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Magnetic resonance microscopy of biofouling induced scale dependent transport in porous media

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Abstract

Non-invasive magnetic resonance microscopy (MRM) methods are applied to study biofouling of a homogeneous model porous media. MRM of the biofilm biomass using magnetic relaxation weighting shows the heterogeneous nature of the spatial distribution of the biomass as a function of growth. Spatially resolved MRM velocity maps indicate a strong variation in the pore scale velocity as a function of biofilm growth. The hydrodynamic dispersion dynamics for flow through the porous media is quantitatively characterized using a pulsed gradient spin echo technique to measure the propagator of the motion. The propagator indicates a transition in transport dynamics from a Gaussian normal diffusion process following a normal advection diffusion equation to anomalous transport as a function of biofilm growth. Continuous time random walk models resulting in a time fractional advection diffusion equation are shown to model the transition from normal to anomalous transport in the context of a conceptual model for the biofouling. The initially homogeneous porous media is transformed into a more complex heterogeneous porous media by the biofilm growth.

Keywords: NMR; MRI; Biofouling; Biofilms; Porous media; Anomalous transport

1. Introduction

Experimental data on the impact of biological activity on transport in porous media in three dimensions has been limited to bulk measurements of pressure drop and tracer breakthrough curves [1,2]. Magnetic resonance (MR) methods provide the ability to non-invasively monitor transport processes and biomass accumulation and distribution [3–6]. The application of MR methods to study environmental science (special issue J Env Qual 2002;31(2)) and biological activity in porous systems [3] has been reviewed. MR studies of transport reported in the hydrology literature have tended to focus on application of spatially resolved velocity imaging [7–9] from which the permeability can be rigorously determined [10]. In this paper, a short overview of the measurement of scale dependent dispersion dynamics by pulsed gradient spin echo (PGSE) MR is given, as much of the research in this area has been reported in the physics, engineering and MR literature [11–18].

The analysis of bioactivity applying these scale dependent techniques, provides quantitative measurement of the change in hydrodynamic dispersion dynamics due to biofouling [6]. The MR data indicate a transition caused by biofilm growth, from normal Gaussian dynamics due to a Fickian dispersion process for flow through the homogenous model media, to anomalous non-Gaussian dynamics [19–21] due to non-Fickian dispersion [6]. The data is discussed in the context of fractional advection dispersion equations and indicate the applicability of continuous time random walk (CTRW) theory to model the transition [22–25]. The biofilm growth alters the homogeneous porous media system structure, increasing the system complexity

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[26] and transforming the porous media into a heterogeneous system. The dispersion data presented, clearly indicate the ability of displacement scale dependent PGSE MR methods to characterize anomalous transport [27].

2. Hydrodynamic dispersion

The classic theory of hydrodynamic dispersion in porous media is based in large part on the work of G.I. Taylor who first derived the celebrated Ornstein–Uhlenbeck process of stochastic dynamics [28,29]. Taylor later demonstrated that coarse graining [30] through averaging of continuum models results in effective Brownian motion processes [31,32]. The fundamental aspect of models of dispersion in porous media is the resultant advection diffusion equation (ADE) governing mass, or probability, conservation in the system. An effective diffusion coefficient, *i.e.* dispersion coefficient, dependent upon the fluctuations about the mean velocity, quantifies transport in the system. Many theoretical approaches to derive the conservation equation and the corresponding transport coefficients have been applied and result in consistent governing equations [33].

Averaging of the continuum mass conservation equations using generalized macrotransport theory based on the method of moments [34] and stochastic process methods based on the central limit theorem [35], return the classical ADE. The ADE is applicable to homogeneous porous media systems in which the dynamics are Gaussian in the asymptotic time limit. In this case the dispersion coefficient is a constant and the mean squared displacement, or positional variance, scales linearly in time. In systems in which long range correlations in the transport dynamics occur due to the heterogeneous nature of the porous media system, non-local transport theories based on ensemble averaging of the continuum momentum and mass conservation equations [36] and nonequilibrium statistical mechanics [37], among other methods, have been applied. Dispersion coefficients in the non-local formulation are dependent on the displacement length and time scale and result in a mean squared displacement which scales non-linearly in time, the definition of anomalous diffusion [38]. More recently transport in heterogeneous porous media has been modeled using the theory of CTRWs which lead to fractional advection diffusion equation (FADE) mass conservation models [19,20,25].

