Hybrid multiscale simulation of a mixing-controlled reaction

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ARTICLE INFO

ABSTRACT

Continuum-scale models, which employ a porous medium conceptualization to represent properties and processes averaged over a large number of solid grains and pore spaces, are widely used to study subsurface flow and reactive transport. Recently, pore-scale models, which explicitly resolve individual soil grains and pores, have been developed to more accurately model and study pore-scale phenomena, such as mineral precipitation and dissolution reactions, microbially-mediated surface reactions, and other complex processes. However, these highly-resolved models are prohibitively expensive for modeling domains of sizes relevant to practical problems. To broaden the utility of pore-scale models for larger domains, we developed a hybrid multiscale model that initially simulates the full domain at the continuum scale and applies a pore-scale model only to areas of high reactivity. Since the location and number of pore-scale model regions in the model varies as the reactions proceed, an adaptive script defines the number and location of pore regions within each continuum iteration and initializes pore-scale simulations from macroscale information. Another script communicates information from the pore-scale simulation results back to the continuum scale. These components provide loose coupling between the pore- and continuum-scale codes into a single hybrid multiscale model implemented within the SWIFT workflow environment. In this paper, we consider an irreversible homogeneous bimolecular reaction (two solutes reacting to form a third solute) in a 2D test problem. This paper is focused on the approach used for multiscale coupling between pore- and continuum-scale models, application to a realistic test problem, and implications of the results for predictive simulation of mixing-controlled reactions in porous media. Our results and analysis demonstrate that the hybrid multiscale method provides a feasible approach for increasing the accuracy of subsurface reactive transport simulations.

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1. Introduction

Granular porous media are made up of solid grains of variable size and shape with intervening pore spaces filled with one or more fluids (e.g., water, air, non-aqueous-phase liquids). However, because of the difficulty of characterizing properties and simulating processes at a scale that explicitly resolves these phases, conventional simulations of flow and reactive transport in porous media treat the medium as an effective continuum. The rates and directions of flow, transport and reaction are defined over a "representative elementary volume" (REV) of porous media for which the effects of variations in fluid flow and solute diffusion within individual pores are approximated by apparent or upscaled model forms and parameters (e.g., Darcy's law and the advection–dispersion–reaction equation, parameterized by the hydraulic conductivity tensor, porosity, longitudinal and transverse dispersivities, and effective reaction rate models). Here, we refer to models that rely on this conceptualization as continuum-scale models. In contrast, it is also possible to explicitly simulate processes at the pore scale, at which the details of solid and pore geometry can be directly incorporated and velocity variations can be resolved at the scale of tens of micrometers or smaller (i.e., "pore-scale models"). Well-developed pore-scale modeling methodologies include the lattice–Boltzmann (LB) method [1–8], the smoothed particle hydrodynamics (SPH) method [9–16], computational fluid dynamics (CFD) methods [17–26], and pore network models [27–38]. These models have proven useful for gaining new insights into the relationship between fundamental processes defined at the pore scale and upscaled phenomena observed at Darcy scales. Significant advances have been made in three-dimensional (3D) characterization of porous media structure, using methods such as X-ray computed tomography (XCT) and magnetic resonance imaging (MRI), and quantitative descriptions of pore geometry with

sufficient resolution to facilitate pore-scale simulations [39]. Simultaneous advances in high-performance computing have provided the computational power necessary to perform pore-scale simulations over complex 3D domains. However, despite these advances pore-scale simulations remain extremely computationally expensive, and have typically been performed on millimeter-scale or smaller domains, with the most advanced high-performance computing applications to date enabling simulations on centimeter- to decimeter-scale domains [17,26,40,41]. Therefore, it is currently infeasible to perform simulations on domain sizes of practical interest (scales of meters to kilometers) with complete pore-scale resolution, and will likely remain so for some time to come.

Nevertheless, recent studies have pointed to the potential need for pore-scale resolution of processes for accurate simulation of some types of reactive transport problems [42,43]. Of particular note are reactive transport processes in which reaction rates are not limited by fundamental molecular interactions, but instead are controlled by mixing processes (primarily diffusion) at interfaces between fluid and/or solid phases containing the various reactants [44–46]. These mixing-controlled reactions are often characterized by sharp local gradients and localized reaction zones; example problems include biologically-mediated natural attenuation of dissolved contaminant plumes [47,48], precipitation/dissolution reactions [10,49,50], incomplete mixing effects on reaction rates [51–57] and biofilm dynamics [11,58]. There have been many representative pore-scale modeling works on transverse mixing-controlled reactive transport [58–66]. A brief review of mixing, spreading and reaction in heterogeneous media has been given in [46]. Battiato et al. [67–69] developed a multiscale analysis of a mixing-controlled precipitation reaction that identified sufficient conditions (in terms of ranges of dimensionless Péclet (Pe) and Damköhler (Da) numbers) under which macroscopic advection–dispersion–reaction equations provided an accurate description of the pore-scale processes. Outside of these ranges, it was postulated that pore-scale processes must be explicitly simulated to obtain an accurate solution.

