



Non-equilibrium in multiphase multicomponent flow in porous media: An evaporation example



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ABSTRACT

In this paper, the influence of relaxing the local thermal and chemical equilibrium assumptions in multiphase, multicomponent flow in porous media is studied. In order to do so, balance equations allowing for the coupled description of thermal and chemical non-equilibrium are developed. The example application presented is taken from the field of evaporation from porous media. Necessary input parameters are obtained from an extended quasi-static three-dimensional pore-network model. These relations are put into a macro-scale model and simulations not employing local thermal and chemical equilibrium assumptions are conducted. The influence of the rate of heat and mass transfer on evaporation rates is shown and convergence of the model demonstrated.

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1. Introduction

Probably the most basic assumption of modeling multiphase flow in porous media is the assumption of local thermodynamic equilibrium. It is assumed that flow processes are sufficiently slow for full equilibration to be considered instantaneous. However, this assumption might be violated for cases of fast invasion (e.g. in the case of soil remediation [1]). The assumption is even more questionable if there is a heat source present in the porous medium. It could be either present in the solid phase (cooling of nuclear debris after accidents [2]) or on the fluid–fluid interface (in situ combustion of oil [3]).

Smits et al. [4] have tailored a model to the case of evaporation from porous media. They found that incorporating chemical non-equilibrium captures the experimentally observed evaporation from a porous medium into the atmosphere better than an equilibrium model. In this case, the reason for the system not to be in equilibrium is the coupled heat and mass transfer from the porous medium to the atmosphere with constant supply of dry (i.e. chemically not equilibrated) air. This is taken as motivation to use our more general chemical and thermal non-equilibrium model in order to simulate evaporation from a porous medium.

It has to be stressed though that the motivation for this work is not primarily to correctly simulate evaporation from a porous medium. We want to study the implications of relaxing the routinely made assumption of local thermal and chemical equilibrium in multiphase flow in porous media. Nonetheless the presented model encloses the equilibrium situation as limiting case. Model calibration will be conducted in a subsequent step as this would be beyond the scope of this work.

A schematic depiction of the motivating setup is shown in Fig. 1: a confined, initially fully water saturated porous medium is passed by free-flowing dry air. The forced convection of air leads to a continuous supply of dry air and thus evaporation from the porous domain. The passing air is not fully water saturated i.e. it is in a chemical non-equilibrium state. This inevitably leads to an equilibration process, i.e. evaporation which in turn is accompanied by strong thermal effects due to the latent heat of vaporization of water. There is continuous supply of non-equilibrium / dry air and therefore we are not merely looking at an asymptotic equilibration process.

In this paper we will study the influence of the rate of thermal and chemical equilibration on the global evaporation rate as well as the local situation on the free flow – porous-media flow interface. These non-equilibrium effects are the main focus of this paper. In order to take a first step towards incorporation of these effects other relevant processes will be simplified or neglected for the time being. Of course this does not mean that we deem these processes less important, this is rather a step-by-step approach.

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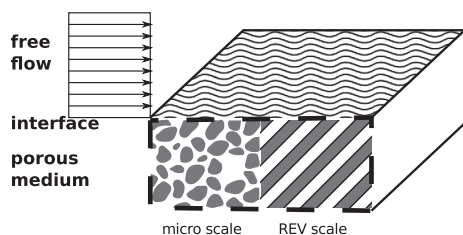


Fig. 1. Terminology and volume averaging used in this paper. The left part shows some grain configuration, which is only incorporated in an averaged (Representative Elementary Volume, REV) sense as shown in the right part.

2. State of the art

This paper deals with the influence of non-equilibrium on multiphase flow in porous media. The example application for this work is evaporation from a porous medium into a free-flow domain. The modeling of coupled porous-medium and free-flow systems has been the subject of lively research over the past years and is of major importance for many different technical, biological and environmental applications [5–7]. Therefore, a brief introduction to the most important aspects of coupled free flow porous-media flow as well as non-equilibrium in multiphase flow in porous media will be given. However, the free-flow system, and its characteristics, are not the focus of this work, and we refer to the literature e.g. Bird et al. [8].

2.1. Porous medium

Darcy's law [9] has been empirically extended to the so-called "extended Darcy's law" in order to describe multiphase flow (e.g. [10,11]). With more than one fluid phase present in the porous medium, the non-linearity of the system is drastically increased: phase pressures differ (capillary pressure) and the presence of one phase inhibits the movement of the other phase (relative permeability).

