Particle methods for simulation of subsurface multiphase fluid flow and biogeochemical processes.

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Abstract

A number of particle models that are suitable for simulating multiphase fluid flow and biogeochemical processes have been developed during the last few decades. Here we discuss three of them: a microscopic model - molecular dynamics; a mesoscopic model - dissipative particle dynamics; and a macroscopic model - smoothed particle hydrodynamics. Particle methods are robust and versatile, and it is relatively easy to add additional physical, chemical and biological processes into particle codes. However, the computational efficiency of particle methods is low relative to continuum methods. Multiscale particle methods and hybrid (particle–particle and particle–continuum) methods are needed to improve computational efficiency and make effective use of emerging computational capabilities. These new methods are under development

Introduction

The computational methods used to simulate single- and multi-phase fluid flow can be divided into two general classes: continuum methods and particle methods. Hybrid particle-continuum methods have also been developed, and some models, such as smoothed particle hydrodynamics and lattice Boltzmann models, can be considered to be either continuum or particle methods. Particle models that can be used to simulate single- and multi-phase fluid dynamics include lattice gas models [Frisch *et al.*, 1986], Monte Carlo methods [Bird, 1994], vortex particle methods [Cottett and Koumoutsakos, 2000] and the fluid-particle model [Espanol, 1998]. Here we focus on a microscopic model - molecular dynamics; a mesoscopic model - dissipative particle dynamics; and a macroscopic model - smoothed particle hydrodynamics.

Molecular Dynamics

Molecular dynamics has been used to simulate both single-phase [Rapaport and Clementi, 1986, for example] and multiphase [Thompson and Robbins, 1989, for example] fluid dynamics. However, the strain rates associated with molecular dynamics simulations are much higher than those encountered under typical experimental conditions. The primary reason for this is the very short time step, $O(10^{-15}\ sec)$ required to obtain accurate results. In the case of a two phase fluid composed of simple molecular constituents (water and carbon tetrachloride, for example) a simulation with 10^6 molecules consisting of 10^9 time steps would simulate the system for a physical time of the order of $10^{-6}\ sec$. Even by today's standards, this would be a large scale molecular dynamics simulation. The characteristic scale of the system, *L*, is a few tens of nanometers, and a strain of at least 10, and in some cases much greater, would be required to simulate the characteristics of multiphase fluid flow. Consequently, the strain rate would be $O(10^7\ sec^{-1})$, and the fluid velocity would be a few tens of *cm sec*$^{-1}$. The no-slip boundary conditions used in continuum hydrodynamics are violated at molecular scales, and the velocity of the fluid in contact with a stationary solid surface, the slip velocity V_S is given approximately by

$V_S = L_S \dot{\varepsilon}$, where L_S is the slip length and $\dot{\varepsilon}$ is the strain rate. Typically slip lengths are on the order of a few nanometers. In order for the molecular dynamics simulation to accurately simulate fluid flow on large scales, the slip length must be much smaller than the characteristic scale of the flow ($L_S << L$). Unless artificial no-slip behavior is imposed at the solid-liquid interface, this condition is likely to be violated in molecular dynamics simulations. An additional problem arises because the structure of liquids adjacent to (within a few molecular diameters from) a solid surface is different from the bulk liquid structure, and properties such as the viscosity can also be expected to be different. For the hypothetical 10^6 molecule simulation, on the order of 1/10 of the fluid could be significantly effected by these "solid wall" effects.

The effect of slip at the solid-liquid interface could be reduced by increasing the number of molecules used in the simulation. For example, the number of particles could be increased by a factor of 1000 (to 10^9), and the number of time steps reduced by a factor of 1000 (to 10^6) to approximately match the computational resources required for the two simulations. This would reduce the effects of slip by a factor of 10. However, the strain rate would increase to 10^{10} sec^{-1} (a value typical of a nuclear explosion) and the fluid velocity would increase to about 3000 m sec^{-1} (the flow would be supersonic). In order for compressibility effects to be small the Mach number ($Ma = V / C$, where V is the fluid velocity and C is the sound velocity in the fluid) must be less than ≈ 0.1. Clearly, compressibility effects would be very large in this simulation.

