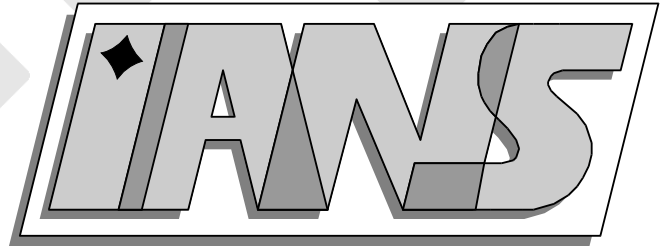


**Universität
Stuttgart**



Phase separation in solid mixtures under elastic
loadings with application to solder materials

Thomas Merkle

**Berichte aus dem Institut für
Angewandte Analysis und Numerische Simulation**

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PHASE SEPARATION IN SOLID MIXTURES UNDER ELASTIC LOADINGS WITH APPLICATION TO SOLDER MATERIALS

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1. INTRODUCTION

The separation of a solid mixture, for example an alloy, into two or more phases becomes recently of significant interest in joining technique. In particular, the reliability and life time of a solder joint used in Surface Mount Technology (SMT) becomes more and more important to electronic industry. In SMT solder is used to connect silicon wafers and Printed Circuit Boards (PCB) electronically and mechanically. The thermal mismatch between various materials involved in the microelectronic composite in combination with advanced temperatures and mechanical loadings is the key issue in solder joint reliability.

Figure 1 shows a cross section cut through a tin/silver solder bump, which has been subjected to several thousand power cycles. The regions of gray and white shades demonstrates, that the original fine mixture of tin and silver separates into islands of high silver and high tin concentration.

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This process is called coarsening and results from diffusion in combination with mechanical loadings under high temperatures ($> 80^{\circ}\text{C}$).

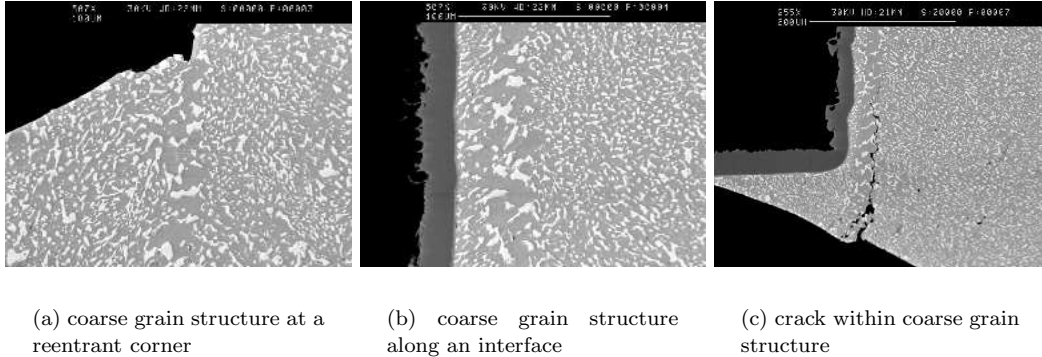


FIGURE 1. Robert Bosch all rights reserved

A phenomenological theory of phase separation in alloys is developed in [3, 4, 5], where also a mathematical model is derived. The driving force for the coarsening phenomena results from the free energy of the interfaces between tin and silver regions. Different continua diffuse within the mixture to reduce the amount of interface and thus to reduce the interface energy, but making the mixture coarser.

For many solder, where the kind of atoms assembling the alloy are not too different, the atoms create a common lattice. They can achieve this state only at the costs of essential distortion of the lattice. In the beginning the energy arising from distortion is relatively unimportant. It becomes important due to the fact, that coarsening decrease the interfacial energy and the mechanical interactions predominates. In a late stage the mechanical interactions affect the coarsening process and the structure of the mixture in a dominant way. The influence of mechanical loadings on the separation process in solder joints was studied in [7, 22, 24, 27, 28], for general alloys this considerations are done in [12].

Figure 1(a) shows a coarse material structure beginning at an reentrant corner, where it is well known, that there are locally high mechanical stresses. In figure 1(b) a coarse grain structure develops along an interface between the solder and the surrounding material. This is also a part of the microelectronic composite, where high local mechanical stresses appear. Finally, figure 1(c) shows a crack resulting from high mechanical stresses within a coarse grain region. These figures demonstrate the influence of mechanical stresses on the phase separation process.

This paper is organised as follows:

In section 2 the kinematics of the mixture is developed. The motion as well as all other kinematic quantities of various continua are described as for a single continuum, see [17]. Mixture's kinematics are formulated by a mass averaged technique, which is quite standard in mixture theory [1, 2, 31]. We show, that the mass averaged motion of the mixture satisfies the definition of a deformation.

In section 3 we introduce Reynold's transport theorem, which plays an important role in the following section for deriving the conservation equations. Further, we formulate in this section a version of Piola's transformation for vector valued functions as well as for tensor valued functions.

Section 4 deals with the balance of mass for each single continuum as well as for the whole mixture. In this section we give a mathematical rigorous formulation of the diffusion process based on the ideas outlined in [1, 2, 31]. The diffusion equation holds in the reference configuration, whereas the transformation into the reference configuration is done by Piola's transformation.

The balance of linear momentum for each single continuum and for the whole mixture in a mass averaged sense is considered in section 5. Here the attention is turned onto the relation between the stress tensor of the mixture, the stress tensors of each continua and the influence of diffusion.

Similar relations are formulated in [1, 2, 31] within the actual configuration and in this paper these relations are transformed into the reference configuration by using Piola's transformation.

In section 6 the phase separation process is modelled under consideration of microforces, also known as configurational forces, see [18, 20]. Microforces are related to the integrity of materials structure, hence they provide a generalisation of Eshelby's theory of lattice defects [11]. Although there exists a theory of Eshelby's relation for mixtures [30], in this paper microforces are used to model the evolution of concentration surfaces based on the ideas in [13, 14, 19]. As we will see in section 8 and similar to [13, 14] microforces appear as variational derivatives of the Helmholtz free energy.

Section 7 deals with the energy conservation of each single continuum as well as energy conservation of the whole mixture. Microforces perform work in the evolution of lattice defects, in our considerations they achieve work in the evolution of concentration surfaces. Thus the energy balance of mixture derived in [31] must be augmented by the energy performed by the microforces similar to [13, 14, 20], to get a generalised energy formulation.

In section 8 the entropy inequality of the mixture is examined by using the ideas developed in [31]. The entropy inequality will be formulated in the reference configuration by applying Piola's transformation. In order to derive a dissipation inequality the energy balance of section 7 is used. Hence we get a dissipation inequality, which contains energetic effects resulting from microforces.

In section 9 constitutive assumptions are formulated with respect to the application on solder material under elastic loadings. These constitutive assumptions in combination with the dissipation inequality of section 7 yield constitutive relations in order to derive a determined material model. By using an elastic strain energy, a double well exchange energy and a quadratic surface energy we derive a generalised Cahn-Hilliard equation similar to [9, 12, 19, 22].

2. KINEMATICS

Kinematics describes the change of geometry undergone by continuous bodies during an evolutionary process. In the context of solid mixtures a formulation of the kinematics of different continua as well as of the whole mixture is necessary. The kinematic quantities of each continuum will be formulated similar as in the case of a single continuum see [17, 31]. In contrast to a single material the kinematic quantities of the mixture will be defined by an appropriate averaging technique, which takes the distribution of mass of different continua into account. This is an almost standard technique and used in [1, 2, 31].

A body occupies different regions of the Euclidean space \mathbb{R}^d in different times, thus we have to define a domain, which can be identified with the body in a reference state..

Definition 2.1. Let $\Omega \subset \mathbb{R}^d$ an open, bounded and simply connected set with piecewise smooth boundary, then Ω is called a domain.

In this paper $d \in \mathbb{N}$ denotes the space dimension. For practical considerations we only observe the cases $d = 2; 3$.

The mixture consists of a finite number of $1 \leq k \leq K$ continua. Within this paper a solid mixture is modelled as a superposition of several continua, where each material point is occupied by different continua at any time. In continuum mechanics this technique is known as principle of overlapping continua, see [31].

Although the mixture as well as several continua occupy different region at different times, one of this region can intrinsically associated with the solid body and used as reference configuration.

Definition 2.2. A continuum k occupies at time $t = 0$ ($t = 0$ without loss of generality) the domain $\Omega_k \subset \mathbb{R}^d$. Ω_k is called reference configuration of continuum k , in addition the reference configuration of the mixture is given by

$$\Omega := \bigcup_{k=1}^K \Omega_k.$$

In this context coordinate $\boldsymbol{x} \in \Omega$ is called reference or Lagrange coordinate. In general we can not follow from the superposition of the different continua that

$$\Omega_k \cap \Omega_l = \emptyset \quad \text{for } k \neq l.$$

In this paper we only observe a positive time parameterisation and therefore we define the set of non-negative real numbers

$$\mathbb{R}^+ := \{t \in \mathbb{R} : t \geq 0\}.$$

Most important in continuum mechanics is the study of motions and deformations of a body. Mathematically this can be expressed by a mapping, which carries material points into a new configuration.

Definition 2.3. The motion of a continuum k is given by a vector valued mapping

$$\boldsymbol{\varphi}_k : \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d,$$

which satisfies properties as follows:

- (1) $\boldsymbol{\varphi}_k$ is a smooth vector field.
- (2) For all $t \in \mathbb{R}^+$ $\boldsymbol{\varphi}_k(t, \cdot)$ is injective.
- (3) For all $t \in \mathbb{R}^+$ $\boldsymbol{\varphi}_k(t, \cdot)$ is orientation preserving.
- (4) $\boldsymbol{\varphi}_k(0, \boldsymbol{x}) = \boldsymbol{x}$ represents the initial configuration.

In this context we remark, if $t \in \mathbb{R}^+$ is arbitrary, but fixed, then $\boldsymbol{\varphi}_k(t, \cdot)$ is called deformation. Accordingly, a motion consists of a one parametric bundle of deformations. In addition in this paper only classical differentiable functions are used, but for a mathematical analysis it is possible to extend this considerations to distributional derivatives.

Definition 2.4. The displacement of continuum k is defined by the smooth vector field

$$\begin{aligned} \boldsymbol{u}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \boldsymbol{u}_k(t, \boldsymbol{x}) &:= \boldsymbol{\varphi}_k(t, \boldsymbol{x}) - \boldsymbol{x}. \end{aligned}$$

For modelling mixtures we differ, as usual in continuum mechanics, between reference and actual configuration.

Definition 2.5. The actual configuration of continuum k at time $t \in \mathbb{R}^+$ is a subset of the Euclidean space \mathbb{R}^d defined by

$$\Omega_k^\varphi(t) := \{\boldsymbol{y} \in \mathbb{R}^d : \exists \boldsymbol{x} \in \Omega : \boldsymbol{y} = \boldsymbol{\varphi}_k(t, \boldsymbol{x})\}.$$

In addition the actual configuration of the mixture under consideration of the principle of overlapping continua is given by

$$\Omega^\varphi(t) := \bigcup_{k=1}^K \Omega_k^\varphi(t).$$

In agreement with this definition the actual space coordinate or Euler coordinate is denoted by $\boldsymbol{y} \in \Omega^\varphi(t)$. During motion continuum k occupies a region in time and space, which is called trajectory, see [17].

Definition 2.6. The trajectory of continuum k is defined by

$$\mathcal{T}_k := \{(t, \boldsymbol{y}) : t \in \mathbb{R}^+, \boldsymbol{y} \in \Omega_k^\varphi(t)\},$$

for the mixture the trajectory is declared by

$$\mathcal{T} := \bigcup_{k=1}^K \mathcal{T}_k.$$

Figure 2 illustrates the times-space-cylinder and the trajectory of the mixture as well as the mapping properties of the motion.

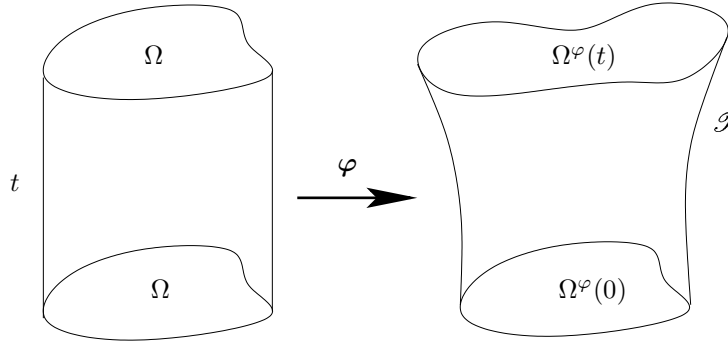


FIGURE 2. time-space-cylinder and trajectory

Definition 2.7. The velocity of continuum k in the reference configuration is defined by the smooth vector field

$$\begin{aligned} \mathbf{v}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \mathbf{v}_k(t, \mathbf{x}) &:= \frac{\partial}{\partial t} \boldsymbol{\varphi}_k(t, \mathbf{x}). \end{aligned}$$

The appropriate velocity field of continuum k in the actual configuration is given by the smooth vector field

$$\begin{aligned} \mathbf{v}_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^d, \\ \mathbf{v}_k^\varphi(t, \mathbf{y}) &:= \mathbf{v}_k(t, \mathbf{x}) \quad \text{for } \mathbf{y} = \boldsymbol{\varphi}_k(t, \mathbf{x}). \end{aligned}$$

In this context index φ denotes a field in Eulerian description, while a field without this index is formulated in Lagrangian description.

Note, if γ^φ is any smooth field in the actual configuration, the spatial time derivative is denoted by $\dot{\gamma}^{\varphi'}(t, \mathbf{y}) := \frac{\partial}{\partial t} \gamma^\varphi(t, \mathbf{y})$. In addition, if γ is the appropriate field in the reference configuration, then the material time derivative is denoted by $\dot{\gamma}(t, \mathbf{x}) := \frac{\partial}{\partial t} \gamma(t, \mathbf{x})$. We denote by $D_{\mathbf{y}}$ the derivative with respect to \mathbf{y} and apply the chain rule, then it follows directly,

$$\dot{\gamma}(t, \mathbf{x}) = \dot{\gamma}^{\varphi'}(t, \mathbf{y}) + D_{\mathbf{y}} \gamma^\varphi(t, \mathbf{y}) \mathbf{v}_k^\varphi(t, \mathbf{y}) \quad \text{for } \mathbf{y} = \boldsymbol{\varphi}_k(t, \mathbf{x}). \quad (2.1)$$

In solid mechanics the study of strain is of great interest, whereas the geometrical aspects of strain are explained in [17]. Similar to the theory of single materials we need a deformation gradient to analyse the strain.

Definition 2.8. The deformation gradient of continuum k is given by the smooth tensor field

$$\begin{aligned} \mathbf{F}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^{d \times d}, \\ \mathbf{F}_k(t, \mathbf{x}) &:= D_{\mathbf{x}} \boldsymbol{\varphi}_k(t, \mathbf{x}). \end{aligned}$$

Because $\boldsymbol{\varphi}_k$ is an orientation preserving mapping, from definition 2.3 and definition 2.8 follows directly

$$\det(\mathbf{F}_k) > 0 \quad \forall t \in \mathbb{R}^+, \forall \mathbf{x} \in \Omega, \quad (2.2)$$

and as consequence of the chain rule we derive from definition 2.8

$$\mathbf{F}_k^{-1}(t, \mathbf{x}) = D_{\mathbf{y}} \boldsymbol{\varphi}_k^{-1}(t, \mathbf{y}) \quad \text{for } \mathbf{y} = \boldsymbol{\varphi}_k(t, \mathbf{x}). \quad (2.3)$$

Further there exists a relation between the displacement and the deformation gradient. In view of displacement's definition 2.4 and definition 2.8 it is easy to see

$$\mathbf{F}_k = \mathbf{I} + D_{\mathbf{x}} \mathbf{u}_k. \quad (2.4)$$

The introduction of all basic kinematic quantities for each single continuum is finished, but additionally the kinematics of the mixture are of great interest. In view of the principle of overlapping continua we define all kinematic quantities of the mixture by an averaging technique using

the distribution of mass of different continua. This averaging technique is standard and often used in mixture theory, see [1, 2, 31].

By $\mathfrak{B}(\Omega^\varphi(t))$ we denote the Borel- σ -algebra of the subset $\Omega^\varphi(t) \subset \mathbb{R}^d$ as defined in [10]. Similar to [17, 31, 32] the mass is assumed to be given by a bulk density in Eulerian description.

Condition 2.9. The mass $M_k(\mathcal{P}^\varphi(t))$ of continuum k at time $t \in \mathbb{R}^+$ in any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by a smooth scalar valued bulk density function in the actual configuration

$$\begin{aligned} \rho_k^\varphi : \mathcal{T} &\longrightarrow \mathbb{R}^+, \\ M_k(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \rho_k^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

A common property of all conservation quantities is their additivity. Mass is an conservation quantity and thus the definition of mixture's bulk density is quite natural.

Definition 2.10. The bulk density of the mixture is given by a smooth density function in the actual configuration

$$\begin{aligned} \rho^\varphi : \mathcal{T} &\longrightarrow \mathbb{R}^+, \\ \rho^\varphi(t, \mathbf{y}) &:= \sum_{k=1}^K \rho_k^\varphi(t, \mathbf{y}). \end{aligned}$$

The additivity of mass as well as the linearity of integrals show, that for mixture's mass $M(\mathcal{P}^\varphi(t))$ at time $t \in \mathbb{R}^+$ in a subset $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ holds

$$M(\mathcal{P}^\varphi(t)) = \sum_{k=1}^K M_k(\mathcal{P}^\varphi(t)) = \sum_{k=1}^K \int_{\mathcal{P}^\varphi(t)} \rho_k^\varphi \, d\mathbf{y} = \int_{\mathcal{P}^\varphi(t)} \rho^\varphi \, d\mathbf{y}. \quad (2.5)$$

In order to describe the structure of the mixture, we need a quantity, which represents the percentage amount of continuum k 's mass inside the whole mixture.

Definition 2.11. The mass concentration of continuum k in the actual configuration is given by the smooth scalar valued function

$$\begin{aligned} c_k^\varphi : \mathcal{T} &\longrightarrow [0, 1], \\ c_k^\varphi(t, \mathbf{y}) &:= \frac{\rho_k^\varphi(t, \mathbf{y})}{\rho^\varphi(t, \mathbf{y})}. \end{aligned}$$

Note, that c_k^φ is well defined, because in view of mixture's bulk density given by definition 2.10 it holds $\rho^\varphi > 0$. The mapping properties of c_k^φ follow directly from condition 2.9 and definition 2.10. In addition the mass concentration satisfies a further condition, which is obvious in view of definition 2.10 and definition 2.11,

$$\sum_{k=1}^K c_k^\varphi = 1. \quad (2.6)$$

On the one hand c_k^φ describes the structure of the mixture, hence it will be a unknown quantity in the material model. On the other hand, we will use c_k^φ like in [1, 2, 31] for averaging the kinematic quantities, because it satisfies relation (2.6).

A mean velocity of the mixture is defined by averaging the velocities of all continua with respect to the distribution of mass, see [1, 2, 31].

Definition 2.12. The mass averaged velocity of the mixture in the actual configuration is defined by the smooth vector field

$$\begin{aligned} \mathbf{v}^\varphi : \mathcal{T} &\longrightarrow \mathbb{R}^d, \\ \mathbf{v}^\varphi(t, \mathbf{y}) &:= \sum_{k=1}^K c_k^\varphi(t, \mathbf{y}) \mathbf{v}_k^\varphi(t, \mathbf{y}). \end{aligned}$$

The definition of a mass averaged velocity makes it possible to declare a mass averaged motion of the mixture. This will be done by the following ordinary differential equations:

Problem 2.13. Find $\varphi \in C^1(\mathbb{R}^+ \times \Omega; \mathbb{R}^d)$, the mass averaged motion of the mixture, such that

$$\frac{\partial}{\partial t} \varphi = \mathbf{v}^\varphi(t, \mathbf{x}) \quad \text{with } \varphi(0, \mathbf{x}) = \mathbf{x}.$$

Definition 2.12 provides, that \mathbf{v}^φ is a smooth vector field and it holds at least $\mathbf{v}^\varphi \in C^1(\mathcal{I}; \mathbb{R}^d)$. From Picard-Lindelöf's theorem, see [33] follows for all $\mathbf{x} \in \Omega$ the existence of a unique solution $\varphi \in C^1(\mathbb{R}^+; \mathbb{R}^d)$. Moreover, while $\mathbf{v}^\varphi \in C^1(\mathbb{R}^+ \times \Omega; \mathbb{R}^d)$ we can determine, that $\varphi \in C^1(\mathbb{R}^+ \times \Omega; \mathbb{R}^d)$. The initial condition and smoothness of φ yield $D_{\mathbf{x}}\varphi(0, \mathbf{x}) = \mathbf{I}$ and thus $\det(D_{\mathbf{x}}\varphi(0, \mathbf{x})) = 1 > 0$. On the other hand from [17] it is well known, that \det is a smooth mapping from the set of invertible matrices denoted by $\text{GL}(\mathbb{R}^d)$ into the real numbers

$$\begin{aligned} \det &: \text{GL}(\mathbb{R}^d) \longrightarrow \mathbb{R}, \\ D_{\mathbf{A}} \det(\mathbf{A})[\mathbf{U}] &= \det(\mathbf{A}) \text{tr}(\mathbf{U}\mathbf{A}^{-1}). \end{aligned}$$

Using this relation, a Taylor expansion of the function \det leads to

$$\begin{aligned} \det(D_{\mathbf{x}}\varphi(t, \mathbf{x})) &= \det(D_{\mathbf{x}}\varphi(0, \mathbf{x})) + D_{\mathbf{A}} \det(D_{\mathbf{x}}\varphi(0, \mathbf{x}))[D_{\mathbf{x}}\varphi(t, \mathbf{x}) - D_{\mathbf{x}}\varphi(0, \mathbf{x})] \\ &\quad + \mathcal{O}(\|D_{\mathbf{x}}\varphi(t, \mathbf{x}) - D_{\mathbf{x}}\varphi(0, \mathbf{x})\|) \\ &= 1 + \text{tr}(D_{\mathbf{x}}\varphi(t, \mathbf{x}) - D_{\mathbf{x}}\varphi(0, \mathbf{x})) + \mathcal{O}(\|D_{\mathbf{x}}\varphi(t, \mathbf{x}) - D_{\mathbf{x}}\varphi(0, \mathbf{x})\|). \end{aligned}$$

In the case of small deformations we have $\det(D_{\mathbf{x}}\varphi(t, \mathbf{x})) > 0$ and so φ is a motion in the sense of definition 2.3. This consideration enables us to define all kinematic quantities for the mixture in a mass averaged sense.

Definition 2.14. The mass averaged velocity of the mixture in the reference configuration is consequently given by the smooth vector field

$$\begin{aligned} \mathbf{v} &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \mathbf{v}(t, \mathbf{x}) &:= \frac{\partial}{\partial t} \varphi(t, \mathbf{x}). \end{aligned}$$

Definition 2.15. The mass averaged displacement of the mixture is the smooth vector field defined by

$$\begin{aligned} \mathbf{u} &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \mathbf{u}(t, \mathbf{x}) &:= \varphi(t, \mathbf{x}) - \mathbf{x}. \end{aligned}$$

In order to analyse the strain of the solid mixture an introduction of a deformation gradient of the mass averaged motion is necessary.

Definition 2.16. The mass averaged deformation gradient of the mixture is the smooth tensor field defined as follows

$$\begin{aligned} \mathbf{F} &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \mathbf{F}(t, \mathbf{x}) &:= D_{\mathbf{x}}\varphi(t, \mathbf{x}). \end{aligned}$$

From definition 2.15 and definition 2.16 we can derive a relation between the deformation gradient of the mixture and the mass averaged displacement.

$$\mathbf{F} = \mathbf{I} + D_{\mathbf{x}}\mathbf{u}. \tag{2.7}$$

Focus the attention on the change of length inside the solid mixture, the appropriate quantity for this purpose is the Cauchy-Green's strain tensor as defined in [17].

Definition 2.17. The left Cauchy-Green strain tensor of the mixture in the reference configuration is given by the smooth tensor field

$$\begin{aligned} \mathbf{C} &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^{d \times d}, \\ \mathbf{C}(t, \mathbf{x}) &:= \mathbf{F}(t, \mathbf{x})\mathbf{F}^T(t, \mathbf{x}). \end{aligned}$$

Note, that there exists also a right Cauchy-Green strain tensor, see [17], but in this paper we make no use of it. Later we assume very small deformation of the solid mixture, hence we can work with linearised strains.

Definition 2.18. The linearised strain tensor of the mixture in the reference configuration is a smooth, symmetric tensor field given by

$$\begin{aligned} \boldsymbol{\varepsilon} &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}_{\text{sym}}^{d \times d}, \\ \boldsymbol{\varepsilon}(t, \boldsymbol{x}) &:= \frac{1}{2} (\mathbf{D}_{\boldsymbol{x}} \mathbf{u}(t, \boldsymbol{x}) + \mathbf{D}_{\boldsymbol{x}} \mathbf{u}^T(t, \boldsymbol{x})). \end{aligned}$$

From definition 2.15 and definition 2.18 we can get another representation of the Cauchy-Green strain tensor

$$\mathbf{C} = \mathbf{I} + 2\boldsymbol{\varepsilon} + (\mathbf{D}_{\boldsymbol{x}} \mathbf{u})^T (\mathbf{D}_{\boldsymbol{x}} \mathbf{u}). \quad (2.8)$$

3. AUXILIARY THEOREMS

In this section we formulate some auxiliary theorems, which are important for further considerations.

