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Diffusive mass transport in the fluid–porous medium inter-region: Closure problem solution for the one-domain approach

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Abstract

A challenging problem for diffusive mass transport is to describe and model the phenomena concerning the fluid-porous medium inter-region. Volume averaging techniques that provide a framework for rigorously addressing the issue of obtaining macroscopic models from pointwise models at fluid and porous medium scales have been used to attend the problem. The efforts have resulted in two modeling approaches. The first one, known as the one-domain approach (ODA), considers the system as a continuum where the geometrical (e.g., porosity) and transport parameters (e.g., diffusivity) display rapid spatial changes in the inter-region. The second one, known as the two-domain approach (TDA), uses different models for the fluid and the porous medium scales, and matches them via the development of corresponding jump conditions at the dividing surface. Recent results [Wood, B.D, Quintard, M., Whitaker, S. (2000). Jump condition at non-uniform boundaries: the catalytic surface. Chemical Engineering Science 55, 5231–5245; Valdés-Parada, F.J., Goyeau, B., Ochoa-Tapia, J.A. (2006). Diffusive mass transfer between a microporous medium and an homogeneous fluid: jump boundary conditions. Chemical Engineering Science 61, 1692–1704] have shown that the coefficients involved in the jump conditions can be computed by solving the associated closure problems. However, in the development of the jump conditions some complications arise due to the difficulty of modeling some of the "surface excess" transport mechanisms that take place in the inter-region. To address this problem, an implicit formulation based on the ODA and TDA is proposed. Although the ODA seems to be more suitable for modeling, it requires the knowledge of the spatial variations of the transport parameters. Heuristic interpolations between the fluid and the porous medium parameters have been commonly used; however, there is no guarantee that such models can provide an accurate description of the mass flux. Within a ODA framework, the aims of this paper are: (i) to show that the effective diffusivity coefficient for the case of passive diffusion in a fluid–porous medium inter-region can be posed as a closure problem derived from volume averaging techniques and (ii) to use a simple one-dimensional model to show that a complete knowledge of spatial variations of diffusivity and porosity are necessary for an accurate description of the mass transport phenomena in the entire fluid–porous medium system. The analysis has allowed us to identify a new contribution to the jump at the dividing surface. This contribution consists of the accumulation that occurs at the dividing surface even when there is no chemical reaction or adsorption taking place.

Keywords: Porous media; Diffusion; Volume averaging; Closure problem; One-domain approach; Inter-regional transport

1. Introduction

The derivation of effective medium (i.e., macroscopic) equations for transport phenomena in multiphase systems is of prime importance in many applications, from chemical process in porous catalysts to contaminant dispersion in soils. As examples one can cite mass transfer in packed bed reactors (Carberry, 1976; Froment and Bischoff, 1979; Fogler, 1992) which are used in many industrial applications such as methanol synthesis, catalyst regeneration, desulfurization in the steam phase among many others (Balakotaiah and Luss, 1981). Moreover, the mass transfer at the boundary of homogeneous regions is crucial in chromatographic separations (DeVault, 1943; Reis et al., 1979; Raghavan and Ruthven, 1985; Dalvie et al., 1990; Goto and McCoy, 2000). The importance of heat and mass transfer in refrigerated storage and transpiration cooling has been highlighted by many authors (Eckert and Drake,

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1972; Cho and Eckert, 1994; Bird et al., 2002; Verboven et al., 2006) as well as their role on the transport of solutes in aquifers (Pickens and Grisak, 1981; Brusseau et al., 1991). The idea is to depart from pointwise (i.e., local) governing equations to arrive, by means of a suitable up-scaling procedure, to macroscopic equations that describe transport phenomena for physical scales larger than certain point characteristic scales. The most common rigorous approaches are homogenization (Sanchez-Palencia, 1980; Ene and Polisěvski, 1987) and volume averaging (Bear, 1972; Whitaker, 1999). While up-scaling methods involve relatively simple procedures, the problem of estimating effective (macroscopic) transport parameters must be solved. In general, this estimation problem is addressed by means of either ad hoc, heuristic methods or more rigorous approaches that lead to closure problems. Solution of those closure problems allows one to predict the corresponding effective transport coefficients.

