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A Theory of Hydrodynamic Dispersion and Reaction in Porous Media Beyond the Long-Time Limit

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In the literature, a range of theoretical approaches has been utilized to study solute transport through porous media. Among these, volume-averaging techniques developed by Whitaker and coworkers, alongside probabilistic methods introduced by Brenner and coworkers, have become prominent within the research community. While these approaches have proven useful, they encounter significant challenges when applied to rapidly changing transient conditions that extend beyond pseudo-steady or quasi-steady states. This study presents a novel theoretical framework for investigating transient solute transport in saturated porous media, accounting for the simultaneous effects of advection, diffusion, and reaction. In many geological, biological, and engineering systems the transient evolution of Darcy-scale concentration field is governed by complex interplay between these pore-scale processes (i.e. advection, diffusion and reaction). Due to the presence of different length scales, such systems are often analyzed using upscaled models derived through homogenization techniques. In such models, the evolution of the local volume-averaged concentration is described by a Darcy-scale mass conservation equation, which relies on upscaled transport coefficients, commonly referred to as Darcy-scale phenomenological coefficients.
The introduced transient theory is proposed for modeling solute transport through porous media comprising of both active and inactive solid/liquid interfaces. The transient problem is studied in the frequency domain by taking the continuous time Fourier transform of time dependent parameters. The mathematical formulation of the transport problem studied is presented for a repetitive unit cell that represents the porous medium. A Darcy-scale mass conservation equation is derived, incorporating three upscaled transport coefficients calculated from the periodic unit cell representation of the system. These upscaled transport coefficients are also interpreted as transfer functions defining an output-input correlations between frequency domain parameters. The first transfer function is the effective diffusion coefficient tensor, which relates the average diffusive flux (as output) to the macroscopic concentration gradient (as input). The second, the advection suppression vector, connects the fractional deviation of the advection rate from its expected value (e.g., the product of average velocity and average concentration) as the output to the pore-scale concentration difference as the input. The third transfer function, the effective reaction rate transfer function, links the average surface flux (output) to the microscopic concentration difference (input). Analytical expressions for the longitudinal component of the effective diffusion coefficient tensor in the flow direction for Poiseuille flow through active and inactive parallel plates and circular tubes are also derived. Additionally, the theory is applied to obtain time-domain breakthrough curves for Poiseuille flow through parallel plates. The results are compared with direct pore-scale solutions and conventional theory of pseudo-steady dispersion. This comparison reveals that the proposed transient theory delivers more accurate predictions for the transient evolution of the average concentration over time compared to the conventional theory.

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References

Author: HAMID, Md Abdul (University of Illinois Urbana-Champaign)

Co-author: Dr SMITH, Kyle (University of Illinois Urbana-Champaign)

Presenter: HAMID, Md Abdul (University of Illinois Urbana-Champaign)

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