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## A modified effective stress principle for chemical active multiphase materials with internal mass exchange

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This paper is dedicated to the memory of our wonderful colleague and friend, Professor Dr.-ing. Tom Schanz, who recently passed away.

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## ABSTRACT

A modified effective stress principle based on the Theory of Porous Media (TPM) subjected to internal mass exchange is presented. The material under consideration can be viewed as three overlapping and interacting continua consisting of a solid, a fluid and a superabsorbent polymer phase (SAP). The latter one has the ability to absorb and bind large amounts of the aqueous fluid phase. To this end, the polymer phase swells to a hydrogel and its mechanical properties differentiate tremendous from both the free pore fluid and the dry polymer. A strict exploitation of the entropy inequality in the sense of Liu–Müller leads to admissible, thermodynamic-consistent forms for the partial stress tensors of the involved phases. Subsequently, an effective stress principle is derived by expressing the total stress tensor of the mixture in terms of the partial stress tensors of the phases. In contrast to well-established effective stress concepts, the proposed modified effective stress principle presented in this contribution shows a transient behavior for materials subjected to internal mass exchange, which is controlled by micro-structural processes. Furthermore, a promising technical application of the model to two-component grouting mortars is discussed.

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### 1. Introduction

Mass exchange in porous media occurs in different physical fields, e.g. geomechanics, biomechanics, biochemistry but also in the technology of building materials.<sup>1</sup> The driving force behind internal mass exchange strongly depends on the specific application under consideration. However, phase transition processes in chemically active porous materials can be incorporated in a multi-phase and multi-species framework based on the Theory of Porous Media (TPM). For instance, Ristinmaa et al.<sup>2</sup> developed a framework for a two-phase model with internal mass exchange, while the driving force for mass exchange has been derived and discussed in a general and rigorous manner. In their approach, kinematics and mass exchange are coupled by a multiplicative decomposition of the deformation gradient, whereas similar ideas have been previously developed and published in the biomechanical literature in order to model e.g. tumor growth (cf. 3–5). Besides the multi-phase model description, Ristinmaa et al. further assumed that each phase in their model can be viewed as a miscible mixture of species. This extension to the classical Theory of Porous Media<sup>6,7</sup> traces back to contributions of Bennethum and

Cushman<sup>8,9</sup> and was also applied to modeling approaches towards transport processes in paperboard.<sup>10,11</sup> Furthermore, Ristinmaa et al. remark the advantage that material properties of the phases, e.g. elastic parameters, can be expressed in terms of the composition, however, they gave no explicit representation regarding to this. In contrast, Ricken, Robeck and co-workers developed models incorporating mass exchange embedded in the classical TPM.<sup>12–14</sup> Herein, exchange terms are directly modeled through empirical constitutive equations for the density production.

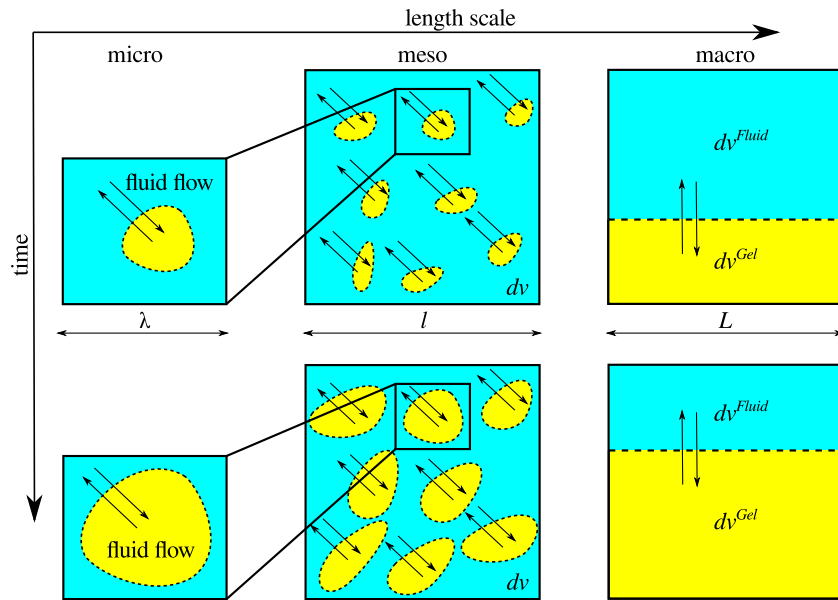
In the present contribution, particular attention is placed on gelation processes induced by inter-phase mass exchange, as a special type of phase transition. The porous material under investigation is composed of a linear-elastic solid phase, whereas the void space is simultaneously filled with an aqueous pore fluid and superabsorbent polymers (SAP). Chemical interactions induce absorption of fluid by the SAP phase, while the latter one swells to a hydrogel. In general, the hydrogel's mechanical properties differentiate tremendous from both the free pore fluid and the dry polymer. Hydrogels with the ability to absorb and bind aqueous solutions have been intensively investigated since the early works of Flory and Rehner.<sup>15–17</sup> The driving forces for absorption and the kinetics of water migration have been widely studied in continuum approaches<sup>18–21</sup> as well as in experimental investigations,<sup>22–24</sup> to mention only a few. The principal difference between modeling