In continuum mechanics the conservation principle is a basic concept and permanently used in this paper. Let us consider an arbitrary conservation quantity G , for example mass or energy. In order to formulate Reynold's transport theorem, we assume, that this conservation quantity G is given by some smooth density γ^φ in the actual configuration

$$\begin{aligned} \gamma^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}, \\ G(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \gamma^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

Furthermore, we observe a volume source of this conservation quantity denoted by \hat{G} , for example a mass or an energy supply. We assume similar, that this volume source is given by an integrable density $\hat{\gamma}^\varphi$ in the actual configuration

$$\begin{aligned} \hat{\gamma}^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}, \\ \hat{G}(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \hat{\gamma}^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

The fact, that G denotes a conservation quantity is mathematically described by

$$\frac{d}{dt} G(\mathcal{P}^\varphi(t)) = \hat{G}(\mathcal{P}^\varphi(t)).$$

Theorem 3.1. *Let G a conservation quantity, its density γ^φ a smooth field and this volume source an integrable density $\hat{\gamma}^\varphi$, then it holds for any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$*

$$\frac{d}{dt} G(\mathcal{P}^\varphi(t)) = \int_{\mathcal{P}^\varphi(t)} \gamma^{\varphi'} + \operatorname{div}_{\mathbf{y}}(\gamma^\varphi \mathbf{v}^\varphi) \, d\mathbf{y} = \int_{\mathcal{P}^\varphi} \hat{\gamma}^\varphi \, d\mathbf{y}.$$

Consequently almost everywhere the local formulation is valid

$$\gamma^{\varphi'} + \operatorname{div}_{\mathbf{y}}(\gamma^\varphi \mathbf{v}^\varphi) = \hat{\gamma}^\varphi.$$

Proof. See [17]. □

In continuum mechanics the mathematical modelling starts in general inside the actual configuration and produces a mathematical model in Eulerian coordinates. From a mathematical point of view it is more convenient to formulate the equations inside the fixed reference configuration. The transformation between actual and reference configuration is called Piola's transformation. To formulate this transformation we need the following definition:

Definition 3.2. Let $\mathbf{A} \in \operatorname{GL}(\mathbb{R}^d)$, then the cofactor matrix of \mathbf{A} is defined by

$$\operatorname{Cof}(\mathbf{A}) := \det(\mathbf{A}) \mathbf{A}^{-T}.$$

The first version of Piola's transformation, which we frequently use in this paper, handles vector-valued function. Let $t \in \mathbb{R}^+$ arbitrary, but fixed, then we consider a vector field in the actual configuration as follows

$$\boldsymbol{\gamma}^\varphi(t, \cdot) : \Omega^\varphi(t) \longrightarrow \mathbb{R}^d.$$

Definition 3.3. Piola's transformation of the vector field γ^φ based on the mass averaged deformation gradient \mathbf{F} of the solid mixture is defined by

$$\begin{aligned} \mathbf{P} &: \mathbb{R}^d \longrightarrow \mathbb{R}^d, \\ (\mathbf{P}\gamma^\varphi)(t, \mathbf{x}) &:= \text{Cof}(\mathbf{F}^T)\gamma^\varphi(t, \varphi(t, \mathbf{x})). \end{aligned}$$

For a short notation we denote by $\gamma := (\mathbf{P}\gamma^\varphi)$ the Piola transformed quantity of γ . The Piola transformed quantity satisfies the following important properties.

Lemma 3.4. *For Piola's transformation \mathbf{P} given by definition 3.3 it holds*

$$\begin{aligned} (1) \quad & \text{div}_{\mathbf{x}}(\gamma) = \det(\mathbf{F}) \text{div}_{\mathbf{y}}(\gamma^\varphi). \\ (2) \quad & \int_{\partial\mathcal{D}^\varphi(t)} \gamma^\varphi \cdot \mathbf{n}^\varphi \, d\mathbf{a}_{\mathbf{y}} = \int_{\partial\mathcal{D}(t)} \gamma \cdot \mathbf{n} \, d\mathbf{a}_{\mathbf{x}}. \end{aligned}$$

Proof. See [6]. □

The second version of Piola's transformation, which we use in this paper, transforms tensor-valued functions. Let us consider in this context a tensor field in the actual configuration at a fixed time $t \in \mathbb{R}^+$,

$$\mathbf{T}^\varphi(t, \cdot) : \Omega^\varphi(t) \longrightarrow \mathbb{R}^{d \times d}.$$

Definition 3.5. Piola's transformation of the tensor field \mathbf{T} based on the mass averaged deformation gradient of the solid mixture is defined by

$$\begin{aligned} \mathbf{P} &: \mathbb{R}^{d \times d} \longrightarrow \mathbb{R}^{d \times d}, \\ (\mathbf{P}\mathbf{T})(t, \mathbf{x}) &:= \mathbf{T}^\varphi(t, \varphi(t, \mathbf{x})) \text{Cof}(\mathbf{F}(t, \mathbf{x})). \end{aligned}$$

Similarly, we define by $\mathbf{T}(t, \mathbf{x}) := (\mathbf{P}\mathbf{T}^\varphi)(t, \mathbf{x})$ the Piola transformed quantity of \mathbf{T}^φ . This transformation also satisfies an important property used in the context of conservation equations.

Lemma 3.6. *Piola's transformation given by definition 3.5 satisfies the following properties:*

$$\begin{aligned} (1) \quad & \text{div}_{\mathbf{x}}(\mathbf{T}) = \det(\mathbf{F}) \text{div}_{\mathbf{y}}(\mathbf{T}^\varphi) \\ (2) \quad & \int_{\partial\mathcal{D}^\varphi(t)} \mathbf{T}^\varphi \mathbf{n}^\varphi \, d\mathbf{a}_{\mathbf{y}} = \int_{\mathcal{D}(t)} \mathbf{T} \mathbf{n} \, d\mathbf{a}_{\mathbf{x}}. \end{aligned}$$

Proof. See [6]. □

4. BALANCE OF MASS

The mass of the body is a measure of its inertia, hence we analyse the balance equation of mass for each single continuum and the balance of mass for the whole mixture in the actual configuration. Both considerations show different relations between conservation quantities of single continua and of the mixture, see [1, 2, 31]. In the end the formulation of balance of mass is given by a diffusion equation in the reference configuration, whereas the change into Lagrangian description is done by Piola's transformation, see [17].

4.1. Mass balance of continuum k . The representation of continuum k 's mass is formulated in condition 2.9. In order to have a simple notation we introduce a mass flux density.

Definition 4.1. The mass flux density of continuum k in the actual configuration is a smooth vector field given by

$$\begin{aligned} \mathbf{m}_k^\varphi &: \mathcal{S} \longrightarrow \mathbb{R}^d, \\ \mathbf{m}_k^\varphi(t, \mathbf{y}) &:= \rho_k^\varphi(t, \mathbf{y}) \mathbf{v}_k^\varphi(t, \mathbf{y}). \end{aligned}$$

In general, inside the solid mixture chemical reactions can happen. This fact makes it necessary to introduce a mass supply of continuum k . We pick up the ideas from [31, 32] and assume, that the mass supply is given by a mass production density.

Condition 4.2. The mass supply $\hat{C}_k(\mathcal{P}^\varphi(t))$ of continuum k at time $t \in \mathbb{R}^+$ in any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is represented by an integrable mass supply density

$$\begin{aligned} \hat{c}_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}, \\ \hat{C}_k(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \hat{c}_k^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

Following [17, 31, 32] we postulate the principle of continuum k ' mass conservation as a non closed thermodynamical system.

Postulate 1 (Principle of continuum k 's mass conservation). *For any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ holds*

$$\frac{d}{dt} M_k(\mathcal{P}^\varphi(t)) = \hat{C}_k(\mathcal{P}^\varphi(t)).$$

Lemma 4.3. *Let the principle of mass conservation be satisfied, then the continuity equation of continuum k holds almost everywhere in the actual configuration*

$$\rho_k^{\varphi'} + \operatorname{div}_{\mathbf{y}}(\mathbf{m}_k^\varphi) = \hat{c}_k^\varphi. \quad (4.1)$$

Proof. This assertion follows directly from Reynold's transport theorem 3.1. \square

4.2. Mass balance of the mixture. Definition 2.10 provides a representation of mixture's mass. Similar to single materials, we introduce a mass flux density for the mixture.

Definition 4.4. The mass flux density of the mixture in the actual configuration is given by a smooth vector field

$$\begin{aligned} \mathbf{m}^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^d, \\ \mathbf{m}^\varphi(t, \mathbf{y}) &:= \sum_{k=1}^K \rho_k^\varphi(t, \mathbf{y}) \mathbf{v}_k^\varphi(t, \mathbf{y}) = \rho^\varphi(t, \mathbf{y}) \mathbf{v}^\varphi(t, \mathbf{y}). \end{aligned}$$

The mass flux density of continuum k introduced by definition 4.1 and the mass flux density of the mixture given by definition 4.4 lead to the relation

$$\mathbf{m}^\varphi = \sum_{k=1}^K \mathbf{m}_k^\varphi. \quad (4.2)$$

Similar as for single materials we postulate the principle of mass conservation of the mixture, see [1, 2], but the whole mixture represents a thermodynamical closed system.

Postulate 2 (Principle of mixture's mass conservation). *For any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ holds*

$$\frac{d}{dt} M(\mathcal{P}^\varphi(t)) = 0.$$

Lemma 4.5. *Let the principle of mass conservation be satisfied, then the continuity equation of the mixture holds almost everywhere in the actual configuration*

$$\rho^{\varphi'} + \operatorname{div}_{\mathbf{y}}(\mathbf{m}^\varphi) = 0. \quad (4.3)$$

Proof. This statement follows from Reynold's transport theorem 3.1. \square

Conclusion 4.6. Because mass is an additive quantity, we summarise the continuity equation (4.1) over all continua and make a comparison of coefficients under consideration of definition 2.10 and equation (4.3),

$$\sum_{k=1}^K \hat{c}_k^\varphi = \sum_{k=1}^K \rho_k^{\varphi'} + \operatorname{div}_{\mathbf{y}}(\mathbf{m}_k^\varphi) = \rho^{\varphi'} + \operatorname{div}_{\mathbf{y}}(\mathbf{m}^\varphi) = 0.$$

This computation shows a further result of the principle of mass conservation

$$\sum_{k=1}^K \hat{c}_k^\varphi = 0. \quad (4.4)$$

Conclusion 4.7. The principle of mass conservation of the mixture shows, for $\mathcal{P}^\varphi(t) = \varphi(t, \mathcal{P})$ with any part $\mathcal{P} \in \mathfrak{B}(\Omega)$ that

$$M(\mathcal{P}^\varphi(t)) = M(\mathcal{P}).$$

This relation leads under consideration of definition 2.10 and transformation of integrals to

$$\begin{aligned} M(\mathcal{P}^\varphi(t)) &= \int_{\mathcal{P}^\varphi(t)} \rho^\varphi(t, \mathbf{y}) \, d\mathbf{y} = \int_{\mathcal{P}} \underbrace{\rho^\varphi(t, \varphi(t, \mathbf{x})) \det(\mathbf{F}(t, \mathbf{x}))}_{=:\rho(t, \mathbf{x})} \, d\mathbf{x} \\ &= \int_{\mathcal{P}} \rho \, d\mathbf{x} = M(\mathcal{P}). \end{aligned}$$

Using this computation and take the principle of mass conservation into account, then we determine

$$\frac{d}{dt} M(\mathcal{P}^\varphi(t)) = \frac{d}{dt} M(\mathcal{P}) = \frac{d}{dt} \int_{\mathcal{P}} \rho(t, \mathbf{x}) \, d\mathbf{x} = \int_{\mathcal{P}} \dot{\rho}(t, \mathbf{x}) \, d\mathbf{x} = 0.$$

This relation holds for any part $\mathcal{P} \in \mathfrak{B}(\Omega)$ and Du Bois-Reymond's lemma, see [10], yields almost everywhere

$$\dot{\rho}(t, \mathbf{x}) = 0 \quad \text{and thus} \quad \rho = \rho(\mathbf{x}).$$

Conclusion 4.7 shows, how the bulk densities for different continua and for the mixture in Lagrangian description should be given.

Definition 4.8. The bulk density of continuum k in the reference configuration is given by a smooth density function

$$\begin{aligned} \rho_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^+, \\ \rho_k(t, \mathbf{x}) &:= \rho_k^\varphi(t, \varphi(t, \mathbf{x})) \det(\mathbf{F}(t, \mathbf{x})). \end{aligned}$$

Definition 4.9. The bulk density of the mixture in the reference configuration is formally defined by the smooth density function

$$\begin{aligned} \rho &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^+, \\ \rho(t, \mathbf{x}) &:= \rho^\varphi(t, \varphi(t, \mathbf{x})) \det(\mathbf{F}(t, \mathbf{x})). \end{aligned}$$

But we keep in mind, that inside the reference configuration ρ is independent of t , which is proofed in conclusion 4.7.

4.3. Diffusion equation. The aim of this section is to develop an equation, which describes the evolution of material structure inside the reference configuration. First we declare the relative motion of continuum k based on the mass averaged motion of the mixture as it is done in [1, 2, 31].

Definition 4.10. The diffusion velocity of continuum k in the actual configuration is given by the smooth vector field

$$\begin{aligned} \mathbf{w}_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^d, \\ \mathbf{w}_k^\varphi(t, \mathbf{y}) &:= \mathbf{v}_k^\varphi(t, \mathbf{y}) - \mathbf{v}^\varphi(t, \mathbf{y}). \end{aligned}$$

In conjunction with a diffusive motion a mass transfer takes place. This mass transport is described by the diffusive flux.

Definition 4.11. The diffusive flux of continuum k in the actual configuration is defined by the smooth vector field

$$\begin{aligned} \mathbf{J}_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^d, \\ \mathbf{J}_k^\varphi(t, \mathbf{y}) &:= \rho_k^\varphi(t, \mathbf{y}) \mathbf{w}_k^\varphi(t, \mathbf{y}). \end{aligned}$$

The diffusive fluxes satisfy a property, which is closely related to the principle of mass conservation. We take mixture's density, definition 2.10, the definition of the mass averaged velocity 2.12 as well as definition 4.11 into account and get

$$\sum_{k=1}^K \mathbf{J}_k^\varphi = \sum_{k=1}^K (\rho_k^\varphi \mathbf{v}_k^\varphi - \rho_k^\varphi \mathbf{v}^\varphi) = \rho^\varphi \mathbf{v}^\varphi - \rho^\varphi \mathbf{v}^\varphi = 0. \quad (4.5)$$

In general a diffusive mass transfer across the boundary of the domain has to be regarded.

Condition 4.12. The mass supply across the boundary $\hat{J}_k(\partial\Omega^\varphi(t))$ at time $t \in \mathbb{R}^+$ over $\partial\Omega^\varphi(t)$ is given by an integrable density function

$$\begin{aligned} \hat{j}_k^\varphi &: \mathbb{R}^+ \times \partial\Omega^\varphi(t) \longrightarrow \mathbb{R}, \\ \hat{J}_k(\partial\Omega^\varphi(t)) &:= \int_{\partial\Omega^\varphi(t)} \hat{j}_k^\varphi \, d\mathbf{a}_\mathbf{y}. \end{aligned}$$

On the other hand the diffusive flux produces also a mass transfer across the boundary and this leads to a global formulation of the boundary condition

$$\hat{J}_k(\partial\Omega^\varphi(t)) = \int_{\partial\Omega^\varphi(t)} \hat{j}_k^\varphi \, d\mathbf{a}_\mathbf{y} = \int_{\partial\Omega^\varphi(t)} \mathbf{J}_k^\varphi \cdot \mathbf{n}^\varphi \, d\mathbf{a}_\mathbf{y}.$$

This relation provides a Neumann boundary condition in the actual configuration formulated in a local form

$$\mathbf{J}_k^\varphi \cdot \mathbf{n}^\varphi = \hat{j}_k^\varphi. \quad (4.6)$$

From equation (4.5) and the boundary condition (4.6) we determine an additional condition for the mass supply across the boundary, which is closely related to the mass conservation

$$\sum_{k=1}^K \hat{j}_k^\varphi = 0. \quad (4.7)$$

The formulation of the diffusion equation inside the reference configuration needs a transformation of mass concentration c_k^φ , mass supply density \hat{c}_k^φ , diffusive flux \mathbf{J}_k^φ and mass supply density across the boundary \hat{j}_k^φ into a Lagrangian description.

Note, that mass concentration is independent of the volume of the body and thus the transformation into the reference configuration consists only in a reparameterisation of the field.

Definition 4.13. The mass concentration of continuum k in the reference configuration is given by a smooth scalar valued field

$$\begin{aligned} c_k &: \mathbb{R}^+ \times \Omega \longrightarrow [0, 1], \\ c_k(t, \mathbf{x}) &:= c_k^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})). \end{aligned}$$

Note, that property (2.6) is also satisfied in the reference configuration, because equation (2.6) and definition 4.13 directly yield

$$\sum_{k=1}^K c_k = 1. \quad (4.8)$$

The mass supply strongly depends on the body's volume as condition 4.2 shows.

Definition 4.14. The mass supply density of continuum k in the reference configuration is given by the integrable density function

$$\begin{aligned} \hat{c}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}, \\ \hat{c}_k(t, \mathbf{x}) &:= \hat{c}_k^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})) \det(\mathbf{F}(t, \mathbf{x})). \end{aligned}$$

The transformed mass supply density \hat{c}_k satisfies also property (4.4), which follows from equation (4.4), equation (2.2) and definition 4.14

$$\sum_{k=1}^K \hat{c}_k = 0. \quad (4.9)$$

The diffusive flux is a surface depending quantity, thus the transformation into the reference configuration uses Piola's transformation as defined in definition 3.3.

Definition 4.15. The diffusive flux of continuum k in the reference configuration is given by the smooth vector field

$$\begin{aligned} \mathbf{J}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \mathbf{J}_k(t, \mathbf{x}) &:= (\mathbf{P}\mathbf{J}_k^\varphi)(t, \mathbf{x}). \end{aligned}$$

Note, that this formulation of the diffusive flux in Lagrangian description is consistent with the mass conservation, because equation (4.5), definition 4.15 and the linearity of Piola's transformation yield

$$\sum_{k=1}^K \mathbf{J}_k = 0. \quad (4.10)$$

The diffusive mass supply across the boundary apparently depends on the surface volume. This dependency has to be regarded in view of surface integral transformation.

Definition 4.16. The mass supply density across the boundary of continuum k in the reference configuration is defined by

$$\begin{aligned} \hat{j}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}, \\ \hat{j}_k(t, \mathbf{x}) &= \hat{j}_k^\varphi(t, \varphi(t, \mathbf{x})) |\text{Cof}(\mathbf{F}(t, \mathbf{x}))\mathbf{n}(t, \mathbf{x})|. \end{aligned}$$

This definition and transformation of surface integrals using the cofactor matrix of the deformation gradient given by definition 3.2 yield for the mass supply across the boundary in the reference configuration

$$\hat{J}_k(\partial\Omega^\varphi(t)) = \int_{\partial\Omega} \hat{j}_k \, d\mathbf{a}_{\mathbf{x}} = \int_{\partial\Omega} \mathbf{J}_k \cdot \mathbf{n} \, d\mathbf{a}_{\mathbf{x}}.$$

A localisation of this calculation leads to a Neumann boundary condition in the reference configuration

$$\mathbf{J}_k \cdot \mathbf{n} = \hat{j}_k. \quad (4.11)$$

Additionally, it is obvious, that in view of equation (4.7), mass conservation and definition 4.16 for the mass supply across the boundary holds

$$\sum_{k=1}^K \hat{j}_k = 0. \quad (4.12)$$

In view of equation (4.8), (4.9), (4.10) and (4.12) it directly follows, that the transformed quantities satisfy the principle of mass conservation in the reference configuration.

Lemma 4.17. *Let the principle of mass conservation be satisfied, then the diffusion equation of continuum k in the reference configuration is almost everywhere valid*

$$\rho \dot{c}_k + \text{div}_{\mathbf{x}}(\mathbf{J}_k) = \hat{c}_k. \quad (4.13)$$

Proof. In view of continuum k 's mass concentration given by definition 2.11 and under consideration of the continuity equation (4.3) we derive

$$\begin{aligned} \rho_k^{\varphi'} &= (\rho_k^\varphi c_k^\varphi)' = \rho_k^{\varphi'} c_k^\varphi + \rho_k^\varphi c_k^{\varphi'} = -c_k^\varphi \text{div}_{\mathbf{y}}(\rho_k^\varphi \mathbf{v}^\varphi) + \rho_k^\varphi c_k^{\varphi'} \\ &= -c_k^\varphi \text{div}_{\mathbf{y}}(\rho_k^\varphi \mathbf{v}^\varphi) - \rho_k^\varphi \nabla_{\mathbf{y}} c_k^\varphi \cdot \mathbf{v}^\varphi + \rho_k^\varphi \nabla_{\mathbf{y}} c_k^\varphi \cdot \mathbf{v}^\varphi + \rho_k^\varphi c_k^{\varphi'} \\ &= -\text{div}_{\mathbf{y}}(\rho_k^\varphi \mathbf{v}^\varphi) + \rho_k^\varphi (c_k^{\varphi'} + \nabla_{\mathbf{y}} c_k^\varphi \cdot \mathbf{v}^\varphi). \end{aligned}$$

Inserting this relation into the continuity equation (4.1), using the material time derivative (2.1), definition 4.9, lemma 3.4, definition 4.15 and definition 4.14 then we get

$$\begin{aligned} \int_{\mathcal{P}^\varphi(t)} \rho_k^{\varphi'} + \operatorname{div}_{\mathbf{y}}(\rho_k^\varphi \mathbf{v}_k^\varphi) d\mathbf{y} &= \int_{\mathcal{P}^\varphi(t)} \rho^\varphi (c_k^{\varphi'} + \nabla_{\mathbf{y}} c_k^\varphi \cdot \mathbf{v}^\varphi) + \operatorname{div}_{\mathbf{y}}(\mathbf{J}_k^\varphi) d\mathbf{y} \\ &= \int_{\mathcal{P}} [\rho^\varphi \underbrace{(c_k^{\varphi'} + \nabla_{\mathbf{y}} c_k^\varphi \cdot \mathbf{v}^\varphi)}_{=\hat{c}_k} + \operatorname{div}_{\mathbf{y}}(\mathbf{J}_k^\varphi)] \det(\mathbf{F}) d\mathbf{x} = \int_{\mathcal{P}} \rho \hat{c}_k + \operatorname{div}_{\mathbf{x}}(\mathbf{J}_k) d\mathbf{x} \\ &= \int_{\mathcal{P}^\varphi(t)} \hat{c}_k^\varphi d\mathbf{y} = \int_{\mathcal{P}} \hat{c}_k^\varphi \det(\mathbf{F}) d\mathbf{x} = \int_{\mathcal{P}} \hat{c}_k d\mathbf{x}. \end{aligned}$$

This calculation yields to the relation

$$\int_{\mathcal{P}} \rho \hat{c}_k + \operatorname{div}_{\mathbf{x}}(\mathbf{J}_k) d\mathbf{x} = \int_{\mathcal{P}} \hat{c}_k d\mathbf{x},$$

which holds for all parts $\mathcal{P} \in \mathfrak{B}(\Omega)$ and Du Bois-Reymond's lemma, see [10], shows the assertion. \square

Lemma 4.18. *If there exists no mass supply into continuum k , this means $\hat{c}_k = 0$ and $\hat{j}_k = 0$, then it holds*

$$\int_{\Omega} c_k(t, \mathbf{x}) d\mathbf{x} = \int_{\Omega} c_k(0, \mathbf{x}) d\mathbf{x} = \text{const.}$$

Proof. From the diffusion equation (4.13), Gauß's theorem and the boundary condition (4.11) directly follows

$$\begin{aligned} \frac{d}{dt} \int_{\Omega} c_k(t, \mathbf{x}) d\mathbf{x} &= \int_{\Omega} \dot{c}_k(t, \mathbf{x}) d\mathbf{x} = - \int_{\Omega} \operatorname{div}_{\mathbf{x}}(\mathbf{J}_k) d\mathbf{x} \\ &= - \int_{\partial\Omega} \mathbf{J}_k \cdot \mathbf{n} d\mathbf{a}_{\mathbf{x}} = 0. \end{aligned}$$

This calculation shows, that $\int_{\Omega} c_k(t, \mathbf{x}) d\mathbf{x}$ does not depend on t and the assertion is proofed. \square

Finally, we have to formulate the initial boundary value problem, which describes the diffusion process of continuum k in the reference configuration. This problem consists of the diffusion equation (4.13), the boundary condition (4.11) and an initial condition, whereas the concentrations c_k are the unknown quantities and the diffusive fluxes \mathbf{J}_k must be described by a constitutive relation,

$$\begin{aligned} \rho \hat{c}_k + \operatorname{div}_{\mathbf{x}}(\mathbf{J}_k) &= \hat{c}_k && \text{in } \mathbb{R}^+ \times \Omega, \\ \mathbf{J}_k \cdot \mathbf{n} &= \hat{j}_k && \text{on } \mathbb{R}^+ \times \partial\Omega, \\ c_k(0, \mathbf{x}) &= c_{k,0}(\mathbf{x}) && \text{in } \Omega. \end{aligned}$$

Further the concentrations have to satisfy the constraint $0 \leq c_k \leq 1$ as well as relation (4.8) and the constitutive relations of the diffusive fluxes have to satisfy the constraint (4.10).

5. BALANCE OF LINEAR MOMENTUM

During motion mechanical interactions between parts of the body or between the body and its environment are described by Newtonian forces. In mixture theory we have to analyse the balance of linear momentum of each single continuum as well as the balance of linear momentum of the whole mixture in the actual configuration. The applied techniques used for these considerations are provided in [1, 2, 31]. In the end the equation of motion of the mixture is formulated in the reference configuration by using Piola's transformation, see [17].

5.1. Linear momentum balance of continuum k . On any continuum k there act surface forces as a result of external loadings of the body. Following [17, 31, 32], we assume, that the surface force is defined by a force density.

Condition 5.1. The surface force $\mathbf{S}_k(\partial\mathcal{P}^\varphi(t))$ acting on continuum k at time $t \in \mathbb{R}^+$ on $\partial\mathcal{P}^\varphi(t)$ of $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by the smooth and symmetric Cauchy stress tensor in the actual configuration

$$\begin{aligned} \mathbf{S}_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}_{\text{sym}}^{d \times d}, \\ \mathbf{S}_k(\partial\mathcal{P}^\varphi(t)) &:= \int_{\partial\mathcal{P}^\varphi(t)} \mathbf{S}_k^\varphi(t, \mathbf{y}) \mathbf{n}^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

Note, that the existence and uniqueness of \mathbf{S}_k^φ is proofed by Cauchy's theorem, see [17]. The symmetry of Cauchy's stress tensor is due to the balance of angular momentum, analysed in [17]. Further note, that in general Neumann boundary conditions have to be formulated, which describe the external loading of continuum k , but this will be done only for the whole body.

Moreover, volume forces resulting from external fields like gravitation, electrical or magnetic fields have to be considered and similar to [17, 31, 32] we introduce them by a force density.