The extended Darcy's law is established as the standard for the REV scale description of such diverse applications as remediation of contaminated sites, CO₂ storage, and modeling of the cathode side of fuel cells [12–14]. Although the theoretical foundation and correctness of the extended Darcy's law is the subject of ongoing scientific debate (e.g. [15]), its application is largely justified by satisfactorily reproducing experimental data and field scale measurements.

2.2. Equilibrium

In porous-medium systems, local thermodynamic equilibrium between the fluid and solid phases is usually assumed. Local thermodynamic equilibrium is composed of a local thermal, chemical and mechanical equilibrium.

As these terms are crucial to this work, it is justified to introduce them in some more detail. Due to the volume averaged description, mathematically there are potentially several phases present at the same point. If these (fluid) phases do not have the same chemical potential, we call this *local chemical non-equilibrium*.

If the (fluid and solid) phases do not have the same temperature at the same point, we call this *local thermal non-equilibrium*. We acknowledge that especially the last term is also used in a different meaning (the possibility to define temperature at a point). However, with the given definition we find that in this context there is little danger of confusion.

The routinely made assumption of local thermodynamic equilibrium may be violated close to the interface of the free-flow and the porous-medium compartment. Here, the equilibration between the chemically not equilibrated inflowing air and the porous medium takes place. Due to the continuous supply of air, characteristic times might not be long enough to allow for an immediate equilibration of the system. This means that the assumption that transfer processes (mass, momentum and energy) are instantaneous has to be scrutinized.

In the following we will give a brief review of the chemical and thermal part of the thermodynamic equilibrium. In the case of air-water systems it is common to assume local chemical equilibrium and to use *Raoult's* and *Henry's* law for describing the composition of the coexisting fluid phases [10,16]. In the field of groundwater remediation, for example, it can be important to describe the temporal course of the mass transfer process of contaminants between the liquid and the gas phase as opposed to just assuming that mass is transferred instantaneously. Armstrong et al. [17] find that a standard model for multiphase flow in porous media, which assumes local chemical equilibrium, overestimates the mass transfer between the phases. However, Falta [18] argues that this observation is just an apparent kinetic effect. He attributes the observed non-equilibrium to the formation of air channels in the water-saturated porous medium.

Similarly to the discussion of local chemical equilibrium above, local thermal equilibrium might not be a good assumption in the case of processes causing a lot of heat transfer – like evaporation of water – or in the presence of a heat source – e.g. if the cooling of a technical system is to be modeled.

In order to capture the relevant physics, the idea of modeling such situations as several temperatures in the same REV has been developed [19,20]. This means, that the temperature distribution of each coexisting phase is described by individual energy conservation equations. Today, local thermal non-equilibrium for the case of single-phase flow in porous media is covered in textbooks, see Nield and Bejan [21] and references therein. Local thermal non-equilibrium might be caused by transient effects, like the injection of a hot fluid into a cold domain or vice versa [22], the heat generation due to chemical [23] or nuclear [24] reaction or very different thermal properties [25].

The body of literature covering local thermal non-equilibrium in the case of more than one mobile fluid phase present in a porous medium is not extensive. There are approaches [26] which assign the same temperature to both fluid phases, but acknowledge that those might be different from the solid temperature. Models which also allow the fluid phases to have different temperatures are mostly motivated by nuclear safety research, concerning post-accident nuclear debris cooling [2]. Here, the cause for local thermal non-equilibrium is the high initial temperature as well as the heat source in the solid phase. Other authors use three energy equations to describe the thermal equilibration process (along with compositional effects) of a two-phase invasion process into a porous medium [27]. In that work only invasion processes are studied. Quite naturally equilibration is quick because this is a purely asymptotic equilibration process. Therefore, the non-equilibrium situation can only be shown for early times. Nonetheless, the balance equations therein were the starting point for the equations used in this work.

However, the situation for this work is slightly different. There is not only two-phase flow in a porous medium subject to boundary conditions but also a domain with a single free-flowing fluid with a potentially high velocity. Other authors have studied such configurations without assuming local thermal equilibrium [28]. However, they justify this approach with the heat source in the solid phase. In this work there are no external heat sources. Here, the cause of local thermal and chemical non-equilibrium in the porous

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