For an experimental model or computer model to quantitatively reproduce the behavior observed in a physical system, all of the important dimensionless ratios in the model and the system must either be equal, both very large or both very small. In the case of fluid flow in the subsurface, the important dimensionless ratios are: 1. the Reynolds number,

$$\mathrm{Re} = VL\rho / \eta, \tag{1}$$

where ρ is the fluid density and η is the viscosity; 2. The Bond number,

$$B_O = \delta\rho g L^2 / \Gamma, \tag{2}$$

where $\delta\rho$ is the difference in density between the two fluids, Γ is the interfacial energy per unit area and g is the gravitational velocity; and 3. the capillary number,

$$Ca = \eta V / \Gamma. \tag{3}$$

In most cases $\mathrm{Re} << \mathrm{Re}^{(C)}$, where $\mathrm{Re}^{(C)}$ is the critical Reynolds number above which the flow is turbulent.

Using a viscosity of $\eta = 0.01$ g cm^{-1} sec^{-1}, and a surface tension of $\Gamma = 75$ g sec^{-2} (approximate values for water at room temperature) the capillary number, Ca, would have a value of about 4×10^{-4} for the hypothetical 10^6 particle simulation, with a velocity of about 30 cm sec^{-1}, and for the 10^9 particle simulation, the capillary number would be about 4. These values are typical of multiphase fluid flow in subsurface porous media, but a capillary number of 4 would be unusually high under typical natural conditions. Similarly, the Reynolds number would be approximately 10^{-2} for the 10^6 particle simulation and about 10^2 for 10^9 particle simulation. In both cases, Re would be below the critical Reynolds number, and the flow in the model system would be laminar, which is typical of flow in subsurface porous media and small aperture fractures. To drive the fluid at a velocity of 30 cm sec^{-1} in the 10^6 particle simulation a body force corresponding to a gravitational acceleration of the order of 10^{12} cm sec^{-2} (about 10^9 g_O, where g_O is the acceleration in the Earth's gravitational field) would be required to reach an asymptotic flow velocity of 30 cm sec^{-1}. This is much larger than the acceleration required to reach a fluid velocity of 30 cm sec^{-1} in 10^{-6} sec. For these conditions, the Bond

number, for a density difference of $\delta\rho = 1.0$ g cm^{-3} is of the order of 0.1. For the 10^9 particle simulation an acceleration of 10^{14} cm^2 sec^{-1} (about 10^{11} g_O) would be required to sustain a velocity of 3000 m sec-1, and under these conditions the Bond number would have a value of the order of 1000. However, an acceleration of the order of 10^{16} cm^2 sec^{-1} would be required for the fluid to reach an average velocity of 3000 m sec^{-1} in 10^{-9} seconds, and if this value is used for g, a Bond number of O(10^5) is estimated.

These estimates of the relevant dimensionless ratios are based on the assumption that the very large strain rates do not significantly change the fluid structure and the associated fluid properties, such as the viscosity. If this is true, the compressibility effects of large Mach numbers and the effects of slip at the solid-fluid interfaces and the influence of the solid surfaces on the neighboring fluid structure are not too large, the estimates of the dimensionless ratios indicate that it might be possible to use large scale molecular dynamics to simulate multiphase fluid flow in confined systems such as fractured and porous subsurface materials. Molecular dynamics simulations of water [Balasubramanian *et al.*, 1996] indicate that shear thinning becomes significant (O(10%)) only at shear rates of about 10^{11} sec^{-1}. Another issue is the very rapid heating caused by very high strain rates. The rate at which heat is generated per unit volume by thermal dissipation is given by $Q = (\dot{\varepsilon} : \boldsymbol{\eta} : \dot{\varepsilon})$, where $\dot{\varepsilon}$ is the second-rank strain-rate tensor and $\boldsymbol{\eta}$ is the fourth rank viscosity tensor. For the 10^6 particle simulation, with a strain rate of O(10^7 sec-1), the heating rate would be of order 10^6 K sec^{-1}, and the temperature would rise by about 1 K during the simulation. For the 10^9 particle simulation with a strain rate of O(10^{10} sec^{-1}) the heating rate would be of order 10^{12} K sec^{-1}, and the temperature would rise by about 1000 K during the simulation. An effective "thermostat" would be needed to prevent a transition from a two phase liquid-liquid or liquid-vapor system to a single phase supercritical fluid and to prevent the properties of the fluids from deviating substantially from their initial values. Such thermostats are used in most non-equilibrium molecular dynamics simulations [Nose, 1982; Hoover 1985; Andersen, 1980].

One of the important advantages of molecular dynamics is its ability to simulate the velocity dependent contact angle behavior, slip effects at solid-liquid interfaces and the complex dynamics in the vicinity of the fluid-fluid-solid contact line, and to couple it with the fluid dynamics [Koplik *et al.*, 1988; Thompson and Robbins, 1989; Barret and Bocquet, 1999, for example].