Condition 5.2. The volume force $\mathbf{B}_k(\mathcal{P}^\varphi(t))$ acting on continuum k at time $t \in \mathbb{R}^+$ in $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi)$ is given in the actual configuration by an integrable specific force density

$$\begin{aligned} \mathbf{b}_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^d, \\ \mathbf{B}_k(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \rho_k^\varphi(t, \mathbf{y}) \mathbf{b}_k^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

Modelling a mixture, we have to take into account interaction forces between continuum k and any other continua. This is also done in [1, 2, 31], where also a representation by a force density is assumed.

Condition 5.3. The interaction force $\mathbf{I}_k(\mathcal{P}^\varphi(t))$ acting on continuum k at time $t \in \mathbb{R}^+$ in $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by an integrable force density

$$\begin{aligned} \mathbf{i}_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^d, \\ \mathbf{I}_k(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \mathbf{i}_k^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

Definition 5.4. The linear momentum $\mathbf{P}_k(\mathcal{P}^\varphi(t))$ of continuum k at time $t \in \mathbb{R}^+$ in $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by

$$\mathbf{P}_k(\mathcal{P}^\varphi(t)) := \int_{\mathcal{P}^\varphi(t)} \rho_k^\varphi(t, \mathbf{y}) \mathbf{v}_k^\varphi(t, \mathbf{y}) \, d\mathbf{y}.$$

Further, a supply of linear momentum of continuum k results from the mass production in continuum k and from continuum k 's velocity, see [1, 2, 31].

Definition 5.5. The linear momentum supply $\hat{\mathbf{P}}_k(\mathcal{P}^\varphi(t))$ of continuum k at time $t \in \mathbb{R}^+$ in $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is defined by

$$\hat{\mathbf{P}}_k(\mathcal{P}^\varphi(t)) := \int_{\mathcal{P}^\varphi(t)} \hat{c}_k^\varphi(t, \mathbf{y}) \mathbf{v}_k^\varphi(t, \mathbf{y}) \, d\mathbf{y}.$$

We postulate as usual in continuum mechanics the principle of linear momentum conservation, see [17, 31, 32].

Postulate 3 (Principle of continuum k 's linear momentum conservation). *For any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ holds*

$$\frac{d}{dt} \mathbf{P}_k(\mathcal{P}^\varphi(t)) = \mathbf{S}_k(\partial\mathcal{P}^\varphi(t)) + \mathbf{B}_k(\mathcal{P}^\varphi(t)) + \mathbf{I}_k(\mathcal{P}^\varphi(t)) + \hat{\mathbf{P}}_k(\mathcal{P}^\varphi(t)).$$

Lemma 5.6. *Let the principle of linear momentum conservation be satisfied, then for continuum k the equation of motion in the actual configuration holds almost everywhere*

$$(\rho_k^\varphi \mathbf{v}_k^\varphi)' + \text{div}_{\mathbf{y}}(\mathbf{m}_k^\varphi \otimes \mathbf{v}_k^\varphi) = \text{div}_{\mathbf{y}}(\mathbf{S}_k^\varphi) + \rho_k^\varphi \mathbf{b}_k^\varphi + \mathbf{i}_k^\varphi + \hat{c}_k^\varphi \mathbf{v}_k^\varphi. \quad (5.1)$$

Proof. This statement follows from Gauß's theorem, see [23], and Reynold's transport theorem 3.1. \square

Note, that the summary of all forces acting on continuum k provide a mechanical power, which is derived by

$$P_{\text{mech},k}(\mathcal{P}^\varphi(t)) = \int_{\partial\mathcal{P}^\varphi(t)} \mathbf{S}_k^\varphi \mathbf{n}^\varphi \cdot \mathbf{v}_k^\varphi \, d\mathbf{a}_\mathbf{y} + \int_{\mathcal{P}^\varphi(t)} \rho_k^\varphi \mathbf{b}_k^\varphi \cdot \mathbf{v}_k^\varphi + \mathbf{i}_k^\varphi \cdot \mathbf{v}_k^\varphi + 1/2 \hat{c}_k^\varphi \mathbf{v}_k^\varphi{}^2 \, d\mathbf{y}. \quad (5.2)$$

5.2. Linear momentum balance of the mixture. The force acting on the whole body measures the external actions, which lead to its deformation. Similar to [1, 2, 31] we assume a surface traction of the mixture defined by a surface force density.

Condition 5.7. The surface force $\mathbf{S}(\partial\mathcal{P}^\varphi(t))$ acting on the mixture at time $t \in \mathbb{R}^+$ on $\partial\mathcal{P}^\varphi(t)$ for any $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by the smooth and symmetric Cauchy stress tensor

$$\begin{aligned} \mathbf{S}^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}_{\text{sym}}^{d \times d}, \\ \mathbf{S}(\partial\mathcal{P}^\varphi(t)) &:= \int_{\partial\mathcal{P}^\varphi(t)} \mathbf{S}^\varphi(t, \mathbf{y}) \mathbf{n}^\varphi(t, \mathbf{y}) \, d\mathbf{a}_\mathbf{y}. \end{aligned}$$

Note as before, that the existence and uniqueness of Cauchy's stress tensor is proofed by Cauchy's theorem and the symmetry results from the balance of angular momentum, see [17].

We divide the boundary $\partial\Omega$ of the reference body into a Dirichlet part Γ_D and a Neumann part Γ_N . This partition has to satisfy the properties

$$\bar{\Gamma}_D \cup \bar{\Gamma}_N = \partial\Omega \quad \text{and} \quad \Gamma_D \cap \Gamma_N = \emptyset.$$

In the actual configuration the Dirichlet boundary is given by $\Gamma_D^\varphi(t) := \varphi(t, \Gamma_D)$ and similar the Neumann boundary is defined by $\Gamma_N^\varphi(t) := \varphi(t, \Gamma_N)$. Note, that a mapping φ satisfying definition 2.3 carries the boundary of a set into the boundary of the image set, see [17].

Condition 5.8. The surface force $\hat{\mathbf{G}}(\Gamma_N^\varphi(t))$ acting on the mixture at time $t \in \mathbb{R}^+$ on $\Gamma_N^\varphi(t)$ in the actual configuration is given by an integrable surface force density

$$\begin{aligned} \hat{\mathbf{g}}^\varphi &: \mathbb{R}^+ \times \Gamma_N^\varphi(t) \longrightarrow \mathbb{R}^d, \\ \hat{\mathbf{G}}(\Gamma_N^\varphi(t)) &:= \int_{\Gamma_N^\varphi(t)} \hat{\mathbf{g}}^\varphi(t, \mathbf{y}) \, d\mathbf{a}_\mathbf{y}. \end{aligned}$$

On the other hand, surfaces forces are represented by Cauchy's stress tensor, where condition 5.8 provides a Neumann boundary condition

$$\mathbf{S}^\varphi \mathbf{n}^\varphi = \hat{\mathbf{g}}^\varphi. \quad (5.3)$$

The external loading of the whole body in the actual configuration is illustrated in figure 3.

Volume forces are additive quantities, hence we will use the mass averaging technique from [1, 2, 31] to define mixture's volume force density.

Definition 5.9. The volume force density of the mixture in the actual configuration is given by an integrable function

$$\begin{aligned} \mathbf{b}^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^d, \\ \mathbf{b}^\varphi(t, \mathbf{y}) &:= \sum_{k=1}^K c_k^\varphi(t, \mathbf{y}) \mathbf{b}_k^\varphi(t, \mathbf{y}). \end{aligned}$$

The additivity of forces and the linearity of integrals yield for the volume forces $\mathbf{B}(\mathcal{P}^\varphi(t))$ of the mixture at time $t \in \mathbb{R}^+$ in any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$

$$\mathbf{B}(\mathcal{P}^\varphi(t)) = \sum_{k=1}^K \mathbf{B}_k(\mathcal{P}^\varphi(t)) = \sum_{k=1}^K \int_{\mathcal{P}^\varphi(t)} \rho_k^\varphi \mathbf{b}_k^\varphi \, d\mathbf{y} = \int_{\mathcal{P}^\varphi(t)} \rho^\varphi \mathbf{b}^\varphi \, d\mathbf{y}.$$

Note, that there exist no interaction forces for the mixture and no linear momentum supply due to mass production.

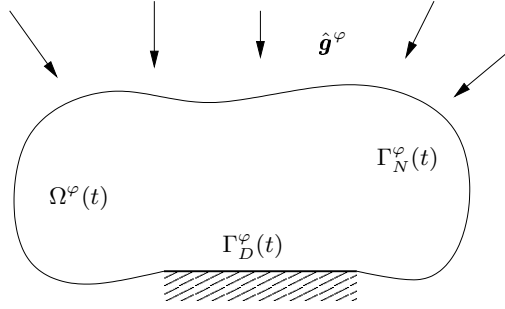


FIGURE 3. external loading of the body

Definition 5.10. The linear momentum of the mixture $\mathbf{P}(\mathcal{P}^\varphi(t))$ at time $t \in \mathbb{R}^+$ of a part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by

$$\mathbf{P}(\mathcal{P}^\varphi(t)) := \int_{\mathcal{P}^\varphi(t)} \rho^\varphi(t, \mathbf{y}) \mathbf{v}^\varphi(t, \mathbf{y}) \, d\mathbf{y}.$$

Similar to [17, 31, 32] we postulate the conservation of mixture's linear momentum.

Postulate 4 (Principle of mixture's linear momentum conservation). *For any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ holds*

$$\frac{d}{dt} \mathbf{P}(\mathcal{P}^\varphi(t)) = \mathbf{S}(\partial \mathcal{P}^\varphi(t)) + \mathbf{B}(\mathcal{P}^\varphi(t)).$$

Lemma 5.11. *Let the principle of linear momentum conservation be satisfied, then for the mixture the equation of motion in the actual configuration holds almost everywhere*

$$(\rho^\varphi \mathbf{v}^\varphi)' + \operatorname{div}_{\mathbf{y}}(\mathbf{m}^\varphi \otimes \mathbf{v}^\varphi) = \operatorname{div}_{\mathbf{y}}(\mathbf{S}^\varphi) + \rho^\varphi \mathbf{b}^\varphi. \quad (5.4)$$

Proof. We get this statement as a result from applying Gauß's theorem, see [23], and Reynold's transport theorem 3.1 to the linear momentum conservation. \square

Note, that these forces achieve a mechanical power, which is calculated by

$$P_{\text{mech}}(\mathcal{P}^\varphi(t)) = \int_{\partial \mathcal{P}^\varphi(t)} \mathbf{S}^\varphi \mathbf{n}^\varphi \cdot \mathbf{v}^\varphi \, d\mathbf{a}_{\mathbf{y}} + \int_{\mathcal{P}^\varphi(t)} \rho^\varphi \mathbf{b}^\varphi \cdot \mathbf{v}^\varphi \, d\mathbf{y}. \quad (5.5)$$

The stress of the mixture is related to the stress of the different continua as well as to the diffusive fluxes. The following results also derived in [1, 2, 31] in the Eulerian description.

Lemma 5.12. *Let the principle of linear momentum conservation for continuum k as well as for the mixture be satisfied, then the following relations hold*

$$\mathbf{S}^\varphi = \sum_{k=1}^K [\mathbf{S}_k^\varphi - \mathbf{w}_k^\varphi \otimes \mathbf{J}_k^\varphi], \quad (5.6)$$

$$\sum_{k=1}^K [\hat{c}_k^\varphi \mathbf{v}_k^\varphi + \mathbf{i}_k^\varphi] = 0. \quad (5.7)$$

Proof. This proof is a technical one and divided into two steps.

Step 1: We use definition 2.10 of mixture's bulk density, the mass averaged velocity given by definition 2.12, as well as the diffusion velocity in form of definition 4.10 and consider the following

term

$$\begin{aligned}
\sum_{k=1}^K \rho_k^\varphi \mathbf{w}_k^\varphi \otimes \mathbf{w}_k^\varphi &= \sum_{k=1}^K \rho_k^\varphi (\mathbf{v}_k^\varphi - \mathbf{v}^\varphi) \otimes (\mathbf{v}_k^\varphi - \mathbf{v}^\varphi) \\
&= \sum_{k=1}^K [\rho_k^\varphi \mathbf{v}_k^\varphi \otimes \mathbf{v}_k^\varphi - \rho_k^\varphi \mathbf{v}_k^\varphi \otimes \mathbf{v}^\varphi - \rho_k^\varphi \mathbf{v}^\varphi \otimes \mathbf{v}_k^\varphi + \rho_k^\varphi \mathbf{v}^\varphi \otimes \mathbf{v}^\varphi] \\
&= \sum_{k=1}^K [\rho_k^\varphi \mathbf{v}_k^\varphi \otimes \mathbf{v}_k^\varphi] - \rho^\varphi \mathbf{v}^\varphi \otimes \mathbf{v}^\varphi - \rho^\varphi \mathbf{v}^\varphi \otimes \mathbf{v}^\varphi + \rho^\varphi \mathbf{v}^\varphi \otimes \mathbf{v}^\varphi \\
&= \sum_{k=1}^K [\rho_k^\varphi \mathbf{v}_k^\varphi \otimes \mathbf{v}_k^\varphi] - \rho^\varphi \mathbf{v}^\varphi \otimes \mathbf{v}^\varphi. \tag{5.8}
\end{aligned}$$

Step 2: Because linear momentum is an additive quantity, we summarise the equation of motion (5.1) over all continua, use the mass averaged velocity declared by definition 2.12 and equation (5.8), this yields

$$\begin{aligned}
&\sum_{k=1}^K [(\rho_k^\varphi \mathbf{v}_k^\varphi)' + \operatorname{div}_{\mathbf{y}}(\rho_k^\varphi \mathbf{v}_k^\varphi \otimes \mathbf{v}_k^\varphi)] \\
&= (\rho^\varphi \mathbf{v}^\varphi)' + \operatorname{div}_{\mathbf{y}}(\rho^\varphi \mathbf{v}^\varphi \otimes \mathbf{v}^\varphi) + \sum_{k=1}^K \operatorname{div}_{\mathbf{y}}(\rho_k^\varphi \mathbf{v}_k^\varphi \otimes \mathbf{v}_k^\varphi) - \operatorname{div}_{\mathbf{y}}(\rho^\varphi \mathbf{v}^\varphi \otimes \mathbf{v}^\varphi) \\
&= (\rho^\varphi \mathbf{v}^\varphi)' + \operatorname{div}_{\mathbf{y}}(\rho^\varphi \mathbf{v}^\varphi \otimes \mathbf{v}^\varphi) + \operatorname{div}_{\mathbf{y}} \left(\sum_{k=1}^K \rho_k^\varphi \mathbf{w}_k^\varphi \otimes \mathbf{w}_k^\varphi \right) \\
&= \sum_{k=1}^K [\operatorname{div}_{\mathbf{y}}(\mathbf{S}_k^\varphi) + \rho_k^\varphi \mathbf{b}_k^\varphi + \mathbf{i}_k^\varphi + \hat{c}_k^\varphi \mathbf{v}_k^\varphi] \\
&= \operatorname{div}_{\mathbf{y}} \left(\sum_{k=1}^K \mathbf{S}_k^\varphi \right) + \rho^\varphi \mathbf{b}^\varphi + \sum_{k=1}^K [\mathbf{i}_k^\varphi + \hat{c}^\varphi \mathbf{v}_k^\varphi]
\end{aligned}$$

The above calculations provide a different description of the balance equation of linear momentum under consideration of the diffusive flux defined by definition 4.11

$$(\rho^\varphi \mathbf{v}^\varphi)' + \operatorname{div}_{\mathbf{y}}(\rho^\varphi \mathbf{v}^\varphi \otimes \mathbf{v}^\varphi) = \operatorname{div}_{\mathbf{y}} \left(\sum_{k=1}^K \mathbf{S}_k^\varphi - \mathbf{w}_k^\varphi \otimes \mathbf{J}_k^\varphi \right) + \rho^\varphi \mathbf{b}^\varphi + \sum_{k=1}^K [\mathbf{i}_k^\varphi + \hat{c}_k^\varphi \mathbf{v}_k^\varphi]. \tag{5.9}$$

A comparison of coefficients between the equation of motion (5.4) and equation (5.9) leads to the assertion. \square

5.3. Equation of motion. From a mathematical point of view it is convenient to turn the considerations into the Lagrangian description, to have a fixed domain. Therefore we have to transform all forces into the reference configuration.

Cauchy's stress tensor represents surface forces, thus Piola's transformation introduced by definition 3.5 transfers the stress tensor into the reference configuration, see [17].

Definition 5.13. The 1. Piola-Kirchoff stress tensor of continuum k in the reference configuration is defined by the smooth tensor field

$$\begin{aligned}
\mathbf{S}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^{d \times d}, \\
\mathbf{S}_k(t, \mathbf{x}) &:= (\mathbf{P}\mathbf{S}_k^\varphi)(t, \mathbf{x}).
\end{aligned}$$

Similar, the 1. Piola-Kirchoff stress tensor of the mixture in the reference configuration is given by the smooth tensor field

$$\begin{aligned}
\mathbf{S} &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^{d \times d}, \\
\mathbf{S}(t, \mathbf{x}) &:= (\mathbf{P}\mathbf{S}^\varphi)(t, \mathbf{x}).
\end{aligned}$$

Note, that the 1. Piola-Kirchoff stress tensor in general is not symmetric, but it is easy to see in view of definition 5.13 and in view of \mathbf{S}^φ 's symmetry, that

$$\mathbf{S}\mathbf{F}^T = \mathbf{F}\mathbf{S}^T.$$

The linearity of Piola's transformation shows, that property (5.6) holds also for the 1. Piola-Kirchoff stress tensor and the diffusive flux in Lagrangian description

$$\mathbf{S} = \sum_{k=1}^K [\mathbf{S}_k - \mathbf{w}_k \otimes \mathbf{J}_k]. \quad (5.10)$$

The surface loading of the body introduced by condition 5.8 is still a surface depending quantity. Thus the transformation into the reference configuration works similar as the transformation of surface integrals using the cofactor matrix of the deformation gradient given by definition 3.2, see [17].

Definition 5.14. The surface loading of the mixture on the boundary part Γ_N in the reference configuration is defined by the integrable force density

$$\begin{aligned} \hat{\mathbf{g}} &: \mathbb{R}^+ \times \Gamma_N \longrightarrow \mathbb{R}^d, \\ \hat{\mathbf{g}}(t, \mathbf{x}) &:= \hat{\mathbf{g}}^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})) |\text{Cof}(\mathbf{F}(t, \mathbf{x}))\mathbf{n}(t, \mathbf{x})|. \end{aligned}$$

Definition 5.14 yields for the surface loading under transformation of surface integrals the relation

$$\hat{\mathbf{G}}(\Gamma_N^\varphi(t)) = \int_{\Gamma_N} \hat{\mathbf{g}}(t, \mathbf{x}) \, d\mathbf{a}_\mathbf{x} = \int_{\Gamma_N} \mathbf{S}(t, \mathbf{x})\mathbf{n}(t, \mathbf{x}) \, d\mathbf{a}_\mathbf{x}.$$

This calculation leads to a Neumann boundary condition in the reference configuration given by

$$\mathbf{S}\mathbf{n} = \hat{\mathbf{g}}. \quad (5.11)$$

The volume forces are formulated by condition 5.2 in a mass independent way, hence the transformation is defined only by a reparameterisation of the field.

Definition 5.15. The volume force density acting on continuum k in the reference configuration is given by the integrable vector field

$$\begin{aligned} \mathbf{b}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \mathbf{b}_k(t, \mathbf{x}) &:= \mathbf{b}_k^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})); \end{aligned}$$

the volume force density acting on the mixture is defined in the reference configuration by the integrable vector field

$$\begin{aligned} \mathbf{b} &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \mathbf{b}(t, \mathbf{x}) &:= \mathbf{b}^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})). \end{aligned}$$

The mass averaging of mixture's volume force density holds also in the reference configuration, this can be seen directly by definition 5.9 and definition 5.15,

$$\mathbf{b}(t, \mathbf{x}) = \sum_{k=1}^K c_k(t, \mathbf{x})\mathbf{b}_k(t, \mathbf{x}). \quad (5.12)$$

The interaction force of continuum k is not defined in a specific way, see condition 5.3, hence we have to consider the volume change by transformation.

Definition 5.16. The interaction force density of continuum k is given by an integrable vector field

$$\begin{aligned} \mathbf{i}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \mathbf{i}_k(t, \mathbf{x}) &:= \mathbf{i}_k^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})) \det(\mathbf{F}(t, \mathbf{x})). \end{aligned}$$

Note, that the mass supply given by definition 4.14 and the interaction force given by definition 5.16 satisfy property (5.7) in the reference configuration,

$$\sum_{k=1}^K [\hat{c}_k \mathbf{v}_k + \mathbf{i}_k] = 0.$$

Lemma 5.17. *Let the principle of mass conservation and the principle of linear momentum conservation be satisfied, then mixture's equation of motion in the reference configuration holds almost everywhere*

$$\rho \ddot{\mathbf{u}} = \operatorname{div}_{\mathbf{x}}(\mathbf{S}) + \rho \mathbf{b}. \quad (5.13)$$

Proof. At first we consider the left hand side of the equation of motion (5.4) and apply the balance equation of mass (4.3) as well as the material time derivative (2.1), then it follows

$$\begin{aligned} & \int_{\mathcal{D}^\varphi(t)} (\rho^\varphi \mathbf{v}^\varphi)' + \operatorname{div}_{\mathbf{y}}(\rho^\varphi \mathbf{v}^\varphi \otimes \mathbf{v}^\varphi) \, d\mathbf{y} \\ &= \int_{\mathcal{D}^\varphi(t)} \rho^{\varphi'} \mathbf{v}^\varphi + \rho^\varphi \mathbf{v}^{\varphi'} + \operatorname{div}_{\mathbf{y}}(\rho^\varphi \mathbf{v}^\varphi) \mathbf{v}^\varphi + \rho^\varphi \mathbf{D}_{\mathbf{y}}(\mathbf{v}^\varphi) \mathbf{v}^\varphi \, d\mathbf{y} \\ &= \int_{\mathcal{D}} \rho^\varphi \underbrace{(\mathbf{v}^{\varphi'} + \mathbf{D}_{\mathbf{y}}(\mathbf{v}^\varphi) \mathbf{v}^\varphi)}_{=\dot{\mathbf{v}}} \det(\mathbf{F}) \, d\mathbf{x} = \int_{\mathcal{D}} \rho \dot{\mathbf{v}} \, d\mathbf{x}. \end{aligned}$$

On the other hand, the equation of motion and lemma 3.6 lead to

$$\begin{aligned} \int_{\mathcal{D}^\varphi(t)} \operatorname{div}_{\mathbf{y}}(\mathbf{S}^\varphi) + \rho^\varphi \mathbf{b}^\varphi \, d\mathbf{y} &= \int_{\mathcal{D}} [\operatorname{div}_{\mathbf{y}}(\mathbf{S}^\varphi) + \rho^\varphi \mathbf{b}^\varphi] \det(\mathbf{F}) \, d\mathbf{x} \\ &= \int_{\mathcal{D}} \operatorname{div}_{\mathbf{x}}(\mathbf{S}) + \rho \mathbf{b} \, d\mathbf{x}. \end{aligned}$$

The summary of both calculations under consideration of equation (5.4) yields

$$\int_{\mathcal{D}} \rho \dot{\mathbf{v}} \, d\mathbf{x} = \int_{\mathcal{D}} \operatorname{div}_{\mathbf{x}}(\mathbf{S}) + \rho \mathbf{b} \, d\mathbf{x}.$$

This equation holds for any part $\mathcal{D} \in \mathfrak{B}(\Omega)$ and Du Bois-Reymond's lemma, see [10], and the mass averaged displacement given by definition 2.15 yield the assertion. \square

In continuum mechanics it is desirable to work with a symmetric stress tensor. This can be done by the 2. Piola-Kirchoff stress tensor defined in [17].

Definition 5.18. The 2. Piola-Kirchoff stress tensor of continuum k is given by a smooth tensor field in the reference configuration

$$\begin{aligned} \boldsymbol{\sigma}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}_{\text{sym}}^{d \times d}, \\ \boldsymbol{\sigma}_k(t, \mathbf{x}) &:= \mathbf{F}^{-1}(t, \mathbf{x}) \mathbf{S}_k(t, \mathbf{x}). \end{aligned}$$

Similar, the 2. Piola-Kirchoff stress tensor of the mixture is defined by a smooth tensor field in Lagrangian description

$$\begin{aligned} \boldsymbol{\sigma} &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}_{\text{sym}}^{d \times d}, \\ \boldsymbol{\sigma}(t, \mathbf{x}) &:= \mathbf{F}^{-1}(t, \mathbf{x}) \mathbf{S}(t, \mathbf{x}). \end{aligned}$$

Using the definition of the 2. Piola-Kirchoff stress tensor as well as the diffusive flux in Lagrangian description and relation (5.10), then it holds in the reference configuration

$$\boldsymbol{\sigma} = \sum_{k=1}^K \left[\boldsymbol{\sigma}_k - \frac{1}{\rho_k} \mathbf{J}_k \otimes \mathbf{J}_k \right]. \quad (5.14)$$

In view of the 2. Piola-Kirchoff stress tensor given by definition 5.18 we can reformulate the equation of motion (5.13) by

$$\rho \ddot{\mathbf{u}} = \operatorname{div}_{\mathbf{x}}(\mathbf{F} \boldsymbol{\sigma}) + \rho \mathbf{b}.$$

Later on, the deformation as well as the strain is assumed to be very small, this means $D_{\mathbf{x}}\mathbf{u} < \varepsilon$, and therefore we conclude the linearised equation of motion

$$\rho\ddot{\mathbf{u}} = \operatorname{div}_{\mathbf{x}}(\boldsymbol{\sigma}) + \rho\mathbf{b}. \quad (5.15)$$

In the same way we formulate a linearised version of boundary condition (5.11). A further remark is, that as a consequence of Piola's transformation in view of lemma 3.4 and lemma 3.6 for the mechanical power in the reference configuration holds

$$P_{\text{mech},k}(\mathcal{P}^\varphi(t)) = P_{\text{mech},k}(\mathcal{P}) = \int_{\partial\mathcal{P}} \mathbf{S}_k \mathbf{n} \cdot \mathbf{v}_k \, d\mathbf{a}_{\mathbf{x}} + \int_{\mathcal{P}} \rho_k \mathbf{b}_k \cdot \mathbf{v}_k + \mathbf{i}_k \cdot \mathbf{v}_k \, d\mathbf{x}, \quad (5.16)$$

$$P_{\text{mech},k}(\mathcal{P}^\varphi(t)) = P_{\text{mech},k}(\mathcal{P}) = \int_{\partial\mathcal{P}} \mathbf{S} \mathbf{n} \cdot \mathbf{v} \, d\mathbf{a}_{\mathbf{x}} + \int_{\mathcal{P}} \rho \mathbf{b} \cdot \mathbf{v} \, d\mathbf{x}. \quad (5.17)$$

Finally, we have to formulate the initial boundary value problem of mixture's deformation in the reference configuration. This problem is described by the linearised equation of motion (5.15), a Dirichlet condition describing a given deformation, the linearised boundary loading (5.11) and two initial conditions for the deformation. In this initial boundary value problem the deformation \mathbf{u} is the unknown quantity, while we have to determine constitutive relations between the 2. Piola-Kirchoff stress tensor $\boldsymbol{\sigma}_k$, the strain $\boldsymbol{\varepsilon}$ and the diffusive fluxes \mathbf{J}_k ,

$$\begin{aligned} \rho\ddot{\mathbf{u}} - \operatorname{div}_{\mathbf{x}}(\boldsymbol{\sigma}) &= \rho\mathbf{b} && \text{in } \mathbb{R}^+ \times \Omega, \\ \boldsymbol{\sigma} &= \sum_{k=1}^K [\boldsymbol{\sigma}_k - 1/\rho_k \mathbf{J}_k \otimes \mathbf{J}_k] && \text{in } \mathbb{R}^+ \times \Omega, \\ \boldsymbol{\varepsilon} &= 1/2(D_{\mathbf{x}}\mathbf{u} + (D_{\mathbf{x}}\mathbf{u})^T) && \text{in } \mathbb{R}^+ \times \Omega, \\ \mathbf{u} &= \hat{\mathbf{u}} && \text{on } \mathbb{R}^+ \times \Gamma_D, \\ \boldsymbol{\sigma} \mathbf{n} &= \hat{\mathbf{g}} && \text{on } \mathbb{R}^+ \times \Gamma_N, \\ \mathbf{u}(0, \mathbf{x}) &= 0 && \text{in } \Omega, \\ \dot{\mathbf{u}}(0, \mathbf{x}) &= \mathbf{v}_0(\mathbf{x}) && \text{in } \Omega. \end{aligned}$$

6. PHASE EVOLUTION

The previous considerations show, that, in particular, constitutive equations for the diffusive fluxes are necessary to determine a closed system of model equations. This fact leads to a detailed modelling of the evolution of concentration surfaces, which describe the phase separation process inside the solid mixture. Modelling phase separation is done in various papers, for example in [3, 12, 29].

