*remodelling parameters*” through the prescription of “*generalized remodelling forces*”, and the exploitation of the entropy inequality. For example, in the context of remodelling of arterial walls, this approach has been recently tackled by Olsson and Klarbring [40]. In [40], the authors identify the set of remodelling parameters with the Euler angle

determining the orientation of the fibres in an arterial wall. In this case, our eq. (5.25) reads

$$(6.5) \quad A_S^{**} - \frac{D_S \varphi}{Dt} \frac{\partial \widehat{\Psi}_S}{\partial \varphi} - \frac{D_S \vartheta}{Dt} \frac{\partial \widehat{\Psi}_S}{\partial \vartheta} \geq 0.$$

The difference between our approach and the method proposed by Olsson and Klarbring [40] is that, while we prescribe the evolution of the angles  $\vartheta$  and  $\varphi$ , and consequently determine a condition for  $A_S^{**}$  through eq. (5.25), they find Onsager's relations for the derivatives  $D_S \varphi / Dt$  and  $D_S \vartheta / Dt$  based on the assumption that  $A_S^{**}$  has the form

$$(6.6) \quad A_S^{**} = \mathcal{R}_\varphi \frac{D_S \varphi}{Dt} + \mathcal{R}_\vartheta \frac{D_S \vartheta}{Dt},$$

where  $\mathcal{R}_\varphi$  and  $\mathcal{R}_\vartheta$  are prescribed “generalized forces” associated with remodelling<sup>(8)</sup>. In fact, by rewriting inequality (6.5) as

$$(6.7) \quad \left[ \mathcal{R}_\varphi - \frac{\partial \widehat{\Psi}_S}{\partial \varphi} \right] \frac{D_S \varphi}{Dt} + \left[ \mathcal{R}_\vartheta - \frac{\partial \widehat{\Psi}_S}{\partial \vartheta} \right] \frac{D_S \vartheta}{Dt} \geq 0,$$

the evolution law for the angles  $\varphi$  and  $\vartheta$  can be expressed in terms of Onsager's relation

$$(6.8) \quad \frac{D_S \varphi}{Dt} = \mathcal{K}_\varphi \left( \mathcal{R}_\varphi - \frac{\partial \widehat{\Psi}_S}{\partial \varphi} \right), \quad \text{and} \quad \frac{D_S \vartheta}{Dt} = \mathcal{K}_\vartheta \left( \mathcal{R}_\vartheta - \frac{\partial \widehat{\Psi}_S}{\partial \vartheta} \right)$$

where  $\mathcal{K}_\varphi, \mathcal{K}_\vartheta \geq 0$  are material parameters.

If we assume for simplicity that both the growth inhomogeneity gradient,  $\mathbf{L}_I^{\text{an}}$ , and the elastic Green-Lagrange strain tensors,  $\mathbf{E}_I^e$  and  $\mathbf{E}_I^{\text{an}}$ , are diagonal, eq. (6.4), written in terms of the angles  $\vartheta$  and  $\varphi$ , becomes

$$(6.9) \quad \frac{D_S \vartheta}{Dt} = -\frac{M_I^*}{2} \sin(2\vartheta) \{ [(\mathbf{B}_I^e)_{11} - (\mathbf{B}_I^0)_{11}] - [(\mathbf{B}_I^e)_{22} - (\mathbf{B}_I^0)_{22}] \},$$

$$(6.10) \quad \frac{D_S \varphi}{Dt} = \frac{M_I^*}{2} \sin(2\varphi) \{ [(\mathbf{B}_I^e)_{11} - (\mathbf{B}_I^0)_{11}] \cos^2(\vartheta) \\ + [(\mathbf{B}_I^e)_{22} - (\mathbf{B}_I^0)_{22}] \sin^2(\vartheta) - [(\mathbf{B}_I^e)_{33} - (\mathbf{B}_I^0)_{33}] \}.$$

Therefore, by comparing this result with the procedure proposed by Olsson and Klarbring [40], we find that the remodelling forces,  $\mathcal{R}_\vartheta$  and  $\mathcal{R}_\varphi$ , are given by

$$(6.11) \quad \mathcal{R}_\vartheta = -\frac{M_I^*}{2\mathcal{K}_\vartheta} \sin(2\vartheta) \{ [(\mathbf{B}_I^e)_{11} - (\mathbf{B}_I^0)_{11}] - [(\mathbf{B}_I^e)_{22} - (\mathbf{B}_I^0)_{22}] \} + \frac{\partial \widehat{\Psi}_S}{\partial \vartheta},$$

$$(6.12) \quad \mathcal{R}_\varphi = \frac{M_I^*}{2\mathcal{K}_\varphi} \sin(2\varphi) \{ [(\mathbf{B}_I^e)_{11} - (\mathbf{B}_I^0)_{11}] \cos^2(\vartheta) + [(\mathbf{B}_I^e)_{22} - (\mathbf{B}_I^0)_{22}] \sin^2(\vartheta) \\ - [(\mathbf{B}_I^e)_{33} - (\mathbf{B}_I^0)_{33}] \} + \frac{\partial \widehat{\Psi}_S}{\partial \varphi}.$$

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<sup>(8)</sup> In the case of growth, this idea was put forward by Di Carlo and Quiligotti [36].

## 7. – Conclusion

We studied a biological mixture consisting of a fluid and a solid phase. The latter was assumed to comprise two sub-phases, *i.e.* matrix and fibre-like inclusion, moving with the common phase velocity,  $\mathbf{v}_S$ . Each phase was modelled as a mixture on its own with several constituents. Because of mass exchanges between phases, the solid-phase experiences growth (or resorption). Growth, and the material inhomogeneities related to it, were described through the multiplicative decomposition of the solid-phase deformation gradient tensor, and the introduction of the growth velocity gradient. On the basis of previous works [37, 39, 16], the growth velocity gradients of sub-phases  $\mathcal{F}_I$  and  $\mathcal{F}_M$  are given through Onsager’s relation (cf. eqs. (5.32) and (5.33)), in which the Mandel-type stress tensors  $\mathbf{B}_I$ ,  $\mathbf{B}_M$ ,  $\mathbf{B}_{FI}$ , and  $\mathbf{B}_{FM}$  are introduced. Tensors  $\mathbf{B}_I$  and  $\mathbf{B}_M$  contain both an elastic part, and a contribution due to the chemical potential of the constituents exchanged with the fluid-phase. The fluid-phase Mandel-type stress tensor,  $\mathbf{B}_F$ , is defined only in terms of chemical potential of fluid-phase constituents, and, within the theoretical framework presented in this paper, is hydrostatic.

Onsager’s relations (5.32) and (5.33) are also used in order to highlight the role of Mandel-type stress tensors on remodelling. We suggested a possible way of including Mandel-type stress tensors, and other constitutive variables, in the evolution laws of the texture tensor,  $\Xi_I^N$ . Although we obtained results similar to those pointed out by Imatani and Maugin [14], and Maugin and Imatani [15], our procedure is methodological different because it is grounded on the derivation of Onsager’s relations in order to express the growth velocity gradient, (5.32), of the fibre-like inclusion sub-phase,  $\mathcal{F}_I$ .

If the mixture under study is made of matrix and fibre-like inclusions (*i.e.* no fluid-phase), Onsager’s relations for growth velocity gradients  $\mathbf{L}_I^{\text{an}}$  and  $\mathbf{L}_M^{\text{an}}$  could still be found, although they should be accordingly modified.

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