Continuum and Particle Methods

Under a wide range of flow conditions, fluid dynamics can be simulated quite accurately by the Navier-Stokes equation, which consist of a conservation of momentum equation

$$\partial\rho\mathbf{V}/\partial t = -\mathbf{V}\bullet\nabla\rho\mathbf{v} - \nabla P + \eta\nabla\bullet\nabla\mathbf{V} + \mathbf{f} \,, \tag{4}$$

where $\mathbf{V}$ is the fluid velocity, P is the pressure, and $\mathbf{f}$ is the body force per unit volume, and a conservation of fluid volume equation

$$\nabla\bullet\mathbf{v} = 0. \tag{5}$$

The validity of the Navier-Stokes equations does not depend on details such as molecular sizes, shapes and interactions. Instead, it is based on the conservation of momentum and mass on the molecular level, and symmetries such as isotropy and Galilean invariance. Essentially any particle-based microdynamics model that conforms to these conservation principles and symmetries on a microscopic level, and has Knudsen number, K_n, that is much less than unity ($K_n = \lambda_f / L$, where λ_f is the particle mean free path) will conform to the Navier-Stokes

equation if compressibility effects are small (if mass conservation implies conservation of fluid volume). This suggests that the applicability of molecular dynamics to multiphase fluid flow can be improved by using point mass particles (instead of molecules with complex shapes, interactions and rotational degrees of freedom) with "soft" particle-particle interactions that allow long time steps to be taken. The compressibility of soft sphere fluids is much larger than the compressibility of typical molecular liquids, but compressibility effects are still quite small if the Mach number is smaller than O(0.1).

Dissipative Particle Dynamics

The dissipative particle dynamics (DPD) model [Hoogebrugge and Koelman, 1992] is based on the idea that particles can be used to represent clusters of atoms or molecules instead of single atoms or molecules. Because of the internal degrees of freedom, the particle-particle interactions include fluctuating and dissipative components. The DPD equation of motion for the *i*th particle is

$$dm_i\mathbf{V}_i/dt = \mathbf{f}_i^{\text{int}} + \mathbf{f}_i^{ext} = \mathbf{f}_i^C + \mathbf{f}_i^D + \mathbf{f}_i^R + \mathbf{f}_i^{ext} = \sum_{j\neq i}\mathbf{f}_{ij}^{\text{int}} + \mathbf{f}_{ij}^{ext}, \tag{6}$$

where $\mathbf{f}_i^C$ is the conservative force, $\mathbf{f}_i^D$ is the dissipative force, $\mathbf{f}_i^R$ is the random force, $\mathbf{f}_i^{ext}$ is the body force acting on particle *i*, and $\mathbf{f}_{ij}$ is the force acting on particle *i* due to interaction with particle *j*. In models for single phase fluids, the conservative force between particles is given a simple soft purely repulsive form such as $\mathbf{f}_{ij}^C = -S_{ij}(1 - r_{ij}/r_0)\hat{\mathbf{r}}_{ij}$ for $r_{ij} > r_0$ and $\mathbf{f}_{ij}^C = 0$ for $r_{ij} < r_0$, where $\hat{\mathbf{r}}_{ij}$ is the unit vector pointing from particle *i* to particle *j* so that

$$\mathbf{f}_i^C = \sum_{j\neq i}\mathbf{f}_{ij}^C = -\sum_{j\neq i}S_{ij}(1 - r_{ij}/r_0)\hat{\mathbf{r}}_{ij}. \tag{7}$$

The dissipative particle-particle interactions are given by $\mathbf{f}_{ij}^D = -\gamma W^D(r_{ij}(\mathbf{r}_{ij}\cdot\mathbf{v}_{ij})\hat{\mathbf{r}}_{ij}$, where $r_{ij} = |\mathbf{r}_{ij}|$ so that

$$\mathbf{f}_i^D = \sum_{j\neq i}\mathbf{f}_{ij}^D = -\sum_{j\neq i}\gamma W^D(r_{ij})(\mathbf{r}_{ij}\cdot\mathbf{v}_{ij})\hat{\mathbf{r}}_{ij}, \tag{8}$$

and the random forces are given by $\mathbf{f}_{ij}^R = \sigma W^R(r_{ij})\zeta\mathbf{r}_{ij}$ where ζ is a random variable with a zero mean and a unit variance. The random and dissipative particle-particle interactions are related through the fluctuation dissipation theorem, which requires that $\gamma = \sigma^2/2k_BT$, where k_B is the Boltzmann constant, and $W^D(r) = (W^R(r))^2$. The combination of dissipative and fluctuating forces, related by the fluctuation-dissipation theorem [Kubo, 1966; Espanol and Warren, 1995] acts as a thermostat, which maintains the temperature of the system, measured through the average kinetic energy of the particles in a coordinate system moving with the fluid, at a temperature of T, providing that the time step used in the simulation is small enough. Consequently, the DPD model can be regarded as a form of thermostatted nonequilbrium molecular dynamics with a soft interaction potential [Soddermann *et al.* 2003].