In this paper we use the concept of microforces to model the disarrangement of atoms due to diffusion, see [18, 20]. Microforces differ completely from classical Newtonian forces, because they are related to the integrity of a body's material structure.

Another difference between microforces and Newtonian forces is due to the fact, that microforces perform work in the evolution of structural defects, in our case in the evolution of concentration surfaces. From [18, 20] we know, that microforces act within the reference configuration.

6.1. Concentration surfaces. The modelling of state surfaces by using microforces is done in [13, 14, 19] and we use these ideas to describe the evolution of concentration surfaces. Figure 4 shows the evolution process of a concentration surface inside the reference configuration.

Definition 6.1. Let $c \in [0, 1]$ arbitrary, but fixed, then the corresponding concentration surfaces of continuum k at time $t \in \mathbb{R}^+$ in the reference configuration is defined by

$$\mathcal{S}_k(t) := \{\mathbf{x} \in \Omega : c_k(t, \mathbf{x}) = c = \text{const}\}.$$

In this paper we assume, that all concentration surfaces are regular. This assumption is necessary, because a local parameterisation of each surface is necessary.

Condition 6.2. For all $t_0 \in \mathbb{R}^+$ and $\mathbf{x}_0 \in \mathcal{S}_k(t_0)$, there exists an index i with $1 \leq i \leq d$, such that

$$\frac{\partial}{\partial x_i} c_k(t, \mathbf{x})|_{(t_0, \mathbf{x}_0)} \neq 0.$$

Conclusion 6.3. From condition 6.2 it follows by the implicit function theorem, that for all $t_0 \in \mathbb{R}^+$ and $\mathbf{x}_0 \in \mathcal{S}_k(t_0)$ there exists an environment $\mathcal{U}_\varepsilon(t_0, \mathbf{x}_0) \subset \mathbb{R}^d$, a set $\mathcal{V} \subset \mathbb{R}^d$ and a smooth local parameterisation

$$\mathbf{X}_k : \mathbb{R}^+ \times \mathcal{V} \longrightarrow \mathcal{U}_\varepsilon(t_0, \mathbf{x}_0) \cap \mathcal{S}_k(t_0),$$

such that

$$c_k(t, \mathbf{X}_k(t, \boldsymbol{\tau})) = c = \text{const.}$$

Note, that this parameterisation \mathbf{X}_k is in general not unique, but in addition for all indices $1 \leq i \leq d$ holds

$$0 = \frac{\partial}{\partial \tau_i} c_k(t, \mathbf{X}_k(t, \boldsymbol{\tau})) = \nabla_{\mathbf{x}} c_k(t, \mathbf{X}_k(t, \boldsymbol{\tau})) \cdot \frac{\partial}{\partial \tau_i} \mathbf{X}_k(t, \boldsymbol{\tau}).$$

This calculation leads to a definition of a normal vector on the concentration surface.

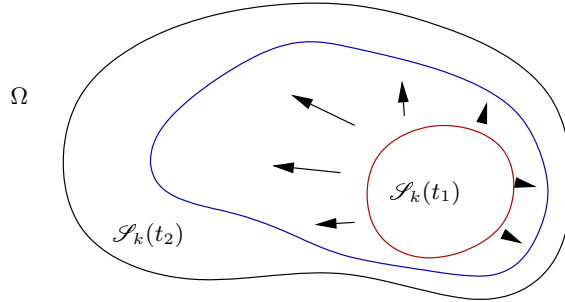


FIGURE 4. evolution of a concentration surface

Definition 6.4. The normal vector on the concentration surface at time $t \in \mathbb{R}^+$ in $\mathbf{x} \in \mathcal{S}_k(t)$ is defined by a smooth vector field

$$\mathbf{n}_k^c : \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d,$$

$$\mathbf{n}_k^c(t, \mathbf{x}) := -\frac{\nabla_{\mathbf{x}} c_k(t, \mathbf{x})}{|\nabla_{\mathbf{x}} c_k(t, \mathbf{x})|}.$$

Note, that \mathbf{n}_k^c is well defined, because in condition 6.2 a regular concentration surface is assumed and therefore we have $|\nabla_{\mathbf{x}} c_k| > 0$. It is obvious, that in general \mathbf{n}_k^c differs from the normal vector of the body \mathbf{n} . Further, the normal vector looks into the other way to the steepest increase of concentration. Figure 5 shows the normal and tangential vector of a concentration surface.

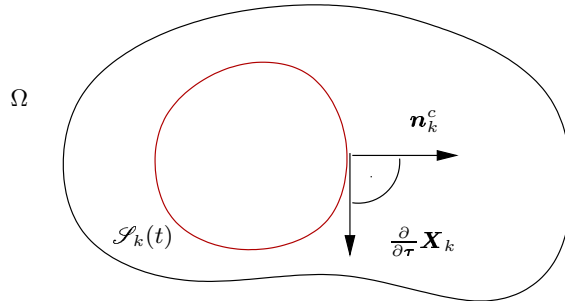


FIGURE 5. normal and tangential vector

Definition 6.5. The normal space on the concentration profile of continuum k at time $t \in \mathbb{R}^+$ in $\mathbf{x} \in \mathcal{S}_k(t)$ is given by

$$\mathcal{N}_{c_k}(t, \mathbf{x}) := \{\lambda \mathbf{n}_k^c(t, \mathbf{x}), \lambda \in \mathbb{R}\},$$

the corresponding tangential space is defined by

$$\mathcal{T}_{c_k}(t, \mathbf{x}) := \{\boldsymbol{\zeta} \in \mathbb{R}^d : \boldsymbol{\zeta} \cdot \mathbf{n}_k^c(t, \mathbf{x}) = 0\}.$$

From [20] we know, that microforces perform work in the evolution of the concentration surface. Therefore we have to define a velocity of the surface, which is possible in two different ways, see [14].

Definition 6.6. The velocity of the concentration surface $\mathcal{S}_k(t)$ is given by the smooth vector field

$$\begin{aligned} \bar{\mathbf{s}}_k^c &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \bar{\mathbf{s}}_k^c(t, \mathbf{x}) &:= \frac{\partial}{\partial t} \mathbf{X}_k(t, \boldsymbol{\tau}) \quad \text{for } \mathbf{x} = \mathbf{X}_k(t, \boldsymbol{\tau}). \end{aligned}$$

Note, that this definition of the velocity is quite natural, but in general not unique, because it depends on the parameterisation \mathbf{X}_k .

Definition 6.7. The normal velocity of the concentration surface of continuum k is defined by the smooth scalar valued field

$$\begin{aligned} V_k^c &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}, \\ V_k^c(t, \mathbf{x}) &:= \frac{\dot{c}_k(t, \mathbf{x})}{|\nabla_{\mathbf{x}} c_k(t, \mathbf{x})|}. \end{aligned}$$

The normal velocity of the concentration profile is uniquely and well defined, because a regular concentration surface is assumed in condition 6.2.

Conclusion 6.8. The following relation between the normal and the state velocity holds for all $\mathbf{x} \in \mathcal{S}_k(t)$

$$V_k^c(t, \mathbf{x}) = \mathbf{n}_k^c(t, \mathbf{x}) \cdot \bar{\mathbf{s}}_k^c(t, \mathbf{x}).$$

We consider a point $\mathbf{x} = \mathbf{X}_k(t, \boldsymbol{\tau})$ and use definition 6.1, the concentration surface $\mathcal{S}_k(t)$, and conclude

$$\begin{aligned} 0 &= \frac{\partial}{\partial t} c_k(t, \mathbf{X}_k(t, \boldsymbol{\tau})) = \dot{c}_k(t, \mathbf{X}_k(t, \boldsymbol{\tau})) + \nabla_{\mathbf{x}} c_k(t, \mathbf{X}_k(t, \boldsymbol{\tau})) \cdot \frac{\partial}{\partial t} \mathbf{X}_k(t, \boldsymbol{\tau}) \\ &= \dot{c}_k(t, \mathbf{x}) + \nabla_{\mathbf{x}} c_k(t, \mathbf{x}) \cdot \bar{\mathbf{s}}_k^c(t, \mathbf{x}). \end{aligned}$$

Dividing this equation by $|\nabla_{\mathbf{x}} c_k(t, \mathbf{x})| > 0$ this yields

$$\frac{\dot{c}_k(t, \mathbf{x})}{|\nabla_{\mathbf{x}} c_k(t, \mathbf{x})|} = - \frac{\nabla_{\mathbf{x}} c_k(t, \mathbf{x})}{|\nabla_{\mathbf{x}} c_k(t, \mathbf{x})|} \cdot \bar{\mathbf{s}}_k^c(t, \mathbf{x}).$$

This consideration leads to the assertion by using definition 6.4 and definition 6.7.

The above considerations make it necessary to divide our considerations of concentration surfaces into the normal and tangential space. Therefore a projection operator is introduced.

Definition 6.9. The projection into the tangential space of the concentration profile is defined by a linear operator

$$\begin{aligned} \mathbf{P}_{\mathcal{T}_{c_k}} &: \mathbb{R}^d \longrightarrow \mathcal{T}_{c_k}, \\ \mathbf{P}_{\mathcal{T}_{c_k}} \boldsymbol{\zeta} &:= (\mathbf{I} - \mathbf{n}_k^c \otimes \mathbf{n}_k^c) \boldsymbol{\zeta}. \end{aligned}$$

It is easy to see by straight forward calculations, that $\mathbf{P}_{\mathcal{T}_{c_k}} \boldsymbol{\zeta} = 0$ for any $\boldsymbol{\zeta} \in \mathcal{N}_{c_k}$. Following [14] we introduce an admissible velocity field of the concentration surface.

Definition 6.10. The admissible velocity field of continuum k 's concentration surface $\mathcal{S}_k(t)$ is given by a smooth vector field

$$\begin{aligned} \mathbf{s}_k^c &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \mathbf{s}_k^c(t, \mathbf{x}) &:= V_k^c(t, \mathbf{x}) \mathbf{n}_k^c(t, \mathbf{x}) + \mathbf{P}_{\mathcal{S}_{c_k}} \boldsymbol{\zeta} \quad \text{for any } \boldsymbol{\zeta} \in \mathbb{R}^d. \end{aligned}$$

This velocity field ensures, that the normal component is uniquely defined in view of definition 6.7 and the tangential component is arbitrary. In view of an admissible velocity it is obvious, that a tangential movement of the concentration surface does not change the material state, figure 6 illustrates this fact.

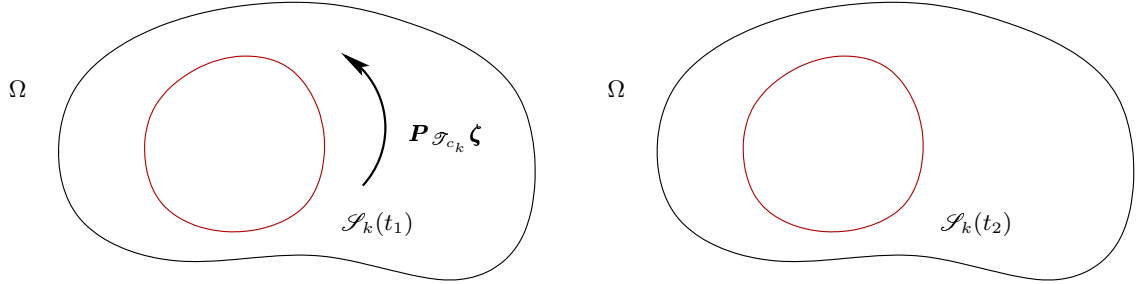


FIGURE 6. tangential movement does not change the material state

6.2. Microforces. In [13, 14] there is a surface traction introduced due to the evolution of a state surface. We pick up this idea and apply it to the evolution of a concentration surfaces.

Condition 6.11. The micro surface force $\mathbf{S}_k^c(\partial\mathcal{P}(t))$ acting on a concentration surface of c_k at time $t \in \mathbb{R}^+$ on the boundary $\partial\mathcal{P}(t)$ for any part $\mathcal{P}(t) \in \mathfrak{B}(\Omega)$ is given by a smooth micro stress tensor

$$\begin{aligned} \mathbf{C}_k^c &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^{d \times d}, \\ \mathbf{S}_k^c(\partial\mathcal{P}(t)) &:= \int_{\partial\mathcal{P}(t)} \mathbf{C}_k^c(t, \mathbf{x}) \mathbf{n}(t, \mathbf{x}) \, d\mathbf{a}_{\mathbf{x}}. \end{aligned}$$

The existence and uniqueness of the micro stress tensor \mathbf{C}_k^c follows from Cauchy's theorem, see [17].

In section 2.3 it is mentioned, that in general a diffusive mass transfer across the boundary takes place. With respect to this fact, we need a micro boundary loading, which describes the evolution of a concentration surface due to the mass transfer across the boundary.

Condition 6.12. The micro boundary loading $\hat{\mathbf{S}}_k^c(\partial\Omega)$ of the body is given by an integrable density function

$$\begin{aligned} \hat{\mathbf{c}}_k^c &: \mathbb{R}^+ \times \partial\Omega \longrightarrow \mathbb{R}^d, \\ \hat{\mathbf{S}}_k^c(\partial\Omega) &:= \int_{\partial\Omega} \hat{\mathbf{c}}_k^c(t, \mathbf{y}) \, d\mathbf{a}_{\mathbf{y}}. \end{aligned}$$

Condition 6.11 as well as condition 6.12 provides a micro surface traction and therefore we get a Neumann boundary condition

$$\mathbf{C}_k^c \mathbf{n} = \hat{\mathbf{c}}_k^c. \quad (6.1)$$

The influence of an external field on the evolution of a concentration surface is described by external microforces. Similar to [19, 20] a representation of the external microforces by a force density is assumed.

Condition 6.13. The external microforce $\mathbf{B}_k^c(\mathcal{P}(t))$ acting on a concentration surface of c_k at time $t \in \mathbb{R}^+$ in $\mathcal{P}(t) \in \mathfrak{B}(\Omega)$ is given by an integrable force density

$$\begin{aligned} \gamma_k^c &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \mathbf{B}_k^c(\mathcal{P}(t)) &:= \int_{\mathcal{P}(t)} \gamma_k^c(t, \mathbf{x}) \, d\mathbf{x}. \end{aligned}$$

The microforces are related to the integrity of material's structure. This fact leads to an internal microforce, which works against the evolution of the concentration profile, see [14, 19].

Condition 6.14. The internal microforce $\mathbf{I}_k^c(\mathcal{P}(t))$ acting on a concentration surface of c_k at time $t \in \mathbb{R}^+$ in $\mathcal{P}(t) \in \mathfrak{B}(\Omega)$ is declared by an integrable force density

$$\begin{aligned} \pi_k^c &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^+, \\ \mathbf{I}_k^c(\mathcal{P}(t)) &:= \int_{\mathcal{P}(t)} \pi_k^c(t, \mathbf{x}) \, d\mathbf{x}. \end{aligned}$$

Microforces describe the internal behaviour of the material and hence we postulate a force balance. In [20] the microforce balance is a consequence of the invariance under changes in material observer.

Postulate 5 (Microforce balance). *For any part $\mathcal{P}(t) \in \mathfrak{B}(\Omega)$ holds*

$$\mathbf{S}_k^c(\partial\mathcal{P}(t)) + \mathbf{B}_k^c(\mathcal{P}(t)) + \mathbf{I}_k^c(\mathcal{P}(t)) = 0.$$

Lemma 6.15. *Let the microforce balance be satisfied, then for continuum k holds almost everywhere in the reference configuration the balance equation*

$$\operatorname{div}_{\mathbf{x}}(\mathbf{C}_k^c) + \gamma_k^c + \pi_k^c = 0.$$

Proof. The balance of microforces and Gauß's theorem, see [23], lead to

$$\begin{aligned} \mathbf{S}_k^c(\partial\mathcal{P}(t)) + \mathbf{B}_k^c(\mathcal{P}(t)) + \mathbf{I}_k^c(\mathcal{P}(t)) &= \int_{\partial\mathcal{P}(t)} \mathbf{C}_k^c \mathbf{n} \, d\mathbf{a}_{\mathbf{x}} + \int_{\mathcal{P}(t)} \gamma_k^c \, d\mathbf{x} + \int_{\mathcal{P}(t)} \pi_k^c \, d\mathbf{x} \\ &= \int_{\mathcal{P}(t)} \operatorname{div}_{\mathbf{x}}(\mathbf{C}_k^c) + \gamma_k^c + \pi_k^c \, d\mathbf{x} = 0. \end{aligned}$$

This calculation holds for any part $\mathcal{P}(t) \in \mathfrak{B}(\Omega)$ and Du Bois-Reymond's lemma, see [10], yields the assertion. \square

The microforces perform work in the evolution of the concentration surface. But we have to remember, that the internal microforces are holding forces and due to this fact, they perform no work, see [20].

$$P_{\text{micro},k}(\mathcal{P}(t)) = \int_{\partial\mathcal{P}(t)} \mathbf{C}_k^c \mathbf{n} \cdot \mathbf{s}_k^c \, d\mathbf{a}_{\mathbf{x}} + \int_{\mathcal{P}(t)} \gamma_k^c \cdot \mathbf{s}_k^c \, d\mathbf{x}. \quad (6.2)$$

The above considerations are very general, thus we have to relate the microforces to the evolution of concentration surfaces. Taking the ideas from [14] into account, we determine the following result.

Lemma 6.16. *For any $t \in \mathbb{R}^+$ and any $\mathbf{x} \in \Omega$, there exists a vector $\mathbf{c}_k^c \in \mathbb{R}^d$ and a scalar $g_k^c \in \mathbb{R}$, respectively a vector $\boldsymbol{\xi}_k^c \in \mathbb{R}^d$ and a scalar $b_k^c \in \mathbb{R}$, such that the micro stress tensor and the external microforce of the concentration surface possess the representation*

$$\mathbf{C}_k^c = \mathbf{c}_k^c \otimes \mathbf{n}_k^c = -\nabla_{\mathbf{x}} c_k \otimes \boldsymbol{\xi}_k^c, \quad (6.3)$$

$$\gamma_k^c = g_k^c \mathbf{n}_k^c = -b_k^c \nabla_{\mathbf{x}} c_k. \quad (6.4)$$

Proof. We consider the working of the microforces given by equation (6.2) and use the admissible velocity of the concentration surface introduced by definition 6.10. It follows

$$\begin{aligned} P_{\text{micro},k}(\mathcal{P}(t)) &= \int_{\partial\mathcal{P}(t)} \mathbf{C}_k^c \mathbf{n} \cdot \mathbf{s}_k^c \, d\mathbf{a}_{\mathbf{x}} + \int_{\mathcal{P}(t)} \gamma_k^c \cdot \mathbf{s}_k^c \, d\mathbf{x} \\ &= \int_{\partial\mathcal{P}(t)} \mathbf{C}_k^c \mathbf{n} \cdot V_k \mathbf{n}_k^c \, d\mathbf{a}_{\mathbf{x}} + \int_{\mathcal{P}(t)} \gamma_k^c \cdot V_k \mathbf{n}_k^c \, d\mathbf{x} \\ &\quad + \int_{\partial\mathcal{P}(t)} \mathbf{C}_k^c \mathbf{n} \cdot (\mathbf{P}_{\mathcal{T}_{c_k}} \boldsymbol{\zeta}) \, d\mathbf{a}_{\mathbf{x}} + \int_{\mathcal{P}(t)} \gamma_k^c \cdot (\mathbf{P}_{\mathcal{T}_{c_k}} \boldsymbol{\zeta}) \, d\mathbf{x}. \end{aligned}$$

In order to have a short notation, we introduce the following definition

$$\mathbf{C}_{\text{tan},k}^c := \mathbf{P}_{\mathcal{T}_{c_k}} \mathbf{C}_k^c \quad \text{and} \quad \gamma_{\text{tan},k}^c := \mathbf{P}_{\mathcal{T}_{c_k}} \gamma_k^c.$$

The work must be independent of the choice of the tangential velocity due to the admissible velocity given by definition 6.10. This leads under consideration of the symmetry of $\mathbf{P}_{\mathcal{T}_{c_k}}$ to

$$\int_{\partial\mathcal{P}(t)} \mathbf{C}_{\text{tan},k}^c \mathbf{n} \cdot \boldsymbol{\zeta} \, d\mathbf{a}_{\mathbf{x}} + \int_{\mathcal{P}(t)} \gamma_{\text{tan},k}^c \cdot \boldsymbol{\zeta} \, d\mathbf{x} = 0.$$

The above relation holds for any smooth vector field $\boldsymbol{\zeta}$ and the fundamental lemma of calculus of variations, see [15], yields

$$\mathbf{C}_{\text{tan},k}^c \mathbf{n} = 0 \quad \text{and} \quad \gamma_{\text{tan},k}^c = 0.$$

Because this work balance holds for any part $\mathcal{P}(t) \in \mathfrak{B}(\Omega)$, we have $\mathbf{C}_{\text{tan},k}^c \mathbf{n} = 0$ for any normal vector \mathbf{n} and therefore it follows additionally

$$\mathbf{C}_{\text{tan},k}^c = \mathbf{0}.$$

The fact, that both tangential amounts disappear, yields the existence of a vector $\mathbf{c}_k^c \in \mathbb{R}^d$ and a scalar $g_k^c \in \mathbb{R}$ such that

$$\begin{aligned} \mathbf{C}_k^c &= \mathbf{n}_k^c \otimes \mathbf{c}_k^c = -\nabla_{\mathbf{x}} c_k \otimes \frac{\mathbf{c}_k^c}{|\nabla_{\mathbf{x}} c_k|} = -\nabla_{\mathbf{x}} c_k \otimes \boldsymbol{\xi}_k^c, \\ \gamma_k^c &= g_k^c \mathbf{n}_k^c = -\frac{g_k^c}{|\nabla_{\mathbf{x}} c_k|} \nabla_{\mathbf{x}} c_k = -b_k^c \nabla_{\mathbf{x}} c_k. \end{aligned}$$

□

Note, that $\mathbf{c}_k^c \cdot \mathbf{n}_k^c$ represents a surface tension, while $\mathbf{c}_k^c \cdot \boldsymbol{\zeta}$ provides a surface shear for $\boldsymbol{\zeta} \in \mathcal{T}_{c_k}$. *Conclusion 6.17.* Using the same arguments as in lemma 6.16, then there exists a scalar $\hat{\xi}_k^c \in \mathbb{R}$, such that the micro surface loading takes the form

$$\hat{\mathbf{c}}_k^c = -\hat{\xi}_k^c \nabla_{\mathbf{x}} c_k. \quad (6.5)$$

Hence we assume a regular concentration surface in condition 6.2, lemma 6.16 and equation (6.5) yields a reduced Neumann boundary condition for the microforce balance

$$\boldsymbol{\xi}_k^c \cdot \mathbf{n} = \hat{\xi}_k^c. \quad (6.6)$$

The representation in lemma 6.16 shows, that the micro surface traction as well as the external microforce acts only in the normal direction of the concentration surface. This considerations provide the idea to decompose the internal microforce into a normal and a tangential component.