Problems in which explicit pore-scale simulation is essential, in the context of computational limitations of pore-scale simulation over large domains, motivate a hybrid multiscale simulation approach in which pore- and continuum-scale models are directly coupled within a single simulation domain. Scheibe et al. [70] provide a review of hybrid multiscale simulation methods and their applications in a number of scientific domains including recent developments in modeling of reactive transport in porous and fractured media. Tartakovsky and Scheibe [71] demonstrated an approach for coupling continuum- and pore-scale models applied to a mixing-controlled precipitation reaction over a small (mm-scale) domain. In this paper, we extend their approach to a much larger domain similar to the benchtop laboratory experiment described in [10]. We couple a pore-scale model based on the SPH code described in [72] with a continuum model based on the subsurface transport over multiple phases (STOMP) simulator [73]. The study of [71] considered a single pore-scale domain corresponding to one continuum-scale grid element as a test case; here we extend this approach to consider multiple pore-scale SPH domains coupled to a two-dimensional array of STOMP grid elements representing a realistic problem of interest. While Tartakovsky and Scheibe [71] simulated a mixing-controlled bimolecular reversible mineral precipitation reaction, here for the sake of computational simplification we consider an irreversible homogeneous bimolecular reaction (two solutes reacting to form a third solute) similar to the problem studied by [9]. We have developed a parallel workflow structure based on the Swift workflow environment [74,75] that manages the complex process of executing many coupled pore- and continuum-scale code runs over the course of a single integrated multiscale hybrid simulation. Scheibe et al. [76] provide a description of the computational workflow management aspects of this work, which take advantage of multiple levels of parallelism inherent in the problem to scale the problem to a massively parallel supercomputing environment. Here, we focus on the approach used for multiscale coupling between pore- and continuum-scale models, application to a realistic test problem, and implications of the results for predictive simulation of mixing-controlled reactions in porous media.

2. Methods

In this section, we present the numerical methods for the pore-scale, continuum-scale and the hybrid multiscale models respectively, which include the governing equations, numerical methods, target problem and setup. The concept and the execution of the coupled hybrid workflow are introduced subsequently.

2.1. Pore-scale simulation

The SPH method is a fully Lagrangian mesh-free particle-based method, and directly solves discretized forms of the partial differential equations describing fluid flow and other porous media dynamics. It was initially developed for applications in astrophysics, but has recently been applied to a number of porous media flow and reactive transport problems [9–15]. SPH is also widely used for computer-generated animation used in gaming and commercial media, particularly to simulate fluid flows with free surfaces (e.g., [81,82]). An overview of the method and applications is provided by [16]. Because discretization points ("particles") can move freely within the simulation domain, the SPH method is particularly well suited to simulation of problems that involve moving interfaces and dynamic pore geometry, including pore-scale precipitation/dissolution reactions. Although in this initial study we consider a solute-phase reaction only, which could be easily solved with a mesh-based method, we intend in future work to apply the same approach to a mixing-controlled precipitation reaction system with dynamic pore geometry. Therefore, and because we have available in-house a scalable parallel implementation of SPH [77], we chose the SPH method for our pore-scale simulator. However, we note that the methodology used here to couple pore- and continuum-scale simulations could be applied to other simulation methods and codes.

For purposes of testing the hybrid multiscale coupling approach, we chose to simulate a two-dimensional model system, both at pore- and continuum-scales. At the pore scale, the fundamental element is taken to be a 0.5 cm by 0.5 cm domain containing a regular pattern of uniform spherical grains as shown in Fig. 1. This domain has periodic geometry, which facilitates handling of boundary conditions in the SPH simulations (described in greater detail below). The porosity of this domain is $\theta = 0.446$, and the diameter of the spherical grains is $d_g = 0.7$ mm. The system is discretized using $200 \times 200 = 40,000$ SPH particles, at an initial uniform spacing of 0.025 mm, which provides a minimum resolution of five particles spanning the narrowest pore throats. A series of preliminary simulations were performed using the SPH code to define an appropriate parameterization. In particular, the applied body force ("gravity" parameter) was adjusted iteratively to obtain an average vertical velocity consistent with the macroscopic simulation, and other parameters (such as the stiffness parameter c) were selected to maximize computational stability and efficiency while maintaining water-like fluid properties. In these simulations, the Reynolds number of the system (Re) is less than unity, indicating creeping flow. Boundary conditions for flow are periodic; that is, any particle that leaves the domain is re-introduced at the corresponding point on the opposite boundary. Therefore, while the average horizontal velocity is zero, local fluxes can occur through the lateral boundaries as dictated by the local pore geometry.

We simulate a mixing-controlled reaction of two non-specific solutes (A and B) that react irreversibly to form a third solute (C)