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**Fig. 1.** Inter-phase mass exchange between pore fluid and hydrogel can be observed over time. On the microscale, swelling is a consequence of local fluid flow. Statistical homogenization on the mesoscale leads finally to a macroscopic description (scale separation:  $L \gg l \gg \lambda$ ).

approaches from the literature and the one given in this work is a macroscopic instead of a microscopic description of the swelling process. Fig. 1 illustrates the reaction within the void space of the porous matrix, i.e. the mass exchange between the hydrogel and the aqueous pore fluid. In recent approaches the hydrogel represents the entire system, whereas in this study the hydrogel is treated as a constituent of a multi-phase material besides a solid and a fluid phase. As a consequence, absorption of fluid mass is no longer modeled in terms of flux boundary conditions<sup>25–27</sup> but in terms of inter-phase mass exchange. In this contribution it is assumed that the mechanical response on the macroscale depends not exclusively on the deformation of the solid matrix. In addition, it is assumed that deviatoric stresses due to shearing are also induced in the hydrogel. To this end, a rigorous derivation of a modified effective stress principle incorporating phase transitions will be introduced and extensively discussed. As far as the authors know, there does not exist porous media models in which the void space of the solid skeleton is simultaneously filled with a pore fluid and a (pore) gel. The model proposed in this contribution takes the phase transition process from pore fluid to (pore) gel into account. We are aiming to concentrate in this contribution on the derivation of a comprehensive framework for a generic material which meets the previously described characteristics.

However, there are many promising technical applications. For instance, the three-phase material could be a special kind of grouting mortar in mechanized tunneling. In such scenarios the solid phase represents the grains of the mixture, e.g. sand and gravel, which form the porous matrix. Especially in mechanized tunneling, where the grouting material needs to be pumped through pipes until it reaches its final spatial position, a sufficient workability (i.e. low viscosity) is necessary in this stage. This requirement can be achieved by a high water content in the mixture. As soon as the grouting mortar reaches its ultimate destination, which is the annular gap between the tunnel structure and the surrounding soil, a rapid development of the shear stiffness is necessary in order to minimize surface settlements. To this end, the excess pore water will be usually pressed in the surrounding soil by means of a consolidation process. In case of nearly impermeable soils, the pressure-diffusion-driven consolidation process would be too slow, hence, SAPs in the mortar mixture can be used in order to achieve an immediate increase in stiffness. Finally, a safe bedding

of the tunnel structure even under difficult circumstances can be ensured with such a modified grouting material.<sup>28</sup>

## 2. Preliminaries

In a microscopic description of a multi-phase material, phases can be distinguished, while each material point can be uniquely assigned to a specific phase. However, capturing all geometric information from the micro-structure is a tremendous task and from a practical point of view not manageable for engineering applications. Therefore, the Theory of Mixtures (TM) provides a macroscopic and continuous description of the material, such that the multi-phasic character is accounted by the superposition of the different phases.<sup>29</sup> On a macroscopic level, the medium can be viewed as an immiscible mixture of different phases, each denoted by  $\varphi^\alpha$ , whereas species are assumed to be miscible and therefore are associated with the same volume as the corresponding phase.<sup>10</sup> Note in passing that even through the invention of species, the macroscopic observation is not violated. Adopting the notation for a multi-phase and multi-species system already proposed by Bennethum et al.,<sup>30</sup> phases are denoted by Greek letters, whereas small Latin letters indicate species. Hence,  $\varphi^{\alpha j}$  labels the species  $j$  in phase  $\alpha$ , while the index  $j$  is omitted when referring to a phase. Following that, the infinitesimal volume element which  $\varphi^\alpha$  occupies is denoted by  $dv^\alpha$  in accordance to the notation convention given above. Therefore, volume fractions for each phase are defined, while the saturation condition imposes an algebraic constraint<sup>7</sup>

$$n^\alpha = \frac{dv^\alpha}{dv}; \quad \sum_{\alpha} n^\alpha = 1. \quad (1)$$

In order to account for the composition of the phases, the mass of the species is related to the volume of its corresponding phase, whereas summing over the species yields to the effective (true) phase density

$$\rho^{\alpha j} = \frac{dm^{\alpha j}}{dv^\alpha}; \quad \rho^{\alpha R} = \frac{dm^\alpha}{dv^\alpha} = \sum_j \rho^{\alpha j}. \quad (2)$$

$\rho^{\alpha j}$  is the mass concentration of a component in a mixture and can also be denoted as the density of a component in a multi-species

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