Definition 6.18. The normal component of the internal microforce is given by a scalar valued field

$$\begin{aligned} \pi_{\text{norm},k}^c &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}, \\ \pi_{\text{norm},k}^c(t, \mathbf{x}) &:= \boldsymbol{\pi}_k^c \cdot \mathbf{n}_k^c(t, \mathbf{x}). \end{aligned}$$

Definition 6.19. The tangential component of the internal microforce is defined by a vector valued field

$$\begin{aligned}\boldsymbol{\pi}_{\text{tan},k}^c &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \boldsymbol{\pi}_{\text{tan},k}^c(t, \boldsymbol{x}) &:= \boldsymbol{P}_{\mathcal{I}_{c_k}} \boldsymbol{\pi}_k^c(t, \boldsymbol{x}).\end{aligned}$$

It is obvious, that both definitions lead to the following decomposition of the internal microforce,

$$\boldsymbol{\pi}_k^c = \pi_{\text{norm},k}^c \boldsymbol{n}_k^c + \boldsymbol{\pi}_{k,\text{tan}}^c. \quad (6.7)$$

Lemma 6.20. *Let the microforces balance be satisfied, then there holds a normal microforce balance*

$$\text{div}_{\boldsymbol{x}}(\boldsymbol{c}_k^c) + g_k^c + \pi_{\text{norm},k}^c = 0,$$

as well as a tangential microforce balance

$$\boldsymbol{\pi}_{\text{tan},k}^c = -\text{D}_{\boldsymbol{x}}(\boldsymbol{n}_k^c) \boldsymbol{c}_k^c.$$

Proof. At first we consider the normal vector field and use the property $\boldsymbol{n}_k^c \cdot \boldsymbol{n}_k^c = 1$, which is obvious in view of definition 6.4. From this relation directly follows

$$\text{D}_{\boldsymbol{x}}(\boldsymbol{n}_k^c \cdot \boldsymbol{n}_k^c) = 2\text{D}_{\boldsymbol{x}}(\boldsymbol{n}_k^c) \boldsymbol{n}_k^c = 0.$$

This calculation shows, that $\text{D}_{\boldsymbol{x}}(\boldsymbol{n}_k^c)$ maps into the tangential space \mathcal{I}_{c_k} . The force balance from lemma 6.15, the representation formula from lemma 6.16 and the decomposition (6.7) yield

$$\begin{aligned}\text{div}_{\boldsymbol{x}}(\boldsymbol{C}_k^c) + \gamma_k^c + \boldsymbol{\pi}_k^c &= \text{div}_{\boldsymbol{x}}(\boldsymbol{n}_k^c \otimes \boldsymbol{c}_k^c) + g_k^c \boldsymbol{n}_k^c + \pi_{\text{norm},k}^c \boldsymbol{n}_k^c + \boldsymbol{\pi}_{\text{tan},k}^c \\ &= \text{div}_{\boldsymbol{x}}(\boldsymbol{c}_k^c) \boldsymbol{n}_k^c + \text{D}_{\boldsymbol{x}}(\boldsymbol{n}_k^c) \boldsymbol{c}_k^c + g_k^c \boldsymbol{n}_k^c + \pi_{\text{norm},k}^c \boldsymbol{n}_k^c + \boldsymbol{\pi}_{\text{tan},k}^c \\ &= \underbrace{(\text{div}_{\boldsymbol{x}}(\boldsymbol{c}_k^c) + g_k^c + \pi_{\text{norm},k}^c) \boldsymbol{n}_k^c}_{\in \mathcal{N}_{c_k}} + \underbrace{\text{D}_{\boldsymbol{x}}(\boldsymbol{n}_k^c) \boldsymbol{c}_k^c + \boldsymbol{\pi}_{\text{tan},k}^c}_{\in \mathcal{I}_{c_k}} = 0.\end{aligned}$$

Both force balances of the assertion are obvious in view of this calculation. \square

Conclusion 6.21. We consider the tangential microforce balance formulated in lemma 6.20 in conjunction with the normal vector of the concentration surface given by definition 6.4 and calculate

$$\begin{aligned}\boldsymbol{\pi}_{\text{tan},k}^c &= -\text{D}_{\boldsymbol{x}}(\boldsymbol{n}_k^c) \boldsymbol{c}_k^c = \text{D}_{\boldsymbol{x}} \left(\frac{\nabla_{\boldsymbol{x}} c_k}{|\nabla_{\boldsymbol{x}} c_k|} \right) |\nabla_{\boldsymbol{x}} c_k| \boldsymbol{\xi}_k^c \\ &= \frac{1}{|\nabla_{\boldsymbol{x}} c_k|} (\text{D}_{\boldsymbol{x}}(\nabla_{\boldsymbol{x}} c_k) |\nabla_{\boldsymbol{x}} c_k| - \nabla_{\boldsymbol{x}} |\nabla_{\boldsymbol{x}} c_k| \otimes \nabla_{\boldsymbol{x}} c_k) \boldsymbol{\xi}_k^c \\ &= \text{D}_{\boldsymbol{x}}(\nabla_{\boldsymbol{x}} c_k) \boldsymbol{\xi}_k^c - \frac{\nabla_{\boldsymbol{x}} |\nabla_{\boldsymbol{x}} c_k|}{|\nabla_{\boldsymbol{x}} c_k|} \nabla_{\boldsymbol{x}} c_k.\end{aligned} \quad (6.8)$$

On the other hand, we decompose the normal internal microforce, which is only a scalar valued quantity, into

$$\pi_{\text{norm},k}^c = i_k^c |\nabla_{\boldsymbol{x}} c_k| - \nabla_{\boldsymbol{x}} |\nabla_{\boldsymbol{x}} c_k| \cdot \boldsymbol{\xi}_k^c. \quad (6.9)$$

Insert equation (6.8) and (6.9) into the decomposition of the internal microforce (6.7), then it follows directly

$$\boldsymbol{\pi}_k^c := -i_k^c \nabla_{\boldsymbol{x}} c_k + \text{D}_{\boldsymbol{x}}(\nabla_{\boldsymbol{x}} c_k) \boldsymbol{\xi}_k^c. \quad (6.10)$$

From conclusion 6.21 directly follows a normal micro force balance similar to [14, 19].

Lemma 6.22. *The normal microforce balance of the concentration surface holds by the following relation*

$$\text{div}_{\boldsymbol{x}}(\boldsymbol{\xi}_k^c) + b_k^c + i_k^c = 0. \quad (6.11)$$

Proof. From the normal microforce balance formulated in lemma 6.20 follows by using equation (6.9)

$$\begin{aligned} \operatorname{div}_{\mathbf{x}}(\mathbf{c}_k^c) + g_k^c + \pi_{\text{norm},k}^c \\ = \operatorname{div}_{\mathbf{x}}(|\nabla_{\mathbf{x}} c_k| \boldsymbol{\xi}_k^c) + |\nabla_{\mathbf{x}} c_k| b_k^c + i_k^c |\nabla_{\mathbf{x}} c_k| - \nabla_{\mathbf{x}} |\nabla_{\mathbf{x}} c_k| \cdot \boldsymbol{\xi}_k^c \\ = |\nabla_{\mathbf{x}} c_k| (\operatorname{div}_{\mathbf{x}}(\boldsymbol{\xi}_k^c) + b_k^c + i_k^c) = 0. \end{aligned}$$

The regularity of the concentration surface assumed in condition 6.2 shows the assertion. \square

Finally, we have to formulate the boundary value problem of the microforce balance, which consists of the normal force balance (6.11) and the reduced boundary condition (6.6). In this problem the normal micro stress $\boldsymbol{\xi}_k^c$ is the unknown quantity.

$$\begin{aligned} \operatorname{div}_{\mathbf{x}}(\boldsymbol{\xi}_k^c) + b_k^c + i_k^c &= 0 && \text{in } \mathbb{R}^+ \times \Omega, \\ \boldsymbol{\xi}_k^c \cdot \mathbf{n} &= \hat{\zeta}_k^c && \text{on } \mathbb{R}^+ \times \partial\Omega. \end{aligned}$$

Note, that in this boundary value problem time $t \in \mathbb{R}^+$ plays the role of a parameter.

The special representation of the microforces in lemma 6.16 determines a relation for the working of the micro forces, which contains the mass concentration.

Lemma 6.23. *The working of the microforces at time $t \in \mathbb{R}^+$ in any part $\mathcal{P}(t) \in \mathfrak{B}(\Omega)$ is given by*

$$P_{\text{micro},k}(\mathcal{P}(t)) = \int_{\mathcal{P}(t)} \boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k - i_k^c \dot{c}_k \, d\mathbf{x}.$$

Proof. In general the working of the microforces is derived by equation 6.2. Taking into account, that the working is independent of the tangential component of the state velocity, using the representation from lemma 6.16 and the normal force balance (6.11), then we determine

$$\begin{aligned} P_{\text{micro},k}(\mathcal{P}(t)) &= \int_{\partial\mathcal{P}(t)} \mathbf{C}_k^c \mathbf{n} \cdot \mathbf{s}_k^c \, d\mathbf{a}_{\mathbf{x}} + \int_{\mathcal{P}(t)} \boldsymbol{\gamma}_k^c \cdot \mathbf{s}_k^c \, d\mathbf{x} \\ &= \int_{\partial\mathcal{P}(t)} \mathbf{C}_k^c \mathbf{n} \cdot V_k^c \mathbf{n}_k^c \, d\mathbf{a}_{\mathbf{x}} + \int_{\mathcal{P}(t)} \boldsymbol{\gamma}_k^c \cdot V_k^c \mathbf{n}_k^c \, d\mathbf{x} \\ &= \int_{\partial\mathcal{P}(t)} \mathbf{n} \cdot \underbrace{(\mathbf{n}_k^c \cdot \mathbf{n}_k^c)}_{=1} c_k^c V_k^c \, d\mathbf{a}_{\mathbf{x}} + \int_{\mathcal{P}(t)} g_k^c \underbrace{(\mathbf{n}_k^c \cdot \mathbf{n}_k^c)}_{=1} V_k^c \, d\mathbf{x} \\ &= \int_{\mathcal{P}(t)} \operatorname{div}_{\mathbf{x}}(c_k^c V_k^c) + g_k^c V_k^c \, d\mathbf{x} = \int_{\mathcal{P}(t)} \operatorname{div}_{\mathbf{x}}(\boldsymbol{\xi}_k^c \dot{c}_k) + b_k^c \dot{c}_k \, d\mathbf{x} \\ &= \int_{\mathcal{P}(t)} \operatorname{div}_{\mathbf{x}}(\boldsymbol{\xi}_k^c) \dot{c}_k + \boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k + b_k^c \dot{c}_k \, d\mathbf{x} = \int_{\mathcal{P}(t)} \boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k - i_k^c \dot{c}_k \, d\mathbf{x}. \end{aligned}$$

\square

In the next section the analysis of continuum k 's energy as well as mixture's energy in the actual configuration are done. For this reason a transformation of the micro stress tensor \mathbf{C}_k^c , the external microforce $\boldsymbol{\gamma}_k^c$ and the state velocity \mathbf{s}_k^c into the actual configuration is necessary. A transformation of this kind is also done in [20], but only of technical interest.

Definition 6.24. The micro stress tensor of a concentration profile c_k in the actual configuration is given by a smooth tensor field

$$\begin{aligned} \mathbf{C}_k^{c,\varphi} : \mathcal{F} &\longrightarrow \mathbb{R}^{d \times d}, \\ \mathbf{C}_k^{c,\varphi}(t, \mathbf{y}) &:= \det \mathbf{F}^{-1}(t, \mathbf{x}) \mathbf{F}^{-T}(t, \mathbf{x}) \mathbf{C}_k^c(t, \mathbf{x}) \mathbf{F}^T(t, \mathbf{x}) \quad \text{for } \mathbf{y} = \varphi(t, \mathbf{x}). \end{aligned}$$

Definition 6.25. The external microforce of a concentration surface c_k in the actual configuration is given by an integrable force density

$$\begin{aligned} \boldsymbol{\gamma}_k^{c,\varphi} : \mathcal{F} &\longrightarrow \mathbb{R}^d, \\ \boldsymbol{\gamma}_k^{c,\varphi}(t, \mathbf{y}) &:= \det \mathbf{F}^{-1}(t, \mathbf{x}) \mathbf{F}^{-T}(t, \mathbf{x}) \boldsymbol{\gamma}_k^c(t, \mathbf{x}) \quad \text{for } \mathbf{y} = \varphi(t, \mathbf{x}). \end{aligned}$$

Definition 6.26. The velocity of a concentration surface in the actual configuration is given by a smooth vector field

$$\begin{aligned} \mathbf{s}_k^{c,\varphi} &: \mathcal{I} \longrightarrow \mathbb{R}^d, \\ \mathbf{s}_k^{c,\varphi}(t, \mathbf{y}) &:= \mathbf{F}(t, \mathbf{x}) \mathbf{s}_k^c(t, \mathbf{x}) \quad \text{for } \mathbf{y} = \varphi(t, \mathbf{x}). \end{aligned}$$

Finally, this definition satisfy by easy calculations the follow representation of microforce's working

$$\begin{aligned} P_{\text{micro},k}(\mathcal{P}^\varphi(t)) &= \int_{\partial \mathcal{P}^\varphi(t)} \mathbf{C}_k^{c,\varphi} \mathbf{n}^\varphi \cdot \mathbf{s}_k^{c,\varphi} \, d\mathbf{a}_\mathbf{y} + \int_{\mathcal{P}^\varphi(t)} \boldsymbol{\gamma}_k^{c,\varphi} \cdot \mathbf{s}_k^{c,\varphi} \, d\mathbf{y} \\ &= \int_{\partial \mathcal{P}(t)} \mathbf{C}_k^c \mathbf{n} \cdot \mathbf{s}_k^c \, d\mathbf{a}_\mathbf{x} + \int_{\mathcal{P}(t)} \boldsymbol{\gamma}_k^c \cdot \mathbf{s}_k^c \, d\mathbf{x} \\ &= P_{\text{micro},k}(\mathcal{P}(t)). \end{aligned}$$

7. BALANCE OF ENERGY

This section is devoted to the first law of thermodynamics. For a general axiomatic foundation see [16], the classical thermodynamics of mixtures is formulated in [31]. In the theory of phase separation we have to consider a more general concept of energy conservation, because microforces perform an additional work, which has to take into account, see [20].

Again, the considerations start with the formulation of continuum k 's energy balance, then we consider the energy conservation of the mixture in a next step and finally we determine some relations between both considerations. The energy consideration provides a generalisation of those in [31], but similar to [19]. In the end we formulate mixture's energy balance in the reference configuration.

7.1. Energy conservation of continuum k . In material theory we are mainly interested in the internal processes of the body, hence we introduce as usual in continuum thermodynamics the internal energy of continuum k . Like in [16, 31, 32] the internal energy is assumed to be given by a density function.

Condition 7.1. The internal energy $E_{\text{int},k}(\mathcal{P}^\varphi(t))$ of continuum k at time $t \in \mathbb{R}^+$ in a part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by a smooth density function

$$\begin{aligned} e_k^\varphi &: \mathcal{I} \longrightarrow \mathbb{R}^+, \\ E_{\text{int},k}(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \rho_k^\varphi(t, \mathbf{y}) e_k^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

The whole energy in continuum k consists of the internal and kinetic energy, thus we determine for the energy

$$E_k(\mathcal{P}^\varphi(t)) = \int_{\mathcal{P}^\varphi(t)} \rho_k^\varphi e_k^\varphi + \frac{1}{2} \rho_k^\varphi \mathbf{v}_k^{\varphi 2} \, d\mathbf{y}. \quad (7.1)$$

Following the classical principles of continuum thermodynamics there is a heat flux into continuum k , see [16, 31, 32].

Condition 7.2. The heat flux $Q_k(\mathcal{P}^\varphi(t))$ into continuum k at time $t \in \mathbb{R}^+$ across the boundary $\partial \mathcal{P}^\varphi(t)$ for any $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi)$ is given by a smooth vector field

$$\begin{aligned} \mathbf{q}_k^\varphi &: \mathcal{I} \longrightarrow \mathbb{R}^d, \\ Q_k(\mathcal{P}^\varphi(t)) &:= \int_{\partial \mathcal{P}^\varphi(t)} \mathbf{q}_k^\varphi(t, \mathbf{y}) \cdot \mathbf{n}^\varphi(t, \mathbf{y}) \, d\mathbf{a}_\mathbf{y}. \end{aligned}$$

Further, we have to take a heat source in continuum k into account, which describes the development of heat resulting for example from chemical reactions, see [16, 31, 32].

Condition 7.3. The heat source $R_k(\mathcal{P}^\varphi(t))$ of continuum k at time $t \in \mathbb{R}^+$ in $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi)$ is given by an integrable density function

$$\begin{aligned} r_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}, \\ R_k(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \rho_k^\varphi(t, \mathbf{y}) r_k^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

Following the ideas of [1, 2, 31], a supply of energy to continuum k happens due to the production of mass and due to chemical reactions.

Condition 7.4. The internal energy supply $\hat{E}_k(\mathcal{P}^\varphi(t))$ of continuum k at time $t \in \mathbb{R}^+$ in any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by an integrable density function

$$\begin{aligned} \hat{e}_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}, \\ \hat{E}_k(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \hat{e}_k^\varphi(t, \mathbf{y}) + \hat{c}_k^\varphi(t, \mathbf{y}) e_k^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

Condition 7.2, condition 7.3 and condition 7.4 provide a representation of the thermal working on continuum k ,

$$P_{\text{therm},k}(\mathcal{P}^\varphi(t)) = \int_{\partial\mathcal{P}^\varphi(t)} \mathbf{q}_k^\varphi \cdot \mathbf{n}^\varphi \, d\mathbf{a}_\mathbf{y} + \int_{\mathcal{P}^\varphi(t)} \rho_k^\varphi r_k^\varphi + \hat{c}_k^\varphi e_k^\varphi + \hat{e}_k^\varphi \, d\mathbf{y}.$$

Similar to [31] we postulate the principle of energy conservation for continuum k , which is generalised towards the microforces, see [19, 20].

Postulate 6 (First law of thermodynamics of continuum k). *For any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ holds*

$$\frac{d}{dt} E_k(\mathcal{P}^\varphi(t)) = P_{\text{mech},k}(\mathcal{P}^\varphi(t)) + P_{\text{therm},k}(\mathcal{P}^\varphi(t)) + P_{\text{micro},k}(\mathcal{P}^\varphi(t)).$$

Lemma 7.5. *Let the first law of thermodynamics be satisfied, then it holds for continuum k in the actual configuration almost everywhere*

$$\begin{aligned} [\rho_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2})]' + \text{div}_\mathbf{y} (\rho_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2}) \mathbf{v}_k^\varphi) &= \text{div}_\mathbf{y} (\mathbf{S}_k^{\varphi T} \mathbf{v}_k^\varphi - \mathbf{q}_k^\varphi + \mathbf{C}^{c,\varphi T} \mathbf{s}_k^{c,\varphi}) \\ &+ \rho_k^\varphi r_k^\varphi + \rho_k^\varphi \mathbf{b}_k^\varphi \cdot \mathbf{v}_k^\varphi + \dot{\mathbf{i}}_k^\varphi \cdot \mathbf{v}_k^\varphi + \boldsymbol{\gamma}_k^{c,\varphi} \cdot \mathbf{s}_k^{c,\varphi} + \hat{e}_k^\varphi + \hat{c}_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2}). \end{aligned}$$

Proof. This lemma follows directly from the first law of thermodynamics, Gauß's theorem and Reynold's transport theorem 3.1. \square

7.2. Energy conservation of the mixture. Similar as for a single continuum and in agreement with [31], we introduce an internal energy for the whole mixture.

Condition 7.6. The internal energy $E_{\text{int}}(\mathcal{P}^\varphi(t))$ of the mixture at time $t \in \mathbb{R}^+$ in any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ in the actual configuration is given by a smooth density function

$$\begin{aligned} e^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}, \\ E_{\text{int}}(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \rho^\varphi(t, \mathbf{y}) e^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

The whole energy in the mixture consists of the internal and kinematic energy resulting from the mass averaged movement of the mixture.

$$E(\mathcal{P}^\varphi(t)) = \int_{\mathcal{P}^\varphi(t)} \rho^\varphi e^\varphi + \frac{1}{2} \rho^\varphi \mathbf{v}^{\varphi 2} \, d\mathbf{y}. \quad (7.2)$$

Following [31] a heat flux Q for the mixture is necessary. The relation between the heat flux Q of the mixture and the heat flux Q_k of a single continuum will be determine later on.

Condition 7.7. The heat flux $Q(\partial\mathcal{P}^\varphi(t))$ into the mixture at time $t \in \mathbb{R}^+$ across the boundary $\partial\mathcal{P}^\varphi(t)$ for any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by a smooth vector field

$$\begin{aligned} \mathbf{q}^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^d, \\ Q(\mathcal{P}^\varphi(t)) &:= \int_{\partial\mathcal{P}^\varphi(t)} \mathbf{q}^\varphi(t, \mathbf{y}) \cdot \mathbf{n}^\varphi(t, \mathbf{y}) \, d\mathbf{a}_\mathbf{y}. \end{aligned}$$

Hence heat is an additive quantity [16], the definition of mixture's heat supply happens in a quite natural way.

Definition 7.8. The heat supply density of the mixture in the actual configuration is defined by an integrable density function

$$\begin{aligned} r^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}, \\ r^\varphi(t, \mathbf{y}) &:= \sum_{k=1}^K c_k^\varphi(t, \mathbf{y}) r_k^\varphi(t, \mathbf{y}). \end{aligned}$$

Note, that there exist no internal energy supply of the mixture, because it is modelled as a thermodynamical closed system. Condition 7.7 and definition 7.8 give us a representation of the thermal working on the mixture,

$$P_{\text{therm}}(\mathcal{P}^\varphi(t)) = \int_{\partial\mathcal{P}^\varphi(t)} \mathbf{q}^\varphi \cdot \mathbf{n}^\varphi \, d\mathbf{a}_\mathbf{y} + \int_{\mathcal{P}^\varphi(t)} \rho^\varphi r^\varphi \, d\mathbf{y}.$$

For mixture's energy we postulate also a conservation principle as it is done in [31], but generalise it with respect to the micro working, see [20].

Postulate 7 (First law of thermodynamics of the mixture). *For any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ holds*

$$\frac{d}{dt} E(\mathcal{P}^\varphi(t)) = P_{\text{mech}}(\mathcal{P}^\varphi(t)) + P_{\text{therm}}(\mathcal{P}^\varphi(t)) + \sum_{k=1}^K P_{\text{micro},k}(\mathcal{P}^\varphi(t)).$$

Lemma 7.9. *Let the first law of thermodynamics be satisfied, then for the mixture holds in the actual configuration almost everywhere*

$$\begin{aligned} &[\rho^\varphi(e^\varphi + 1/2\mathbf{v}^{\varphi 2})]' + \text{div}_\mathbf{y}(\rho^\varphi(e^\varphi + 1/2\mathbf{v}^{\varphi 2})\mathbf{v}^\varphi) \\ &= \text{div}_\mathbf{y} \left(\mathbf{S}^{\varphi T} \mathbf{v}^\varphi - \mathbf{q}^\varphi + \sum_{k=1}^K \mathbf{C}_k^{c,\varphi T} \mathbf{s}_k^{c,\varphi} \right) + \rho^\varphi r^\varphi + \sum_{k=1}^K [\rho_k^\varphi \mathbf{b}_k^\varphi \cdot \mathbf{v}_k^\varphi + \gamma_k \cdot \mathbf{s}_k^{c,\varphi}]. \end{aligned}$$

Proof. This proof is given by the first law of thermodynamics, Gauß's theorem and Reynold's transport theorem 3.1. \square

Energy is an additive quantity, see [16], whereas this fact leads to a relation between the internal energy of the mixture and the internal energy of continuum k . Further, there is a relation between mixture's heat flux and the heat fluxes of different continua. Both relations are formulated within the actual configuration in [1, 2, 31].

Lemma 7.10. *Let the principle of mass conservation and the first law of thermodynamics be satisfied, then the additivity of energy yields*

$$e^\varphi = \sum_{k=1}^K [c_k^\varphi e_k^\varphi + 1/2 c_k^\varphi \mathbf{w}_k^{\varphi 2}], \quad (7.3)$$

$$\mathbf{q}^\varphi = \sum_{k=1}^K [\mathbf{q}_k^\varphi + \mathbf{J}_k^\varphi e_k^\varphi + 1/2 \mathbf{J}_k^\varphi \mathbf{w}_k^{\varphi 2} - \mathbf{S}_k^{\varphi T} \mathbf{w}_k^\varphi], \quad (7.4)$$

$$\sum_{k=1}^K [\mathbf{i}_k^\varphi \cdot \mathbf{v}_k^\varphi + \hat{e}_k^\varphi + \hat{c}_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2})] = 0, \quad (7.5)$$

$$\sum_{k=1}^K [\mathbf{i}_k^\varphi \cdot \mathbf{w}_k^\varphi + \hat{e}_k^\varphi + \hat{c}_k^\varphi (e_k^\varphi + 1/2 \mathbf{w}_k^{\varphi 2})] = 0. \quad (7.6)$$

Proof. This lemma is a very technical one and thus it is divided into five steps.

Step 1: For the proof of relation (7.3) we make use of the fact, that energy is an additive quantity. We summarise equation (7.1) over all continua, take the mass averaged velocity given by definition 4.10 into account and apply the diffusive flux's property (4.5),

$$\begin{aligned} \sum_{k=1}^K \rho_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2}) &= \sum_{k=1}^K \rho_k^\varphi (e_k^\varphi + 1/2 (\mathbf{w}_k^\varphi + \mathbf{v}^\varphi)^2) \\ &= \sum_{k=1}^K \rho_k^\varphi (e_k^\varphi + 1/2 \mathbf{w}_k^{\varphi 2} + \mathbf{w}_k^\varphi \cdot \mathbf{v}^\varphi + 1/2 \mathbf{v}^{\varphi 2}) \\ &= \sum_{k=1}^K \rho_k^\varphi (e_k^\varphi + 1/2 \mathbf{w}_k^{\varphi 2} + 1/2 \mathbf{v}^{\varphi 2}). \end{aligned} \quad (7.7)$$

A comparison of coefficients with the energy representation of the mixture given by equation (7.2) yields assertion (7.3).