In many practical systems with diffusive mass transport, the configuration is composed by a homogeneous fluid region and an adjacent porous medium saturated by the same fluid (Beavers and Joseph, 1967). Examples of this class of configurations are filtration processes, ground water pollution, drying processes, separation membranes, transport in biological tissues, among many others. In general, the transport properties at the fluid bulk are well understood and known. In addition, many efforts have been devoted to the experimental (Hoogschagen, 1955; Currie, 1960; Kim et al., 1987) and theoretical (Wakao and Smith, 1962; Weissberg, 1963; Ryan et al., 1981; Quintard, 1993) determination of effective diffusivities for homogeneous porous media. Within the volume averaging framework, a periodic porous medium model for a two-phase system was proposed to solve a boundary-value problem (the so-called closure problem) that allows one to numerically compute (Ryan et al., 1981; Quintard, 1993) or analytically approximate (Ochoa-Tapia et al., 1986) the components of effective coefficients. However, the description of the transport phenomena around the fluid–porous medium inter-region has received less attention due to the difficulty of understanding the geometrical effects of the transition region on the macroscopic coefficients. For instance, the difficulty of using standard volume averaging results relies on the fact that the length-scale constraints used to perform the up-scaling in the porous medium bulk are not met due to drastic variations of the porous medium properties (e.g., porosity) and transport parameters (e.g., diffusivity) around the fluid–porous medium inter-region. From a modeling standpoint, two approaches have been considered to describe the inter-regional transport phenomena:

- *One-domain approach* (*ODA*): The porous medium is considered as a continuum with effective transport coefficients. The transition from the fluid to the porous medium is achieved through a continuous transition of properties, such as diffusivity and porosity. An advantage of the ODA is that it avoids an explicit formulation of matching conditions at the fluid–porous medium inter-region, which has allowed many applications for extensive numerical simulations of thermal natural convection and double diffusive convection (see Goyeau et al., 2003 and references therein).
- *Two-domain approach* (*TDA*): Here, the porous medium and the fluid are modeled according to the inherent properties of each region. Contrary to the ODA, a model matching problem to couple the transport in both homogeneous regions needs to be addressed, resulting in the so-called jump boundary conditions (Prat, 1989, 1990, 1992; Sahraoui and Kaviany, 1993, 1994; Ochoa-Tapia and Whitaker, 1995, 1997; Valencia-Lopez et al., 2003). These jump conditions often contain coefficients whose dependence of the local geometry of the inter-region is missing. To this end, some approximate approaches have been proposed (Goyeau et al., 2003; Deng and Martinez, 2005; Min and Kim, 2005; Chandesris and Jamet, 2006). In addition, recent developments have been presented to express the jump conditions, for diffusive mass transfer with chemical reaction, in terms of effective coefficients that are computed via the numerical solution of the corresponding closure problems (Wood et al., 2000; Valdés-Parada et al., 2006).

While both the ODA and the TDA can be formulated from a volume averaging framework, some methodological and computational differences can be highlighted. The TDA recognizes the drastic spatial variations at the fluid–porous medium inter-region and poses the problem of describing "excess surface" transport properties, which are used to match the behavior in the fluid to the behavior in the plain porous medium (Ochoa-Tapia and Whitaker, 1995). However, the nature of some excess surface transport quantities is not easily understood and this may limit the applicability of the TDA. Commonly, the resulting jump condition is simplified by neglecting the contribution of some of these transport terms at the dividing surface. On the other hand, reported ODAs use ad hoc descriptions of the spatial variations of geometric parameters. In particular, linear, sinusoidal and error function models, have been used to describe the porosity variations, within a suitably defined boundary layer, when moving from the fluid bulk to the porous medium bulk (Goyeau et al., 2003). It should be obvious that effective transport coefficients, like the effective diffusivity, change continuously from one homogeneous region to another. However, such changes are not necessarily trivial, and this can have a significant effect of the inter-region mass transport. Hence, detailed studies of this issue are required for a better understanding of the mass transport mechanisms around fluid–porous medium inter-regions. Within the ODA modeling framework, this paper focuses on the problem of determining the effective diffusivity in the transition from a fluid to a homogeneous porous medium. The aim of the paper is twofold:

1. To show that, as in the homogeneous porous medium (Whitaker, 1999), boundary-value problems can be posed to determine the effective diffusivity (for the case of a fluid-porous medium inter-region) as closure problems derived from volume averaging techniques. The solution of the resulting closure problems provides the spatial variations of an effective diffusivity coefficient in the inter-region.