2.1. Normal diffusion: ADE

The theory of Brownian motion involves the averaging or coarse graining of fast variables in a system, a separation of scales [30]. In the case of a colloidal particle suspended in a liquid, the velocity fluctuations of the particle occur rapidly relative to the position variation and the treatment results in a Fokker–Planck, or Smoluchowski equation, governing the particle concentration or probability [32]. As indicated above, the long time asymptotic behavior of the process is Gaussian with a constant effective diffusion, *i.e.* dispersion, coefficient, and linear scaling of the mean squared displacement in time. In the non-asymptotic short time regime the time dependence of the mean squared displacement is quadratic in time consistent with the ballistic motion of a particle with a constant velocity. The Brownian motion process has been solved for full time dependence and is the Ornstein–Uhlenbeck stochastic process [28–30]. Preasymptotic time dependence of the normal Brownian motion diffusion process, during which the mean squared displacement transitions from the short time quadratic time scaling behavior of the ballistic regime, to the long time linear scaling of the diffusive regime, to the long time linear scaling of the diffusive regime, is in contrast to anomalous diffusion in which non-linear time scaling of the mean squared displacement persists for asymptotic times [21,38].

The mass, or probability, conservation equation for axial z-direction flow in a porous media is given by the ADE

$$\frac{\partial P(Z,t)}{\partial t} = \left[-\langle v_z\rangle \frac{\partial}{\partial Z} + D^* \frac{\partial^2}{\partial Z^2}\right] P(Z,t). \qquad (1)$$

The time rate of change of the probability of a tracer, or solute particle (molecule) $P(Z,t)$ having displacement Z in time t is dependent on the advection due to the mean velocity $\langle v_z\rangle$ and the dispersion coefficient D^*. The ADE is Galilei invariant, meaning the distribution is invariantly translated with the average velocity. The effective diffusion, or dispersion, coefficient depends on the velocity fluctuation autocorrelation function $D^*(\Delta) = \int_0^{\Delta}(1 - \frac{\tau}{\Delta}) \langle [v_z(t+\tau) - \langle v_z\rangle][v_z(t) - \langle v_z\rangle]\rangle \, d\tau$ and can be written in time dependent fashion where Δ is the observation time scale [30]. The effective diffusion for the time dependent Brownian motion Ornstein–Uhlenbeck process is recovered for an exponential velocity fluctuation autocorrelation. In the asymptotic limit, for times Δ much larger than the correlation time of the process, $D^* = \lim_{\Delta\to\infty} D^*(\Delta) = \int_0^{\infty}\langle [v_z(t+\tau) - \langle v_z\rangle][v_z(t) - \langle v_z\rangle]\rangle \, d\tau$ and the mean squared displacement of the solute or tracer in the long time limit is $\langle Z(t)^2\rangle - \langle Z(t)\rangle^2 = 2D^*t$. The random fluctuations in velocity about the mean velocity at the pore scale generate an effective Brownian motion diffusion process at the macroscopic scale. In classical continuum modeling of porous media transport it is the averaging of the continuum equations of conservation of mass and momentum which provides the coarse graining that results in the Brownian motion nature of the model.

2.2. Anomalous diffusion: FADE model of biofouling

The separation of scales which occurs in development of the Brownian motion theory is the reason the ADE fails to model macroscopic transport in systems with heterogeneity over many scales. Correlated dynamics are generated by the heterogeneities in structure and corresponding transport properties, *e.g.* permeability, that results in non-local