Step 2: The next auxiliary relations deals with different representations of several velocities. We use the definition of the diffusion velocity 4.10 and consider the following term

$$\begin{aligned} &\sum_{k=1}^K [\rho_k^\varphi (\mathbf{w}_k^\varphi \otimes \mathbf{w}_k^\varphi \mathbf{v}^\varphi - 1/2 \mathbf{v}_k^{\varphi 2} \mathbf{w}_k^\varphi)] \\ &= \sum_{k=1}^K [\rho_k^\varphi (\mathbf{w}_k^\varphi \cdot \mathbf{v}^\varphi \mathbf{w}_k^\varphi - 1/2 \mathbf{w}_k^\varphi \cdot \mathbf{v}_k^\varphi \mathbf{w}_k^{\varphi 2} + 1/2 \mathbf{w}_k^\varphi \cdot \mathbf{v}_k^\varphi \mathbf{w}_k^\varphi - 1/2 \mathbf{v}_k^\varphi \cdot \mathbf{v}_k^\varphi \mathbf{w}_k^\varphi)] \\ &= \sum_{k=1}^K [\rho_k^\varphi (\mathbf{w}_k^\varphi \cdot \mathbf{v}^\varphi \mathbf{w}_k^\varphi - 1/2 \mathbf{w}_k^\varphi \cdot \mathbf{v}_k^\varphi \mathbf{w}_k^{\varphi 2} - 1/2 \mathbf{v}^\varphi \cdot \mathbf{v}_k^\varphi \mathbf{w}_k^\varphi)] \\ &= 1/2 \sum_{k=1}^K [\rho_k^\varphi (\mathbf{w}_k^\varphi \cdot \mathbf{v}^\varphi \mathbf{w}_k^\varphi - \mathbf{w}_k^\varphi \cdot \mathbf{v}_k^\varphi \mathbf{w}_k^{\varphi 2} + \mathbf{w}_k^\varphi \cdot \mathbf{v}_k^\varphi \mathbf{w}_k^\varphi - \mathbf{v}^\varphi \cdot \mathbf{v}_k^\varphi \mathbf{w}_k^\varphi)] \\ &= 1/2 \sum_{k=1}^K [\rho_k^\varphi (-\mathbf{w}_k^{\varphi 2} \mathbf{w}_k^\varphi + \mathbf{v}^{\varphi 2} \mathbf{w}_k^\varphi)] = -1/2 \sum_{k=1}^K \rho_k^\varphi \mathbf{w}_k^{\varphi 2} \mathbf{w}_k^\varphi + 1/2 \underbrace{\sum_{k=1}^K \mathbf{J}_k^\varphi \mathbf{v}^{\varphi 2}}_{=0} \\ &= -1/2 \sum_{k=1}^K \rho_k^\varphi \mathbf{w}_k^{\varphi 2} \mathbf{w}_k^\varphi \end{aligned} \quad (7.8)$$

Step 3: In order to proof (7.4) and (7.5) the additivity of energy is used and summarising the balance equation of energy formulated in lemma 7.5 over all continua, then the application of relation (7.3) and the auxiliary relation (7.7) yield

$$\begin{aligned}
& \sum_{k=1}^K [\rho_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2})]' + \operatorname{div}_{\mathbf{y}} (\rho_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2}) \mathbf{v}_k^\varphi) \\
&= [\rho^\varphi (e^\varphi + 1/2 \mathbf{v}^{\varphi 2})]' + \operatorname{div}_{\mathbf{y}} (\rho^\varphi (e^\varphi + 1/2 \mathbf{v}^{\varphi 2}) \mathbf{v}^\varphi) + \sum_{k=1}^K \operatorname{div}_{\mathbf{y}} (\rho_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2}) \mathbf{w}_k^\varphi) \\
&= \sum_{k=1}^K [\operatorname{div}_{\mathbf{y}} (\mathbf{S}_k^{\varphi T} \mathbf{v}_k^\varphi - \mathbf{q}_k^\varphi + \mathbf{C}_k^{c,\varphi T} \mathbf{s}_k^{c,\varphi}) + \rho_k^\varphi r_k^\varphi + \rho_k^\varphi \mathbf{b}_k^\varphi \cdot \mathbf{v}_k^\varphi + \mathbf{i}^\varphi \cdot \mathbf{v}_k^\varphi \\
&\quad + \boldsymbol{\gamma}_k^{c,\varphi} \cdot \mathbf{s}_k^{c,\varphi} + \hat{e}_k^\varphi + \hat{c}_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2})].
\end{aligned}$$

Step 4: This calculation leads to another representation of mixture's energy balance equation, if we take the diffusion velocity given by definition 4.10 and auxiliary equation (7.8) into account,

$$\begin{aligned}
& [\rho^\varphi (e^\varphi + 1/2 \mathbf{v}^{\varphi 2})]' + \operatorname{div}_{\mathbf{y}} (\rho^\varphi (e^\varphi + 1/2 \mathbf{v}^{\varphi 2}) \mathbf{v}^\varphi) \\
&= \sum_{k=1}^K [\operatorname{div}_{\mathbf{y}} (\mathbf{S}_k^{\varphi 2} \mathbf{v}_k^\varphi - \rho_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2}) \mathbf{w}_k^\varphi - \mathbf{q}_k^\varphi + \mathbf{C}_k^{c,\varphi T} \mathbf{s}_k^{c,\varphi}) \\
&\quad + \rho_k^\varphi r_k^\varphi + \rho_k^\varphi \mathbf{b}_k^\varphi \cdot \mathbf{v}_k^\varphi + \mathbf{i}_k^\varphi \cdot \mathbf{v}_k^\varphi + \boldsymbol{\gamma}_k^{c,\varphi} \cdot \mathbf{s}_k^{c,\varphi} + \hat{e}_k^\varphi + \hat{c}_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2})] \\
&= \sum_{k=1}^K [\operatorname{div}_{\mathbf{y}} (\mathbf{S}_k^{\varphi T} \mathbf{v}^\varphi + \mathbf{S}_k^{\varphi T} \mathbf{w}_k^\varphi - \rho_k^\varphi \mathbf{w}_k^\varphi \otimes \mathbf{w}_k^\varphi \mathbf{v}^\varphi + \rho_k^\varphi \mathbf{w}_k^\varphi \otimes \mathbf{w}_k^\varphi \mathbf{v}^\varphi \\
&\quad - \rho_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2}) \mathbf{w}_k^\varphi - \mathbf{q}_k^\varphi + \mathbf{C}_k^{c,\varphi T} \mathbf{s}_k^{c,\varphi}) \\
&\quad + \rho_k^\varphi r_k^\varphi + \rho_k^\varphi \mathbf{b}_k^\varphi \cdot \mathbf{v}_k^\varphi + \mathbf{i}_k^\varphi \cdot \mathbf{v}_k^\varphi + \boldsymbol{\gamma}_k^{c,\varphi} \cdot \mathbf{s}_k^{c,\varphi} + \hat{e}_k^\varphi + \hat{c}_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2})] \\
&= \operatorname{div}_{\mathbf{y}} \left(\mathbf{S}^{\varphi T} \mathbf{v}^\varphi + \sum_{k=1}^K \mathbf{C}_k^{c,\varphi T} \mathbf{s}_k^{c,\varphi} - \sum_{k=1}^K \left(\mathbf{q}_k^\varphi + \mathbf{J}_k^\varphi e_k^\varphi + 1/2 \mathbf{J}_k^\varphi \mathbf{w}_k^{\varphi 2} - \mathbf{S}_k^{\varphi T} \mathbf{w}_k^\varphi \right) \right) \\
&\quad + \rho^\varphi r^\varphi + \sum_{k=1}^K [\rho_k^\varphi \mathbf{b}_k^\varphi \cdot \mathbf{v}_k^\varphi + \boldsymbol{\gamma}_k^{c,\varphi} \cdot \mathbf{s}_k^{c,\varphi}] + \sum_{k=1}^K [\mathbf{i}_k^\varphi \cdot \mathbf{v}_k^\varphi + \hat{e}_k^\varphi + \hat{c}_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2})]
\end{aligned}$$

Using this relation and make a comparison of coefficients with the energy balance equation derived in lemma 7.9, then assertion (7.4) and (7.5) follow.

Step 5: The proof of assertion (7.6) uses relation (7.6), the diffusion velocity given by definition 4.10 and relation (4.4). A straight forward calculation yields

$$\begin{aligned}
& \sum_{k=1}^K [\mathbf{i}_k^\varphi \cdot \mathbf{v}_k^\varphi + \hat{e}_k^\varphi + \hat{c}_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2})] \\
&= \sum_{k=1}^K [\mathbf{i}_k^\varphi \cdot \mathbf{w}_k^\varphi + \mathbf{i}_k^\varphi \cdot \mathbf{v}^\varphi + \hat{e}_k^\varphi + \hat{c}_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^{\varphi 2} - \mathbf{v}_k^\varphi \cdot \mathbf{v}^\varphi + \mathbf{v}_k^\varphi \cdot \mathbf{v}^\varphi)]
\end{aligned}$$

$$\begin{aligned}
&= \sum_{k=1}^K [\mathbf{i}_k^\varphi \cdot \mathbf{w}_k^\varphi + \mathbf{i}_k^\varphi \cdot \mathbf{v}^\varphi + \hat{e}_k^\varphi + \hat{c}_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^\varphi \cdot \mathbf{w}_k^\varphi - 1/2 \mathbf{v}_k^\varphi \cdot \mathbf{v}^\varphi + \mathbf{v}_k^\varphi \cdot \mathbf{v}^\varphi)] \\
&= \sum_{k=1}^K [\mathbf{i}_k^\varphi \cdot \mathbf{w}_k^\varphi + \hat{e}_k^\varphi + \hat{c}_k^\varphi (e_k^\varphi + 1/2 \mathbf{v}_k^\varphi \cdot \mathbf{w}_k^\varphi - 1/2 \mathbf{v}_k^\varphi \cdot \mathbf{v}^\varphi - 1/2 \mathbf{v}_k^\varphi \cdot \mathbf{w}_k^\varphi + \mathbf{v}_k^\varphi \cdot \mathbf{w}_k^\varphi)] \\
&= \sum_{k=1}^K [\mathbf{i}_k^\varphi \cdot \mathbf{w}_k^\varphi + \hat{e}_k^\varphi + \hat{c}_k^\varphi (e_k^\varphi + 1/2 \mathbf{w}_k^{\varphi 2})] + 1/2 \underbrace{\sum_{k=1}^K \hat{c}_k^\varphi \mathbf{v}^{\varphi 2}}_{=0} = 0.
\end{aligned}$$

□

7.3. Energy balance in the reference configuration. This section is devoted to the formulation of mixture's energy balance equation in the reference configuration. The transformation of the internal energy density consist only in a reparameterisation, because the internal energy density is formulated in a mass independent way.

Definition 7.11. The internal energy of continuum k in the reference configuration is given by a smooth scalar valued field

$$\begin{aligned}
e_k &: \mathbb{R}^+ \Omega \longrightarrow \mathbb{R}^+, \\
e_k(t, \mathbf{x}) &:= e_k^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})).
\end{aligned}$$

Definition 7.12. The internal energy of the mixture in the reference configuration is given by a smooth scalar valued field

$$\begin{aligned}
e &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^+, \\
e(t, \mathbf{x}) &:= e^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})).
\end{aligned}$$

From definition 7.11 and definition 7.12 it is easy to see that relation (7.3) holds also in the reference configuration,

$$e = \sum_{k=1}^K [c_k e_k + 1/2 c_k \mathbf{w}_k^2]. \tag{7.9}$$

The transformation of the heat flux into the reference configuration needs Piola's transformation, because it is a surface depending quantity.

Definition 7.13. The heat flux density of continuum k in the reference configuration is defined by a smooth vector field

$$\begin{aligned}
\mathbf{q}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\
\mathbf{q}_k(t, \mathbf{x}) &:= (\mathbf{P} \mathbf{q}_k^\varphi)(t, \mathbf{x}).
\end{aligned}$$

Definition 7.14. The heat flux density of the mixture in the reference configuration is given by a smooth vector field

$$\begin{aligned}
\mathbf{q} &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\
\mathbf{q}(t, \mathbf{x}) &:= (\mathbf{P} \mathbf{q}^\varphi)(t, \mathbf{x}).
\end{aligned}$$

Note, that as a consequence of the linearity of Piola's transformation relation (7.4) holds also in the reference configuration, if we take the diffusive flux in the reference configuration introduced by definition 4.15 and the 1. Piola-Kirchoff stress tensor formulated in definition 5.13 into account

$$\mathbf{q} = \sum_{k=1}^K [\mathbf{q}_k + \mathbf{J}_k e_k + 1/2 \mathbf{J}_k \mathbf{w}_k^2 - \mathbf{S}_k^T \mathbf{w}_k]. \tag{7.10}$$

The heat supply density is also defined in a mass independent way. Therefore the transformation into the reference configuration will be done only by a reparameterisation.

Definition 7.15. The heat supply density of continuum k in the reference configuration is given by an integrable field

$$\begin{aligned} r_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}, \\ r_k(t, \mathbf{x}) &:= r_k^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})). \end{aligned}$$

Definition 7.16. The heat supply density of the mixture in the reference configuration is given by a integrable function

$$\begin{aligned} r &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}, \\ r(t, \mathbf{x}) &:= r^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})). \end{aligned}$$

In view of mixture's heat supply density in the actual configuration given by condition 7.8 in conjunction with definition 7.15 and definition 7.16 holds

$$r = \sum_{k=1}^K c_k r_k.$$

Lemma 7.17. *Let the principle of mass conservation, linear momentum conservation and energy conservation be satisfied, then for the mixture in the reference configuration holds almost everywhere*

$$\rho \dot{e} = \mathbf{S} : \dot{\mathbf{F}} - \operatorname{div}_{\mathbf{x}}(\mathbf{q}) + \rho r + \sum_{k=1}^K [\mathbf{F}^T \mathbf{b}_k \cdot \mathbf{J}_k + \boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k - i_k^c \dot{c}_k]. \quad (7.11)$$

Proof. We consider mixture's energy balance formulated in lemma 7.9, apply the continuity equation (2.5) and relation (7.5), then it follows

$$\begin{aligned} & \int_{\mathcal{P}^\varphi(t)} [\rho^\varphi (e^\varphi + 1/2 \mathbf{v}^{\varphi 2})]' + \operatorname{div}_{\mathbf{y}}(\rho^\varphi (e^\varphi + 1/2 \mathbf{v}^{\varphi 2}) \mathbf{v}^\varphi) d\mathbf{y} \\ &= \int_{\mathcal{P}^\varphi(t)} \rho^{\varphi'} (e^\varphi + 1/2 \mathbf{v}^{\varphi 2}) + \rho^\varphi (e^\varphi + 1/2 \mathbf{v}^{\varphi 2})' \\ & \quad + \operatorname{div}_{\mathbf{y}}(\rho^\varphi \mathbf{v}^\varphi)(e^\varphi + 1/2 \mathbf{v}^{\varphi 2}) + \rho^\varphi \mathbf{D}_{\mathbf{y}}(e^\varphi + 1/2 \mathbf{v}^{\varphi 2}) \mathbf{v}^\varphi d\mathbf{y} \\ &= \int_{\mathcal{P}^\varphi(t)} \rho^\varphi e^{\varphi'} + \rho^\varphi \mathbf{v}^{\varphi'} \cdot \mathbf{v}^\varphi + \rho^\varphi \nabla_{\mathbf{y}} e^\varphi \cdot \mathbf{v}^\varphi + \rho^\varphi \mathbf{D}_{\mathbf{y}}(\mathbf{v}^\varphi) \mathbf{v}^\varphi \cdot \mathbf{v}^\varphi d\mathbf{y} \\ &= \int_{\mathcal{P}^\varphi(t)} \operatorname{div}_{\mathbf{y}} \left(\mathbf{S}^{\varphi T} \mathbf{v}^\varphi - \mathbf{q}^\varphi + \sum_{k=1}^K \mathbf{C}_k^{c, \varphi T} \mathbf{s}_k^{c, \varphi} \right) \\ & \quad + \rho^\varphi r^\varphi + \sum_{k=1}^K [\rho_k^\varphi \mathbf{b}_k^\varphi \cdot \mathbf{v}_k^\varphi + \gamma_k^{c, \varphi} \cdot \mathbf{s}_k^{c, \varphi}] d\mathbf{y}. \end{aligned}$$

This straight forward calculation yields a new representation of mixture's energy balance equation

$$\begin{aligned} & \int_{\mathcal{P}^\varphi(t)} \rho^\varphi e^{\varphi'} + \rho^\varphi \nabla_{\mathbf{y}} e^\varphi \cdot \mathbf{v}^\varphi d\mathbf{y} = \int_{\mathcal{P}^\varphi(t)} -\rho^\varphi (\mathbf{v}^{\varphi'} \cdot \mathbf{v}^\varphi + \mathbf{D}_{\mathbf{y}}(\mathbf{v}^\varphi) \mathbf{v}^\varphi \cdot \mathbf{v}^\varphi) \\ & \quad + \operatorname{div}_{\mathbf{y}} \left(\mathbf{S}^{\varphi T} \mathbf{v}^\varphi - \mathbf{q}^\varphi + \sum_{k=1}^K \mathbf{C}_k^{c, \varphi T} \mathbf{s}_k^{c, \varphi} \right) + \rho^\varphi r^\varphi + \sum_{k=1}^K [\rho_k^\varphi \mathbf{b}_k^\varphi \cdot \mathbf{v}_k^\varphi + \gamma_k^{c, \varphi} \cdot \mathbf{s}_k^{c, \varphi}] d\mathbf{y}. \end{aligned} \quad (7.12)$$

For the first term of (7.12) we get by using the material time derivative (2.1) and the transformation integrals

$$\int_{\mathcal{P}^\varphi(t)} \rho^\varphi (e^{\varphi'} + \nabla_{\mathbf{y}} e^\varphi \cdot \mathbf{v}^\varphi) d\mathbf{y} = \int_{\mathcal{P}} \rho^\varphi (e^{\varphi'} + \nabla_{\mathbf{y}} e^\varphi \cdot \mathbf{v}^\varphi) \det \mathbf{F} d\mathbf{x} = \int_{\mathcal{P}} \rho \dot{e} d\mathbf{x}. \quad (7.13)$$

The consideration of the second term of (7.12) uses transformation of integrals, the properties of Piola's transformation given by lemma 3.6 and mixture's equation of motion (5.13)

$$\begin{aligned}
& \int_{\mathcal{D}^\varphi(t)} -\rho^\varphi(\mathbf{v}^{\varphi'} \cdot \mathbf{v}^\varphi + \mathbf{D}_\mathbf{y}(\mathbf{v}^\varphi)\mathbf{v}^\varphi \cdot \mathbf{v}^\varphi) + \operatorname{div}_\mathbf{y} \left(\mathbf{S}^{\varphi T} \mathbf{v}^\varphi - \mathbf{q}^\varphi + \sum_{k=1}^K \mathbf{C}_k^{c,\varphi T} \mathbf{s}_k^{c,\varphi} \right) \\
& \quad + \rho^\varphi r^\varphi + \sum_{k=1}^K [\rho^\varphi \mathbf{b}_k^\varphi \cdot \mathbf{v}^\varphi + \gamma_k^{c,\varphi} \cdot \mathbf{s}_k^{c,\varphi}] d\mathbf{y} \\
& = \int_{\mathcal{D}^\varphi(t)} -\rho^\varphi(\mathbf{v}^{\varphi'} + \mathbf{D}_\mathbf{y}(\mathbf{v}^\varphi)\mathbf{v}^\varphi) \cdot \mathbf{v}^\varphi + \operatorname{div}_\mathbf{y}(\mathbf{S}^\varphi) \cdot \mathbf{v}^\varphi + \mathbf{S}^\varphi : \dot{\mathbf{F}}' - \operatorname{div}_\mathbf{y}(\mathbf{q}^\varphi) \\
& \quad + \sum_{k=1}^K \operatorname{div}_\mathbf{y}(\mathbf{C}_k^{c,\varphi T} \mathbf{s}_k^{c,\varphi}) + \rho^\varphi r^\varphi \sum_{k=1}^K [\mathbf{b}_k^\varphi \cdot \mathbf{J}_k^\varphi + \rho_k^\varphi \mathbf{b}_k^\varphi \cdot \mathbf{v}^\varphi + \gamma_k^{c,\varphi} \cdot \mathbf{s}_k^{c,\varphi}] d\mathbf{y} \\
& = \int_{\mathcal{D}} \underbrace{(-\rho \dot{\mathbf{v}} + \operatorname{div}_\mathbf{x}(\mathbf{S}) + \sum_{k=1}^K \rho_k \mathbf{b}_k)}_{=0} \cdot \mathbf{v} + \mathbf{S} : \dot{\mathbf{F}} - \operatorname{div}_\mathbf{x}(\mathbf{q}) + \sum_{k=1}^K \operatorname{div}_\mathbf{x}(\mathbf{C}_k^{cT} \mathbf{s}_k^c) \\
& \quad + \rho r + \sum_{k=1}^K [\mathbf{F}^T \mathbf{b}_k \cdot \mathbf{J}_k + \gamma_k^c \cdot \mathbf{s}_k^c] d\mathbf{x}. \tag{7.14}
\end{aligned}$$

The summary of equation (7.12) and equation (7.14) under consideration of lemma 6.23 yields

$$\int_{\mathcal{D}} \rho \dot{\epsilon} d\mathbf{x} = \int_{\mathcal{D}} \mathbf{S} : \dot{\mathbf{F}} - \operatorname{div}_\mathbf{x}(\mathbf{q}) + \sum_{k=1}^K [\mathbf{F}^T \mathbf{b}_k \cdot \mathbf{J}_k + \boldsymbol{\xi}_k^c \cdot \nabla_\mathbf{x} \dot{c}_k - i_k^c \dot{c}_k].$$

This relation holds for all parts $\mathcal{D} \in \mathfrak{B}(\Omega)$ and Du Bois-Reymond's lemma, see [10], yields the assertion. \square

Finally, we consider mixture's energy balance equation by using the 2. Piola-Kirchoff stress tensor from definition 5.18. In addition one often discuss the case of small deformation and small strains, therefore the energy balance takes the form

$$\rho \dot{\epsilon} = \boldsymbol{\sigma} : \dot{\boldsymbol{\epsilon}} - \operatorname{div}_\mathbf{x}(\mathbf{q}) + \rho r + \sum_{k=1}^K [\mathbf{b}_k \cdot \mathbf{J}_k + \boldsymbol{\xi}_k^c \cdot \nabla_\mathbf{x} \dot{c}_k - i_k^c \dot{c}_k]. \tag{7.15}$$

8. ENTROPY INEQUALITY

The consideration of entropy in mixture theory is quite different from entropy considerations for single continuum materials. The thermodynamics of mixtures are formulated in [31]. Following this ideas, we assume, that entropy is a conservation quantity and thus we determine a balance equation of entropy for each single continuum as well as for the mixture.

It is well known from [31] that the entropy inequality only holds for the whole mixture. We get this inequality by formulating the second law of thermodynamics with a non negative entropy supply.

This sections ends with the formulation of the dissipation inequality of the mixture inside the reference configuration.

8.1. Entropy balance of continuum k . Entropy is a quantity, which describes the irreversibility of a thermodynamic process. The existence of an entropy for each single continuum k is assumed and formulated by a density function, see [31].

Condition 8.1. The entropy $S_k(\mathcal{P}^\varphi(t))$ of continuum k at time $t \in \mathbb{R}^+$ in a part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by a specific smooth density function

$$\begin{aligned} \eta_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^+, \\ S_k(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \rho_k^\varphi(t, \mathbf{y}) \eta_k^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

An axiomatic foundation of this assumption is given by [16]. Further we need the existence of absolute temperature. In [16] the existence of an absolute temperature is proofed only by using thermodynamic quantities. In this paper the existence of only one temperature for all continua is assumed, but there are also papers, which work with different temperatures for different continua.

Condition 8.2. There exists an absolute temperature Θ^φ for all continua $1 \leq k \leq K$ in the actual configuration

$$\begin{aligned} \Theta^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^+, \\ \Theta^\varphi &= \Theta^\varphi(t, \mathbf{y}). \end{aligned}$$

Similar to [16, 31, 32] the existence of a conductive entropy flux with an appropriate representation is assumed. An axiomatic foundation of this representation is described in [16].

Condition 8.3. The conductive entropy flux $J_k(\partial \mathcal{P}^\varphi(t))$ of continuum k at time $t \in \mathbb{R}^+$ across the boundary $\partial \mathcal{P}^\varphi(t)$ for any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by the following representation

$$J_k(\partial \mathcal{P}^\varphi(t)) := - \int_{\partial \mathcal{P}^\varphi(t)} \frac{\mathbf{q}_k^\varphi \cdot \mathbf{n}^\varphi}{\Theta^\varphi} \, d\mathbf{a}_\mathbf{y}.$$

Further, we need a radiative entropy flux as usual in continuum thermodynamics.

Condition 8.4. The radiative entropy flux $K_k(\mathcal{P}^\varphi(t))$ of continuum k at time $t \in \mathbb{R}^+$ in any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by the following representation

$$K_k(\mathcal{P}^\varphi(t)) := \int_{\mathcal{P}^\varphi(t)} \frac{\rho_k^\varphi r_k^\varphi}{\Theta^\varphi} \, d\mathbf{y}.$$

In contrast to the classical thermodynamic theory for single materials we need in thermodynamics of mixtures a entropy growth. This concept is developed by [31] and used in this paper.

Condition 8.5. The entropy supply $\hat{S}_k(\mathcal{P}^\varphi(t))$ of continuum k at time $t \in \mathbb{R}^+$ in a part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by an integrable density function

$$\begin{aligned} \hat{\eta}_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}, \\ \hat{S}_k(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \hat{c}_k^\varphi(t, \mathbf{y}) \eta_k^\varphi(t, \mathbf{y}) + \hat{\eta}_k^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \end{aligned}$$

Following [31] we postulate a principle of entropy conservation for continuum k .

Postulate 8 (Entropy conservation of continuum k). *For any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ holds*

$$\frac{d}{dt} S_k(\mathcal{P}^\varphi(t)) = J_k(\partial \mathcal{P}^\varphi(t)) + K_k(\mathcal{P}^\varphi(t)) + \hat{S}_k(\mathcal{P}^\varphi(t)).$$

Lemma 8.6. *Let the principle of entropy conservation be satisfied, then for continuum k holds in the actual configuration almost everywhere*

$$(\rho_k^\varphi \eta_k^\varphi)' + \operatorname{div}_\mathbf{y}(\rho_k^\varphi \eta_k^\varphi \mathbf{v}_k^\varphi) = - \operatorname{div}_\mathbf{y} \left(\frac{\mathbf{q}_k^\varphi}{\Theta^\varphi} \right) + \frac{\rho_k^\varphi r_k^\varphi}{\Theta^\varphi} + \hat{c}_k^\varphi \eta_k^\varphi + \hat{\eta}_k^\varphi.$$

Proof. This relation follows from Gauß's theorem and Reynold's transport theorem 3.1. \square

8.2. Entropy balance of the mixture. In this paper it is assumed, that entropy is an additive quantity. This was also done in [31] and an axiomatic foundation of this assumption is given in [16].

Definition 8.7. The specific entropy density of the mixture in the actual configuration is given by a smooth density function

$$\begin{aligned}\eta^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^+, \\ \eta^\varphi(t, \mathbf{y}) &:= \sum_{k=1}^K c_k^\varphi(t, \mathbf{y}) \eta_k^\varphi(t, \mathbf{y}).\end{aligned}$$

Using this definition the entropy of the mixture at time $t \in \mathbb{R}^+$ in any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ can be calculated by

$$S(\mathcal{P}^\varphi(t)) = \int_{\mathcal{P}^\varphi(t)} \rho^\varphi(t, \mathbf{y}) \eta^\varphi(t, \mathbf{y}) \, d\mathbf{y}. \quad (8.1)$$

Following [31] we take the existence of a conductive entropy flux of the mixture into account.

Condition 8.8. The conductive entropy flux $J(\partial\mathcal{P}^\varphi(t))$ of the mixture at time $t \in \mathbb{R}^+$ across the boundary $\partial\mathcal{P}^\varphi(t)$ of any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by a

$$\begin{aligned}\mathbf{h}^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^d, \\ J(\partial\mathcal{P}^\varphi(t)) &:= - \int_{\partial\mathcal{P}^\varphi(t)} \frac{\mathbf{h}^\varphi(t, \mathbf{y}) \cdot \mathbf{n}^\varphi(t, \mathbf{y})}{\Theta^\varphi(t, \mathbf{y})} \, d\mathbf{a}_\mathbf{y}.\end{aligned}$$

The relation between the conductive entropy flux density \mathbf{h}^φ and the heat flux density \mathbf{q}_k^φ of different continua will be calculated later on.

Condition 8.9. The radiative entropy flux $K(\mathcal{P}^\varphi(t))$ of the mixture at time $t \in \mathbb{R}^+$ in any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by the following representation

$$K(\mathcal{P}^\varphi(t)) := \int_{\mathcal{P}^\varphi(t)} \frac{\rho^\varphi(t, \mathbf{y}) r^\varphi(t, \mathbf{y})}{\Theta^\varphi(t, \mathbf{y})} \, d\mathbf{y}.$$

Note, that this representation is shown by an axiomatic foundation in [16]. In order to describe the conservation of entropy, we introduce similar to [31] an entropy growth.

Condition 8.10. The entropy growth $\hat{S}(\mathcal{P}^\varphi(t))$ of the mixture at time $t \in \mathbb{R}^+$ in any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ is given by an integrable density function

$$\begin{aligned}\hat{\eta}^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}, \\ \hat{S}(\mathcal{P}^\varphi(t)) &:= \int_{\mathcal{P}^\varphi(t)} \hat{\eta}^\varphi(t, \mathbf{y}) \, d\mathbf{y}.\end{aligned}$$

Note, as for single continua and similar to [31], that entropy is an conservation quantity.

Postulate 9 (Entropy conservation of the mixture). *For any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ holds*

$$\frac{d}{dt} S(\mathcal{P}^\varphi(t)) = J(\partial\mathcal{P}^\varphi(t)) + K(\mathcal{P}^\varphi(t)) + \hat{S}(\mathcal{P}^\varphi(t)).$$

In this context the second axiom thermodynamics says, that the growth of entropy is a non negative function.

Postulate 10 (Second law of thermodynamic). *For any part $\mathcal{P}^\varphi(t) \in \mathfrak{B}(\Omega^\varphi(t))$ holds*

$$\hat{S}(\mathcal{P}^\varphi(t)) \geq 0 \quad \iff \quad \hat{\eta}^\varphi \geq 0.$$

The global and local formulation of the second law of thermodynamics are equivalent in view of Du Bois-Reymond's lemma, see [10].

Lemma 8.11. *Let the principle of entropy conservation be satisfied, then for the mixture holds in the actual configuration almost everywhere*

$$(\rho^\varphi \eta^\varphi)' + \operatorname{div}_\mathbf{y}(\rho^\varphi \eta^\varphi \mathbf{v}^\varphi) = - \operatorname{div}_\mathbf{y} \left(\frac{\mathbf{h}^\varphi}{\Theta^\varphi} \right) + \frac{\rho^\varphi r^\varphi}{\Theta^\varphi} + \hat{\eta}^\varphi. \quad (8.2)$$

Let additionally the second law of thermodynamics be satisfied, then for the mixture holds the entropy inequality in the actual configuration almost everywhere

$$(\rho^\varphi \eta^\varphi)' + \operatorname{div}_{\mathbf{y}}(\rho^\varphi \eta^\varphi \mathbf{v}^\varphi) + \operatorname{div}_{\mathbf{y}} \left(\frac{\mathbf{h}^\varphi}{\Theta^\varphi} \right) - \frac{\rho^\varphi r^\varphi}{\Theta^\varphi} \geq 0. \quad (8.3)$$

Proof. This lemma follows from the second law of thermodynamics, Gauß's theorem and Reynold's transport theorem 3.1. \square

Entropy is an additive quantity as well as all conservation quantities. This fact leads to the following relations, which are also formulated in [31] in the actual configuration.

Lemma 8.12. *For the conductive entropy flux of the mixture as well as for the entropy growth of the mixture the following relations hold*

$$\mathbf{h}^\varphi = \sum_{k=1}^K (\mathbf{q}_k^\varphi + \Theta^\varphi \eta_k^\varphi \mathbf{J}_k^\varphi), \quad (8.4)$$

$$\hat{\eta}^\varphi = \sum_{k=1}^K (\hat{c}_k^\varphi \eta_k^\varphi + \hat{\eta}_k^\varphi) \geq 0. \quad (8.5)$$

Proof. We summarise the entropy balance equations over all continua, apply the mass averaged velocity given by definition 2.12 and mixture's entropy introduced by definition 8.7, then we determine

$$\begin{aligned} \sum_{k=1}^K (\rho_k^\varphi \eta_k^\varphi)' + \operatorname{div}_{\mathbf{y}}(\rho_k^\varphi \eta_k^\varphi \mathbf{v}_k^\varphi) &= \sum_{k=1}^K (\rho_k^\varphi \eta_k^\varphi)' + \operatorname{div}_{\mathbf{y}}(\rho_k^\varphi \eta_k^\varphi (\mathbf{v}^\varphi + \mathbf{w}_k^\varphi)) \\ &= (\rho^\varphi \eta^\varphi)' + \operatorname{div}_{\mathbf{y}}(\rho^\varphi \eta^\varphi \mathbf{v}^\varphi) + \sum_{k=1}^K \operatorname{div}_{\mathbf{y}}(\rho_k^\varphi \eta_k^\varphi \mathbf{w}_k^\varphi) \\ &= \sum_{k=1}^K -\operatorname{div}_{\mathbf{y}} \left(\frac{\mathbf{q}_k^\varphi}{\Theta^\varphi} \right) + \frac{\rho_k^\varphi \mathbf{v}_k^\varphi}{\Theta^\varphi} + \hat{c}_k^\varphi \eta_k^\varphi + \hat{\eta}_k^\varphi. \end{aligned}$$

This calculation yields a different representation of mixture's entropy balance equation, described by

$$(\rho^\varphi \eta^\varphi)' + \operatorname{div}_{\mathbf{y}}(\rho^\varphi \eta^\varphi \mathbf{v}^\varphi) = \sum_{k=1}^K \left[-\operatorname{div}_{\mathbf{y}} \left(\frac{\mathbf{q}_k^\varphi}{\Theta^\varphi} + \rho_k^\varphi \eta_k^\varphi \mathbf{w}_k^\varphi \right) \right] + \frac{\rho^\varphi r^\varphi}{\Theta^\varphi} + \sum_{k=1}^K [\hat{c}_k^\varphi \eta_k^\varphi + \hat{\eta}_k^\varphi].$$

A comparison of coefficient with equation (8.2) and the second law of thermodynamics yield the assertion. \square

So far we consider the second law of thermodynamics only in the actual configuration. Now we skip our considerations into the reference configuration. The transformation of entropy into the reference configurations works in form of a reparameterisation, because the entropy density is formulated in a volume independent way.

Definition 8.13. The specific entropy density of continuum k in the reference configuration is given by the smooth scalar valued field

$$\begin{aligned} \eta_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^+, \\ \eta_k(t, \mathbf{x}) &:= \eta_k^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})). \end{aligned}$$

Definition 8.14. The specific entropy density of the mixture in the reference configuration is given by a smooth function

$$\begin{aligned} \eta &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^+, \\ \eta(t, \mathbf{x}) &:= \eta^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})). \end{aligned}$$

It is easy to see, that in view of mass concentration's definition 4.13 and mixture's entropy definition 8.7 the following relation holds

$$\eta = \sum_{k=1}^K c_k \eta_k.$$

The temperature is still a volume independent quantity and therefore the transformation into the reference configurations is quite natural.

Definition 8.15. The absolute temperature of the thermo dynamical system in the reference configuration is given by a smooth scalar valued field

$$\begin{aligned} \Theta &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^+, \\ \Theta(t, \mathbf{x}) &:= \Theta^\varphi(t, \varphi(t, \mathbf{x})). \end{aligned}$$

The conductive entropy flux of the mixture across any surface $\mathcal{P}^\varphi(t)$ needs Piola's transformation introduced by definition 3.3.

Definition 8.16. The conductive entropy flux of the mixture in the reference configuration is given by a smooth vector field

$$\begin{aligned} \mathbf{h} &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^d, \\ \mathbf{h}(t, \mathbf{x}) &:= (\mathbf{P}\mathbf{h})(t, \mathbf{x}). \end{aligned}$$

The linearity of Piola's transformation in combination with the diffusive flux given by definition 4.15 and the heat flux 7.13 yields

$$\mathbf{h} = \sum_{k=1}^K \mathbf{q}_k + \Theta \eta_k \mathbf{J}_k. \quad (8.6)$$

The entropy growth depends on change of volume of the body.

Definition 8.17. The entropy growth of continuum k in the reference configuration is given by an integrable function

$$\begin{aligned} \hat{\eta}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}, \\ \hat{\eta}_k(t, \mathbf{x}) &:= \hat{\eta}_k^\varphi(t, \varphi(t, \mathbf{x})) \det \mathbf{F}(t, \mathbf{x}). \end{aligned}$$

Similar, the entropy growth of the mixture in the reference configuration is given by an integrable function

$$\begin{aligned} \hat{\eta} &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}, \\ \hat{\eta}(t, \mathbf{x}) &:= \hat{\eta}^\varphi(t, \varphi(t, \mathbf{x})) \det \mathbf{F}(t, \mathbf{x}). \end{aligned}$$

By this definition we see, that the second law of thermodynamics $\hat{\eta} \geq 0$ holds also in the reference configuration, because φ is an orientation preserving motion. Further it is obvious, that equation (8.5) is also valid in the reference configuration,

$$\hat{\eta} = \sum_{k=1}^K (\hat{c}_k \eta_k + \hat{\eta}_k) \geq 0. \quad (8.7)$$

Lemma 8.18. *Let the principle of entropy conservation and the principle of mass conservation be satisfied, then for the mixture holds in the reference configuration almost everywhere*

$$\rho \dot{\eta} = -\operatorname{div}_{\mathbf{x}} \left(\frac{\mathbf{h}}{\Theta} \right) + \frac{\rho r}{\Theta} + \hat{\eta}. \quad (8.8)$$

Let additionally the second law of thermodynamic be satisfied, then the entropy inequality of the mixture holds in the reference configuration

$$\rho \dot{\eta} \geq -\operatorname{div}_{\mathbf{x}} \left(\frac{\mathbf{h}}{\Theta} \right) + \frac{\rho r}{\Theta} \quad (8.9)$$

Proof. First we consider the left hand side of mixture's entropy balance equation (8.2), apply mixture's mass balance equation (2.5) and the material time derivative (2.1), then we get

$$\begin{aligned}
& \int_{\mathcal{D}^\varphi(t)} (\rho^\varphi \eta^\varphi)' + \operatorname{div}_{\mathbf{y}}(\rho^\varphi \eta^\varphi \mathbf{v}^\varphi) \, d\mathbf{y} \\
&= \int_{\mathcal{D}^\varphi(t)} \rho^{\varphi'} \eta^\varphi + \rho^\varphi \eta^{\varphi'} + \operatorname{div}_{\mathbf{y}}(\rho^\varphi \mathbf{v}^\varphi) \eta^\varphi + \rho^\varphi \nabla_{\mathbf{y}} \eta^\varphi \cdot \mathbf{v}^\varphi \, d\mathbf{y} \\
&= \int_{\mathcal{D}^\varphi(t)} \rho^\varphi \eta^{\varphi'} + \rho^\varphi \nabla_{\mathbf{y}} \eta^\varphi \cdot \mathbf{v}^\varphi \, d\mathbf{y} \\
&= \int_{\mathcal{D}} \rho^\varphi (\eta^{\varphi'} + \nabla_{\mathbf{y}} \eta^\varphi \cdot \mathbf{v}^\varphi) \det \mathbf{F} \, d\mathbf{x} = \int_{\mathcal{D}} \rho \dot{\eta} \, d\mathbf{x}.
\end{aligned}$$

The right hand side of mixture's entropy balance equation (8.2) leads under consideration of Piola's transformation and lemma 3.4 to

$$\int_{\mathcal{D}^\varphi(t)} -\operatorname{div}_{\mathbf{y}} \left(\frac{\mathbf{h}^\varphi}{\Theta^\varphi} \right) + \frac{\rho^\varphi r^\varphi}{\Theta^\varphi} + \hat{\eta}^\varphi \, d\mathbf{y} = \int_{\mathcal{D}} -\operatorname{div}_{\mathbf{x}} \left(\frac{\mathbf{h}}{\Theta} \right) + \frac{\rho r}{\Theta} + \hat{\eta} \, d\mathbf{x}.$$

The summary of both calculations yields the relation

$$\int_{\mathcal{D}} \rho \dot{\eta} \, d\mathbf{x} = \int_{\mathcal{D}} -\operatorname{div}_{\mathbf{x}} \left(\frac{\mathbf{h}}{\Theta} \right) + \frac{\rho r}{\Theta} + \hat{\eta} \, d\mathbf{x},$$

which holds for all parts $\mathcal{D} \in \mathfrak{B}(\Omega)$ and Du Bois-Reymond's lemma, see [10], as well as the second law of thermodynamics lead to the assertion. \square

8.3. Dissipation inequality. In this section we study the free energy of the body. The analysis of the dissipation of energy within the mixture is important to determine restrictions on the constitutive equations of the material model.

As in continuum thermodynamics usual, all quantities are defined in the actual configuration and then skip the considerations into the reference configuration.

Definition 8.19. The specific Helmholtz free energy of continuum k in the actual configuration is defined by a smooth density function

$$\begin{aligned}
\psi_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}, \\
\psi_k^\varphi(t, \mathbf{y}) &:= e_k^\varphi(t, \mathbf{y}) - \Theta^\varphi(t, \mathbf{y}) \eta_k^\varphi(t, \mathbf{y}).
\end{aligned}$$

Because this definition of Helmholtz free energy is formulated in a mass independent way, the transformation into the reference configuration is only given by a reparameterisation.

Definition 8.20. The specific Helmholtz free energy of continuum k in the reference configuration is given by a smooth density function

$$\begin{aligned}
\psi_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}, \\
\psi_k(t, \mathbf{x}) &:= \psi_k^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})).
\end{aligned}$$

Note, that due to this reparameterisation in the reference configuration still holds

$$\psi_k = e_k - \Theta \eta_k.$$

Definition 8.21. The specific Helmholtz free energy of the mixture in the actual configuration is given by a smooth density function

$$\begin{aligned}
\psi^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}, \\
\psi^\varphi(t, \mathbf{y}) &:= e^\varphi(t, \mathbf{y}) - \Theta^\varphi(t, \mathbf{y}) \eta^\varphi(t, \mathbf{y}).
\end{aligned}$$

Similar to a single continuum, the specific Helmholtz free energy is formulated in a mass independent way and the transformation into the reference configuration is done only by a reparameterisation.

Definition 8.22. The specific Helmholtz free energy of the mixture in the reference configuration is defined by a smooth density function

$$\begin{aligned}\psi &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}, \\ \psi(t, \mathbf{x}) &:= \psi^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})).\end{aligned}$$

It is obvious in view of definition 8.22, that in the reference configuration still holds

$$\psi = e - \Theta\eta.$$

The Helmholtz free energy of the mixture consists of the Helmholtz free energies of different continua and of a kinematic energy due to the diffusion process. This fact is also mentioned by [31] in the actual configuration.

Conclusion 8.23. Between Helmholtz free energy of continuum k and Helmholtz free energy of the mixture there exist following relation

$$\psi^\varphi = \sum_{k=1}^K c_k^\varphi (\psi_k^\varphi + 1/2 \mathbf{w}_k^{\varphi 2}), \quad (8.10)$$

$$\psi = \sum_{k=1}^K c_k (\psi_k + 1/2 \mathbf{w}_k^2). \quad (8.11)$$

Both relations follow from equation (7.3), mixture's entropy definition 8.7 as well as Helmholtz free energy of continuum k given by definition 8.19.

The analysis of a diffusion processes needs a chemical potential, which describes an entropy flux due to the diffusion velocity. In general the chemical potential is a tensor valued quantity and we use the definition given by [31].

Definition 8.24. The tensor of chemical potential of continuum k in the actual configuration is defined by a smooth tensor field

$$\begin{aligned}\mathbf{K}_k^\varphi &: \mathcal{T} \longrightarrow \mathbb{R}^{d \times d}, \\ \mathbf{K}_k^\varphi(t, \mathbf{y}) &:= \left(\psi_k^\varphi(t, \mathbf{y}) + \frac{1}{2} \mathbf{w}_k^{\varphi 2}(t, \mathbf{y}) \right) \mathbf{I} - \frac{1}{\rho_k^\varphi(t, \mathbf{y})} \mathbf{S}_k^{\varphi T}(t, \mathbf{y}).\end{aligned}$$

The transformation of the tensor of chemical potential into the reference configuration is defined by a simple matrix transformation with respect to the deformation gradient.

Definition 8.25. The tensor of chemical potential of continuum k in the reference configuration is defined by

$$\begin{aligned}\mathbf{K}_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}^{d \times d}, \\ \mathbf{K}_k(t, \mathbf{x}) &:= \mathbf{F}^{-1}(t, \mathbf{x}) \mathbf{K}_k^\varphi(t, \boldsymbol{\varphi}(t, \mathbf{x})) \mathbf{F}(t, \mathbf{x}).\end{aligned}$$

From definition 8.25 we can follow a representation of the chemical potential, which contains the 1. Piola-Kirchhoff stress tensor.

Lemma 8.26. *In the reference configuration the tensor of chemical potential take the following representation*

$$\mathbf{K}_k = \left(\psi_k + \frac{1}{2} \mathbf{w}_k^2 \right) \mathbf{I} - \frac{1}{\rho_k} \mathbf{S}_k^T \mathbf{F}.$$

Proof. From definition 8.24 and definition 8.25 under consideration of the 1. Piola-Kirchoff stress tensor given by definition 5.13 and continuum k 's bulk density one gets

$$\begin{aligned} \mathbf{K}_k &= \mathbf{F}^{-1} \mathbf{K}_k^\varphi \mathbf{F} \\ &= \mathbf{F}^{-1} (\psi_k + 1/2 \mathbf{w}_k^{\varphi 2}) \mathbf{F} - \mathbf{F}^{-1} \frac{\det \mathbf{F}}{\rho_k^\varphi \det \mathbf{F}} \mathbf{S}_k^{\varphi T} \mathbf{F} \\ &= (\psi_k + 1/2 \mathbf{w}_k^2) \mathbf{I} - \frac{1}{\rho_k} \det \mathbf{F} (\mathbf{S}_k^\varphi \mathbf{F}^{-T})^T \mathbf{F} \\ &= (\psi_k + 1/2 \mathbf{w}_k^2) \mathbf{I} - \frac{1}{\rho_k} \mathbf{S}_k^T \mathbf{F}. \end{aligned}$$

□

In order to introduce the diffusion process into the entropy considerations, we have to formulate the influence of the chemical potential onto the conductive entropy flux. In the actual configuration, this is also done in [31].

Lemma 8.27. *For the conductive entropy flux of the mixture in the actual configuration as well as in the reference configuration holds*

$$\mathbf{h}^\varphi = \mathbf{q}^\varphi - \sum_{k=1}^K \mathbf{K}_k^\varphi \mathbf{J}_k^\varphi, \quad (8.12)$$

$$\mathbf{h} = \mathbf{q} - \sum_{k=1}^K \mathbf{K}_k \mathbf{J}_k. \quad (8.13)$$

Proof. From relation (8.4) for mixture's entropy flux it follows by applying the representation of mixture's heat flux (7.4) and using the tensor of chemical potential,

$$\begin{aligned} \mathbf{h}^\varphi &= \sum_{k=1}^K [\mathbf{q}_k^\varphi + \rho_k^\varphi \Theta^\varphi \eta_k^\varphi \mathbf{w}_k^\varphi] \\ &= \sum_{k=1}^K [\mathbf{q}_k^\varphi + \rho_k^\varphi e_k^\varphi \mathbf{w}_k^\varphi + 1/2 \rho_k^\varphi \mathbf{w}_k^{\varphi 2} \mathbf{w}_k^\varphi - \mathbf{S}_k^{\varphi T} \mathbf{w}_k^\varphi \\ &\quad - \rho_k^\varphi e_k^\varphi \mathbf{w}_k^\varphi - 1/2 \rho_k^\varphi \mathbf{w}_k^{\varphi 2} \mathbf{w}_k^\varphi + \mathbf{S}_k^{\varphi T} \mathbf{w}_k^\varphi + \rho_k^\varphi \Theta^\varphi \eta_k^\varphi \mathbf{w}_k^\varphi] \\ &= \mathbf{q}^\varphi - \sum_{k=1}^K [(\rho_k^\varphi \psi_k^\varphi + 1/2 \rho_k^\varphi \mathbf{w}_k^{\varphi 2}) \mathbf{I} - \mathbf{S}_k^{\varphi T}] \mathbf{w}_k^\varphi = \mathbf{q}^\varphi - \sum_{k=1}^K \mathbf{K}_k^\varphi \mathbf{J}_k^\varphi. \end{aligned}$$

This calculation shows assertion (8.12). The second formula of this lemma follows directly from (8.12) and the linearity of Piola's transformation. □

Finally, we formulate the dissipation inequality of the mixture within the reference configuration.

Lemma 8.28 (Dissipation inequality). *Let the principle of mass conservation, the principle of energy conservation and the second law of thermodynamic be satisfied, then the dissipation inequality of mixture holds in the reference configuration*

$$\begin{aligned} \rho(\dot{\psi} + \dot{\Theta}\eta) + \frac{1}{\Theta} \nabla_{\mathbf{x}} \Theta \cdot \mathbf{h} - \mathbf{S} : \dot{\mathbf{F}} \\ - \sum_{k=1}^K [\operatorname{div}_{\mathbf{x}} (\mathbf{K}_k \mathbf{J}_k) + \mathbf{F}^T \mathbf{b}_k \cdot \mathbf{J}_k + \boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k - i_k^c \dot{c}_k] \leq 0. \end{aligned}$$

Proof. From the energy balance of the mixture (7.11) we can follow an expression of mixture's heat supply

$$-\rho r = -\rho \dot{e} + \mathbf{S} : \dot{\mathbf{F}} - \operatorname{div}_{\mathbf{x}}(\mathbf{q}) + \sum_{k=1}^K [\mathbf{F}^T \mathbf{b}_k \cdot \mathbf{J}_k + \boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k - i_k^c \dot{c}_k] \quad (8.14)$$

We insert equation (8.14) into mixture's entropy inequality (8.9) and derive by straight forward calculations

$$\begin{aligned} \Theta \rho \dot{\eta} + \Theta \operatorname{div}_{\mathbf{x}} \left(\frac{\mathbf{h}}{\Theta} \right) - \rho r &= \Theta \rho \dot{\eta} + \operatorname{div}_{\mathbf{x}}(\mathbf{h}) - \frac{1}{\Theta} \nabla_{\mathbf{x}} \Theta \cdot \mathbf{h} - \rho r \\ &= \Theta \rho \dot{\eta} + \operatorname{div}_{\mathbf{x}}(\mathbf{h}) - \frac{1}{\Theta} \nabla_{\mathbf{x}} \Theta \cdot \mathbf{h} - \rho \dot{e} + \mathbf{S} : \dot{\mathbf{F}} - \operatorname{div}_{\mathbf{x}}(\mathbf{q}) \\ &\quad + \sum_{k=1}^K [\mathbf{F}^T \mathbf{b}_k \cdot \mathbf{J}_k + \boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k - i_k^c \dot{c}_k] \\ &= \rho(\Theta \dot{\eta} - \dot{e}) + \operatorname{div}_{\mathbf{x}}(\mathbf{h} - \mathbf{q}) - \frac{1}{\Theta} \nabla_{\mathbf{x}} \Theta \cdot \mathbf{h} + \mathbf{S} : \dot{\mathbf{F}} \\ &\quad + \sum_{k=1}^K [\mathbf{F}^T \mathbf{b}_k \cdot \mathbf{J}_k + \boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k - i_k^c \dot{c}_k] \geq 0. \end{aligned}$$

This calculation leads to a new representation of mixture's entropy inequality

$$-\rho(\Theta \dot{\eta} - \dot{e}) - \operatorname{div}_{\mathbf{x}}(\mathbf{h} - \mathbf{q}) + \frac{1}{\Theta} \nabla_{\mathbf{x}} \Theta \cdot \mathbf{h} - \mathbf{S} : \dot{\mathbf{F}} - \sum_{k=1}^K [\mathbf{F}^T \mathbf{b}_k \cdot \mathbf{J}_k + \boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k - i_k^c \dot{c}_k] \leq 0. \quad (8.15)$$

Using mixture's Helmholtz free energy given by definition 8.13, then we derive

$$\dot{\psi} = \dot{e} - \dot{\Theta} \eta - \Theta \dot{\eta} \quad \text{respectively,} \quad -(\Theta \dot{\eta} - \dot{e}) = \dot{\psi} + \dot{\Theta} \eta. \quad (8.16)$$

Further, we take lemma 8.27 into account and derive

$$\operatorname{div}_{\mathbf{x}}(\mathbf{h} - \mathbf{q}) = \sum_{k=1}^K \operatorname{div}_{\mathbf{x}}(\mathbf{K}_k \mathbf{J}_k). \quad (8.17)$$

Finally, insert (8.16) and (8.17) into (8.15) then mixture's dissipation inequality follows directly. \square

In the end of this section, we focus our attention on the formulation of the dissipation inequality by using the 2. Piola-Kirchoff stress tensor as well as take care of small strains. This leads to the dissipation inequality

$$\begin{aligned} \rho(\dot{\psi} + \dot{\Theta} \eta) + \frac{1}{\Theta} \nabla_{\mathbf{x}} \Theta \cdot \mathbf{h} - \boldsymbol{\sigma} : \dot{\boldsymbol{\epsilon}} \\ - \sum_{k=1}^K [\operatorname{div}_{\mathbf{x}}(\mathbf{K}_k \mathbf{J}_k) + \mathbf{b}_k \cdot \mathbf{J}_k + \boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k - i_k^c \dot{c}_k] \leq 0. \end{aligned} \quad (8.18)$$

9. MATERIAL MODEL

9.1. Constitutive assumptions. Up to this point, the presented considerations describe in a very general way the mechanics and thermodynamics of phase separation in solid mixtures. Now we focus our analysis on the special case of general solder materials. Therefore constitutive conditions are assumed in order to derive constitutive equations for a determined system of model equations.

The first kind of constitutive assumptions are related to the diffusion process.

Condition 9.1. Mixture's bulk density is assumed to be constant, this means

$$\rho = \text{const} > 0.$$

An assumption of this kind is often postulated in such a context, whereas we choose a mean value of mixture's bulk density. Note, that the structure of the mixture is described by the mass concentrations c_k , which are still not constant.

Condition 9.2. There are no chemical reactions within the mixture, consequently continuum k 's mass supply disappears,

$$\hat{c}_k = 0.$$

Note, that in general a solder joint is part of a composite and there can happen chemical reactions with different substructures, but from an experimental point of view the amount of reaction species are very small and thus we neglected them.

Condition 9.3. There takes no diffusive mass transfer across the boundary place, this means

$$\hat{j}_k = 0.$$

Note, that in connection with condition 9.2 and condition 9.3, lemma 4.18 is valid, which provides an important property for the mathematical analysis. Applying the above conditions on the diffusion equation, the diffusion process in a solder material is described by the initial boundary value problem

$$\rho \dot{c}_k + \operatorname{div}_{\mathbf{x}}(\mathbf{J}_k) = 0 \quad \text{in } \mathbb{R}^+ \times \Omega, \quad (9.1)$$

$$\mathbf{J}_k \cdot \mathbf{n} = 0 \quad \text{on } \mathbb{R}^+ \times \partial\Omega, \quad (9.2)$$

$$c_k(0, \mathbf{x}) = c_{k,0}(\mathbf{x}) \quad \text{in } \Omega. \quad (9.3)$$

The second kind of constitutive assumption is denoted to the deformation behaviour of the solder material.

Condition 9.4. There are no volume forces acting on the body, this induces

$$\mathbf{b}_k = 0 \quad \text{and consequently} \quad \mathbf{b} = 0.$$

This condition provides an admissible assumption, if there are no external fields, which deform the body or accelerate the phase separation process.

Condition 9.5. Only small deformations and small strains of the solid mixture will be considered, this means

$$|\mathbf{D}_{\mathbf{x}} \mathbf{u}| < \varepsilon.$$

Condition 9.5 ensures the usage of the linearised versions of the equations derived in the previous sections. Especially, we use the linearised equation of motion (5.15), but this equations takes no care about the characteristic length scale of a solder joint.

The restriction of the equation of motion on the characteristic length scale will be done by a dimension analysis. Therefore we consider the following characteristic parameters resulting from measurements:

- **characteristic length:** For practical applications a solder joint has a dimension of a cube with side length

$$L_0 = 10^{-3} [m].$$

- **characteristic velocity:** From measurements we know, that the wave speed of the deformation is bounded by a value

$$v_0 = 10 \left[\frac{m}{s} \right]$$

- **characteristic time:** We are mostly interested in the long time behaviour of the solder material, where a characteristic time of at least one year is assumed,

$$t_0 = 10^8 [s].$$

Lemma 9.6. *Let condition 9.4, condition 9.5 and the above characteristic parameter be satisfied, then the mechanics of the solid mixture is described by a static equilibrium equation*

$$-\operatorname{div}_{\mathbf{x}}(\boldsymbol{\sigma}) = 0.$$

Proof. Using the characteristic parameters, we have to transform the equation of motion (5.15) into dimensionless parameters

$$\mathbf{x}^* = \frac{\mathbf{x}}{L_0}, \quad t^* = \frac{t}{t_0}, \quad \mathbf{v}^* = \frac{\mathbf{v}}{v_0}, \quad \boldsymbol{\sigma}^* = \frac{\boldsymbol{\sigma}}{\rho v_0^2}.$$

Further, we have to transform the derivative operators into dimensionless parameters

$$\frac{\partial}{\partial x_i^*} = L_0 \frac{\partial}{\partial x_i}, \quad \frac{\partial}{\partial t^*} = t_0 \frac{\partial}{\partial t}.$$

The application of this transformation yields for the linearised equation of motion (5.15) under consideration of condition 9.4,

$$\rho \dot{\mathbf{v}} - \operatorname{div}_{\mathbf{x}}(\boldsymbol{\sigma}) = \frac{\rho v_0}{t_0} \frac{\partial}{\partial t^*} \mathbf{v}^* - \frac{\rho v_0^2}{L_0} \operatorname{div}_{\mathbf{x}^*}(\boldsymbol{\sigma}^*) = 0.$$

This calculation directly leads to the relation

$$\frac{L_0}{t_0 v_0} \frac{\partial}{\partial t^*} \mathbf{v}^* = \operatorname{div}_{\mathbf{x}^*}(\boldsymbol{\sigma}^*). \quad (9.4)$$

The factor on the left hand side of equation (9.4) is called Strouhol's number,

$$Sr = \frac{L_0}{t_0 v_0}$$

and by using the characteristic parameters, an estimate of this quantity yields

$$Sr \leq 10^{-12}.$$

This estimate shows, that the mechanical wave propagation can be neglected, because the right hand side of (9.4) is of order one. \square

In view of lemma 9.6 we neglect the wave propagation and the mechanical part of the model will be described by the following static equilibrium boundary value problem of the Newtonian forces,

$$-\operatorname{div}_{\mathbf{x}}(\boldsymbol{\sigma}) = 0 \quad \text{in } \mathbb{R}^+ \times \Omega, \quad (9.5)$$

$$\boldsymbol{\sigma} \mathbf{n} = \hat{\mathbf{g}} \quad \text{on } \mathbb{R}^+ \times \Gamma_N, \quad (9.6)$$

$$\mathbf{u} = 0 \quad \text{on } \mathbb{R}^+ \times \Gamma_D. \quad (9.7)$$

The next kind of constitutive assumptions are related to the microforces. Due to the fact, that we assume no flow boundary constraints in condition 9.3, there happens no propagation of concentration surfaces across the boundary.

Condition 9.7. There are no micro boundary loadings acting on the concentration surfaces, this means

$$\hat{\boldsymbol{\xi}}_k^c = 0.$$

Further, we assume no Newtonian volume forces which results from external fields like electrical or magnetic fields. Similar we assume no external microforces, which affect the diffusion process.

Condition 9.8. There acts no external microforce on the concentration surface, which means

$$\mathbf{b}_k^c = 0.$$

From condition 9.7 and condition 9.8 directly follow a static equilibrium boundary value problem of the microforces

$$\operatorname{div}_{\mathbf{x}}(\boldsymbol{\xi}_k^c) + i_k^c = 0 \quad \text{in } \mathbb{R}^+ \times \Omega, \quad (9.8)$$

$$\boldsymbol{\xi}_k^c \cdot \mathbf{n} = 0 \quad \text{on } \mathbb{R}^+ \times \partial\Omega. \quad (9.9)$$

The following kind of constitutive assumption is related to energy considerations. We consider in this paper only an isothermal process, this is due to the fact, that on the one side we assume no chemical reactions and on the other side we assume no external heating.

Condition 9.9. The phase separation process is assumed to be an isothermal process, this means

$$\Theta = \text{const} > 0, \quad \mathbf{q} = \mathbf{0}, \quad r = 0.$$

Applying condition 9.9 on the linearised energy equation (7.15), than we get a restricted linearised energy equation

$$\rho \dot{\varepsilon} = \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}} + \sum_{k=1}^K [\boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k - i_k^c \dot{c}_k]. \quad (9.10)$$

A further general constitutive assumption deals with the diffusion process and energy dissipation. We assume, that the chemical potential is represented by a scalar valued field. This assumption is often used in most thermodynamic literature.

Condition 9.10. The tensor of chemical potential is assumed to be given by scalar value

$$\begin{aligned} \mu_k &: \mathbb{R}^+ \times \Omega \longrightarrow \mathbb{R}, \\ \mathbf{K}_k &= -\mu_k \mathbf{I}. \end{aligned}$$

Applying the above formulated constitutive constrains on the linearised dissipation inequality (8.18) we get a restricted dissipation inequality

$$\rho \dot{\psi} - \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}} - \sum_{k=1}^K [\boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k - (i_k^c - \rho \mu_k) \dot{c}_k - \nabla_{\mathbf{x}} \mu_k \cdot \mathbf{J}_k] \leq 0. \quad (9.11)$$

For a shorter notation we introduce $\mathbf{c} := (c_1, \dots, c_K)$, $\nabla_{\mathbf{x}} \mathbf{c} := (\nabla_{\mathbf{x}} c_1, \dots, \nabla_{\mathbf{x}} c_K)$, $\boldsymbol{\mu} := (\mu_1, \dots, \mu_K)$ and $\nabla_{\mathbf{x}} \boldsymbol{\mu} := (\nabla_{\mathbf{x}} \mu_1, \dots, \nabla_{\mathbf{x}} \mu_K)$. Using this notation we consider a thermodynamic process defined by the following condition.

Condition 9.11. The thermodynamic process of phase separation in a solid mixture is described by the following constitutive approach:

$$\begin{aligned} \psi &= \psi(\boldsymbol{\varepsilon}, \mathbf{c}, \nabla_{\mathbf{x}} \mathbf{c}, \boldsymbol{\mu}, \nabla_{\mathbf{x}} \boldsymbol{\mu}), & \boldsymbol{\sigma} &= \boldsymbol{\sigma}(\boldsymbol{\varepsilon}, \mathbf{c}, \nabla_{\mathbf{x}} \mathbf{c}, \boldsymbol{\mu}, \nabla_{\mathbf{x}} \boldsymbol{\mu}), \\ \boldsymbol{\xi}_k^c &= \boldsymbol{\xi}_k^c(\boldsymbol{\varepsilon}, \mathbf{c}, \nabla_{\mathbf{x}} \mathbf{c}, \boldsymbol{\mu}, \nabla_{\mathbf{x}} \boldsymbol{\mu}), & i_k^c &= i_k^c(\boldsymbol{\varepsilon}, \mathbf{c}, \nabla_{\mathbf{x}} \mathbf{c}, \boldsymbol{\mu}, \nabla_{\mathbf{x}} \boldsymbol{\mu}), \\ \mathbf{J}_k &= \mathbf{J}_k(\boldsymbol{\varepsilon}, \mathbf{c}, \nabla_{\mathbf{x}} \mathbf{c}, \boldsymbol{\mu}, \nabla_{\mathbf{x}} \boldsymbol{\mu}). \end{aligned}$$

Condition 9.11 leads directly to the following formula of the material time derivative of Helmholtz free energy,

$$\dot{\psi} = \frac{\partial \psi}{\partial \boldsymbol{\varepsilon}} : \dot{\boldsymbol{\varepsilon}} + \sum_{k=1}^K \left[\frac{\partial \psi}{\partial c_k} \dot{c}_k + \frac{\partial \psi}{\partial \nabla_{\mathbf{x}} c_k} \cdot \nabla_{\mathbf{x}} \dot{c}_k + \frac{\partial \psi}{\partial \mu_k} \dot{\mu}_k + \frac{\partial \psi}{\partial \nabla_{\mathbf{x}} \mu_k} \cdot \nabla_{\mathbf{x}} \dot{\mu}_k \right]. \quad (9.12)$$

The constitutive conditions formulated in this section enforces constitutive restriction towards the 2. Piola-Kirchoff stress tensor, the microstresses, the internal microforces and the diffusive fluxes. Similar restriction are calculated in [19].

Theorem 9.12. *Let the constitutive assumption formulated in this section be satisfied, than it holds*

$$\begin{aligned} \boldsymbol{\sigma} &= \rho \frac{\partial \psi}{\partial \boldsymbol{\varepsilon}}, & \boldsymbol{\xi}_k^c &= \rho \frac{\partial \psi}{\partial \nabla_{\mathbf{x}} c_k}, \\ i_k^c &= -\rho \frac{\partial \psi}{\partial c_k} + \rho \mu_k, & \sum_{k=1}^K \mathbf{J}_k \cdot \nabla_{\mathbf{x}} \mu_k &\leq 0, \\ \psi &= \psi(\boldsymbol{\varepsilon}, \mathbf{c}, \nabla_{\mathbf{x}} \mathbf{c}). \end{aligned}$$

Proof. This restriction of the constitutive equations follow by inserting (9.12) into the dissipation inequality (9.11) and derive

$$\begin{aligned}
\rho \frac{\partial \psi}{\partial \boldsymbol{\varepsilon}} : \dot{\boldsymbol{\varepsilon}} &+ \sum_{k=1}^K \left[\rho \frac{\partial \psi}{\partial c_k} \dot{c}_k + \rho \frac{\partial \psi}{\partial \nabla_{\mathbf{x}} c_k} \cdot \nabla_{\mathbf{x}} \dot{c}_k + \rho \frac{\partial \psi}{\partial \mu_k} \dot{\mu}_k + \rho \frac{\partial \psi}{\partial \nabla_{\mathbf{x}} \mu_k} \cdot \nabla_{\mathbf{x}} \dot{\mu}_k \right] \\
&- \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}} - \sum_{k=1}^K [\boldsymbol{\xi}_k^c \cdot \nabla_{\mathbf{x}} \dot{c}_k - (i_k^c - \rho \mu_k) \dot{c}_k - \mathbf{J}_k \cdot \nabla_{\mathbf{x}} \dot{\mu}_k] \\
&= \left(\rho \frac{\partial \psi}{\partial \boldsymbol{\varepsilon}} - \boldsymbol{\sigma} \right) : \dot{\boldsymbol{\varepsilon}} + \sum_{k=1}^K \left[\left(\rho \frac{\partial \psi}{\partial c_k} + i_k^c - \rho \mu_k \right) \dot{c}_k + \left(\rho \frac{\partial \psi}{\partial \nabla_{\mathbf{x}} c_k} - \boldsymbol{\xi}_k^c \right) \cdot \nabla_{\mathbf{x}} \dot{c}_k \right. \\
&\quad \left. + \rho \frac{\partial \psi}{\partial \mu_k} \dot{\mu}_k + \rho \frac{\partial \psi}{\partial \nabla_{\mathbf{x}} \mu_k} \cdot \nabla_{\mathbf{x}} \dot{\mu}_k + \mathbf{J}_k \cdot \nabla_{\mathbf{x}} \dot{\mu}_k \right] \leq 0.
\end{aligned}$$

The constitutive assumptions formulated in condition 9.11 are independent of $\dot{\boldsymbol{\varepsilon}}$, $\dot{\mathbf{c}}$, $\nabla_{\mathbf{x}} \dot{\mathbf{c}}$, $\dot{\boldsymbol{\mu}}$, $\nabla_{\mathbf{x}} \dot{\boldsymbol{\mu}}$. By the virtue of this fact, the above calculation holds for arbitrary $\dot{\boldsymbol{\varepsilon}}$, $\dot{\mathbf{c}}$, $\nabla_{\mathbf{x}} \dot{\mathbf{c}}$, $\dot{\boldsymbol{\mu}}$, $\nabla_{\mathbf{x}} \dot{\boldsymbol{\mu}}$ and thus we can follow

$$\begin{aligned}
\rho \frac{\partial \psi}{\partial \boldsymbol{\varepsilon}} - \boldsymbol{\sigma} &= 0 && \text{leads to} && \boldsymbol{\sigma} = \rho \frac{\partial \psi}{\partial \boldsymbol{\varepsilon}}, \\
\rho \frac{\partial \psi}{\partial \nabla_{\mathbf{x}} c_k} - \boldsymbol{\xi}_k^c &= 0 && \text{leads to} && \boldsymbol{\xi}_k^c = \rho \frac{\partial \psi}{\partial \nabla_{\mathbf{x}} c_k}, \\
\rho \frac{\partial \psi}{\partial c_k} + i_k^c - \rho \mu_k &= 0 && \text{leads to} && i_k^c = -\rho \frac{\partial \psi}{\partial c_k} + \rho \mu_k, \\
\rho \frac{\partial \psi}{\partial \mu_k} &= 0 && \text{leads to} && \psi = \psi(\boldsymbol{\varepsilon}, \mathbf{c}, \nabla_{\mathbf{x}} \mathbf{c}), \\
\rho \frac{\partial \psi}{\partial \nabla_{\mathbf{x}} \mu_k} &= 0 \\
\sum_{k=1}^K [\mathbf{J}_k \cdot \nabla_{\mathbf{x}} \dot{\mu}_k] &\leq 0.
\end{aligned}$$

□

From theorem 9.12 we determine a formula of the chemical potential of continuum k .

Lemma 9.13. *The chemical potential of continuum k takes the form*

$$\mu_k = -\operatorname{div}_{\mathbf{x}} \left(\rho \frac{\partial \psi}{\partial \nabla_{\mathbf{x}} c_k} \right) + \frac{\partial \psi}{\partial c_k}.$$

Proof. We use the representation of the microstress tenor $\boldsymbol{\xi}_k^c$ and the internal microforce i_k^c given by theorem 9.12 and determine by the virtue of the microforce balance (9.8)

$$\operatorname{div}_{\mathbf{x}} \left(\rho \frac{\partial \psi}{\partial \nabla_{\mathbf{x}} c_k} \right) - \rho \frac{\partial \psi}{\partial c_k} + \rho \mu_k = 0.$$

In view of condition 9.1 the assertion follows. □

9.2. Phase separation under elastic loading. Pure phase separation in mixtures was studied in [3, 4, 5] and mathematically described by Cahn-Hilliard's equation. In [19] Cahn-Hilliard's equation was derived by using microforces and further a generalisation towards elastic material behaviour was developed.

A different consideration of phase separation with elasticity can be found in [12] and an application to tin lead solder is published in [7, 8, 9].

In the linearised theory we need in this subsection an additional condition concerning the chemical potential.

Condition 9.14. The chemical potentials of all continua are assumed to be small, this means

$$|\nabla_{\mathbf{x}}\mu_k| \leq \varepsilon \quad \text{for all } 1 \leq k \leq K.$$

Condition 9.14 leads to a constitutive relation for the diffusive fluxes inside the reference configuration.

Lemma 9.15. *Let condition 9.14 be satisfied, then there exist positive semidefinite matrices $\mathbf{M}_{k,l} \in \mathbb{R}^{d \times d}$ for $1 \leq k, l \leq K$, such that*

$$\mathbf{J}_k = -\rho \sum_{l=1}^K \mathbf{M}_{k,l} \nabla_{\mathbf{x}}\mu_l. \tag{9.13}$$

Proof. We obtain the dissipation working from theorem 9.12, given by

$$\sum_{k=1}^K \mathbf{J}_k(\varepsilon, \mathbf{c}, \nabla_{\mathbf{x}}\mathbf{c}, \boldsymbol{\mu}, \nabla_{\mathbf{x}}\boldsymbol{\mu}) \cdot \nabla_{\mathbf{x}}\mu_k \leq 0.$$

Let $\lambda > 0$ an arbitrary parameter, then it also holds

$$\sum_{k=1}^K \mathbf{J}_k(\varepsilon, \mathbf{c}, \nabla_{\mathbf{x}}\mathbf{c}, \boldsymbol{\mu}, \lambda \nabla_{\mathbf{x}}\boldsymbol{\mu}) \cdot \nabla_{\mathbf{x}}\mu_k \leq 0$$

We consider the limit $\lambda \rightarrow 0$ and obtain in view of \mathbf{J}_k 's smoothness, that

$$\sum_{k=1}^K \mathbf{J}_k(\varepsilon, \mathbf{c}, \nabla_{\mathbf{x}}\mathbf{c}, \boldsymbol{\mu}, \mathbf{0}) \cdot \nabla_{\mathbf{x}}\mu_k \leq 0.$$

This relation holds for arbitrary $\nabla_{\mathbf{x}}\mu_k$ and hence in equilibrium we have

$$\mathbf{J}_k(\varepsilon, \mathbf{c}, \nabla_{\mathbf{x}}\mathbf{c}, \boldsymbol{\mu}, \mathbf{0}) = \mathbf{0}.$$

Further, we determine a Taylor expansion of \mathbf{J}_k in $\nabla_{\mathbf{x}}\boldsymbol{\mu} = \mathbf{0}$, which leads to

$$\mathbf{J}_k = \sum_{l=1}^K \frac{\partial \mathbf{J}_k}{\partial \nabla_{\mathbf{x}}\mu_l} \Big|_{\nabla_{\mathbf{x}}\boldsymbol{\mu}=\mathbf{0}} \nabla_{\mathbf{x}}\mu_l + \mathcal{O}(\varepsilon).$$

This Taylor expansion give the definition of the mobility tensors of the solder material, which are introduced in [21],

$$\mathbf{M}_{kl} := -\frac{1}{\rho} \frac{\partial \mathbf{J}_k}{\partial \nabla_{\mathbf{x}}\mu_l} \Big|_{\nabla_{\mathbf{x}}\boldsymbol{\mu}=\mathbf{0}}.$$

Inserting this linearised representation of the diffusive fluxes into the dissipation working, it has to hold for any $\nabla_{\mathbf{x}}\mu_k, \nabla_{\mathbf{x}}\mu_l$

$$-\rho \sum_{k=1}^K \sum_{l=1}^K \mathbf{M}_{kl} \nabla_{\mathbf{x}}\mu_l \cdot \nabla_{\mathbf{x}}\mu_k \leq 0.$$

If we choose $\nabla_{\mathbf{x}}\mu_k = \nabla_{\mathbf{x}}\mu_l$ then it follows, that all \mathbf{M}_{kl} must be positive semidefinite. \square

Due to Onsager's reciprocity law [25, 26] the mobilities must be symmetric matrices. Condition 9.14 also effects the constitutive relations of mixture's stress tensor.

Lemma 9.16. *Let condition 9.14 be satisfied, then the 2. Piola-Kirchoff stress tensor takes the form*

$$\boldsymbol{\sigma} = \sum_{k=1}^K \boldsymbol{\sigma}_k.$$

Proof. We consider mixture's 2. Piola-Kirchoff stress tensor given by equation (5.14) and insert the constitutive relations of the diffusive fluxes (9.13). This leads with respect to condition 9.14 to

$$\begin{aligned}\boldsymbol{\sigma} &= \sum_{k=1}^K [\boldsymbol{\sigma}_k - 1/\rho_k \mathbf{J}_k \otimes \mathbf{J}_k] \\ &= \sum_{k=1}^K \left[\boldsymbol{\sigma}_k - \rho^2/\rho_k \left(\sum_{k=1}^K \mathbf{M}_{kl} \nabla_{\mathbf{x}} \mu_l \right) \otimes \left(\sum_{k=1}^K \mathbf{M}_{kl} \nabla_{\mathbf{x}} \mu_l \right) \right] = \sum_{k=1}^K \boldsymbol{\sigma}_k + \mathcal{O}(\varepsilon).\end{aligned}$$

□

A further constitutive relation in this subsections deals with the Helmholtz free energy of the mixture. The free energy is decomposed into the strain energy $W(\boldsymbol{\varepsilon}, \mathbf{c})$, the exchange energy $f(\mathbf{c})$ and the gradient energy $g(\mathbf{c}, \nabla_{\mathbf{x}} \mathbf{c})$ characterising the transition layer.

Condition 9.17. Mixture's Helmholtz free energy is decomposed in the following way

$$\begin{aligned}\psi(\boldsymbol{\varepsilon}, \mathbf{c}, \nabla_{\mathbf{x}} \mathbf{c}) &= W(\boldsymbol{\varepsilon}, \mathbf{c}) + f(\mathbf{c}) + g(\mathbf{c}, \nabla_{\mathbf{x}} \mathbf{c}), \\ W(\boldsymbol{\varepsilon}, \mathbf{c}) &= \frac{1}{2} \boldsymbol{\sigma}(\boldsymbol{\varepsilon}, \mathbf{c}) : \boldsymbol{\varepsilon}, \\ f(\mathbf{c}) &= \frac{1}{2} \prod_{k=1}^K (\nu_k - c_k)^2, \\ g(\mathbf{c}, \nabla_{\mathbf{x}} \mathbf{c}) &= \frac{1}{2} \sum_{k=1}^K \nabla_{\mathbf{x}} c_k : \boldsymbol{\Gamma}_k(\mathbf{c}) \nabla_{\mathbf{x}} c_k.\end{aligned}$$

Note, that for the exchange energy f there is often used a logarithmic energy function, for example see [9, 12].

Finally, all these constitutive relations provide a determined equation system modelling the phase separation process under elastic loading. In this initial boundary value problem the unknown quantities are the mass concentrations c_k of all continua and the mass averaged deformation of the mixture \mathbf{u} .

$$\begin{aligned}\dot{c}_k &= \operatorname{div}_{\mathbf{x}} \left(\sum_{l=1}^K \mathbf{M}_{kl} \nabla_{\mathbf{x}} \mu_l \right) && \text{in } \mathbb{R}^+ \times \Omega, \\ \mu_k &= -\operatorname{div}_{\mathbf{x}} (\boldsymbol{\Gamma}_k \nabla_{\mathbf{x}} c_k) + \frac{\partial g(\mathbf{c}, \nabla_{\mathbf{x}} \mathbf{c})}{\partial c_k} + \frac{\partial f(\mathbf{c})}{\partial c_k} + \frac{\partial W(\boldsymbol{\varepsilon}, \mathbf{c})}{\partial c_k} && \text{in } \mathbb{R}^+ \times \Omega, \\ -\operatorname{div}_{\mathbf{x}} (\boldsymbol{\sigma}) &= 0 && \text{in } \mathbb{R}^+ \times \Omega, \\ \boldsymbol{\sigma} &= \sum_{k=1}^K \mathbf{C}_k(c_k) (\boldsymbol{\varepsilon} - \bar{\boldsymbol{\varepsilon}}(c_k)) && \text{in } \mathbb{R}^+ \times \Omega, \\ \boldsymbol{\varepsilon} &= \frac{1}{2} (\mathbf{D}_{\mathbf{x}}(\mathbf{u}) + \mathbf{D}_{\mathbf{x}}(\mathbf{u})^T) && \text{in } \mathbb{R}^+ \times \Omega,\end{aligned}$$

Additionally, this partial differential equation system is equipped with an initial constrain for the mass concentrations and boundary conditions for the diffusive fluxes, the mass concentration as well as for the 2. Piola-Kirchoff stress and for the deformation of the mixture.

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