When DPD models are used to simulate liquids, the particles overlap extensively (there are several tens of particles in a volume of $4\pi r_o^3/3$). As the temperature is lowered from a large value, the DPD system undergoes a Kirkwood-Alder transition to a solid [Kirkwood 1939, Alder and Wainright, 1962]. The transition temperature depends on the magnitude of the thermal energy relative to the interaction energy. As the number of atoms or molecules, N_p, represented

by the DPD particle increases, the thermal energy, $k_B T / 2$ per degree of freedom remains constant while the DPD particle-particle interaction increases in magnitude. This drives the system through the Kirkwood-Alder transition and limits the size of the cluster of atoms or molecules that the DPD particles can represent.

In the standard DPD model, the particle-particle interaction is purely repulsive. Multi-component, multi-phase systems can be simulated by labeling the particles (1, ...) to identify which component they represent, and using different repulsive interaction strengths (equation 7) ($S_{11}, S_{12}, S_{22} ...$) with $S_{11} << S_{12}, S_{22} << S_{12}$ etc. to bring about phase separation. However, this approach cannot be used to simulate single component two phase (liquid – vapor) systems. An alternative approach is to use a combination of short range repulsive interactions and (relatively) long range attractive interactions [Liu *et al.,* 2007]. This borrows from the physical origins of phase separation in single component fluids (the combination of short range repulsive and long-range attractive atom-atom and molecule-molecule interactions [van der Waals, 1873]). A combination of short range repulsive and relatively long range attractive interactions between fluid particles and particles used to represent confining solid materials can also be used to realistically simulate different wetting conditions (velocity dependent contact angle angles and contact line dynamics). In addition a combination of "bounce back" boundary conditions and particle–particle interactions can be used to reduce slip at solid-liquid interfaces to negligible levels. These features were used in the simulation shown in Figure 1.
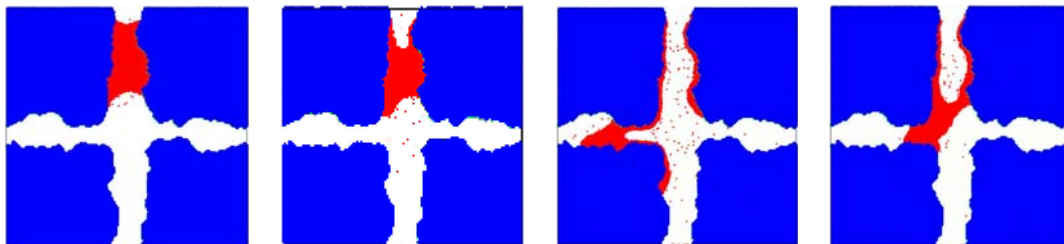

Figure 1: Four stages in a two-dimensional dissipative particle dynamics simulation of the penetration of a wetting fluid through a fracture junction

DPD models have been used to simulate a variety of soft condensed matter systems. For example, polymer molecules can be simulated by connecting together a string of DPD particles using harmonic springs [Groot and Madden, 1998, for example], finitely extensible nonlinear elastic (FENE) links [Fan *et al.*, 2003, for example], or other links. Concentrated colloids [Boek *et al.*, 1997, for example] amphiphilic vesicles [Yamamo *et al.*, 2002, for example] and membranes [Li *et al.*, 2004] have also been simulated using DPD models. When DPD models are used to simulate suspensions of colloidal particles or polymer solutions, gels etc., the particles are used to represent both the particles or polymers and the surrounding fluid. As a consequence particle- particle and both intramolecular and intermolecular hydrodynamic interactions are automatically included. From a rheo-mechanical point of view, a wide range of biological systems lie within the realm of soft condensed matter physics. These include biofilm that forms under nutrient rich conditions caused by organic contaminants and/or the addition of nutrient to accelerate natural bioremediation contains high concentrations of colloids (living and dead cells) and polymers (polysaccharides and other polymers secreted by subsurface microorganisms). In the SciDAC project on "Hybrid numerical methods for multiscale simulations of biogeochemical processes" we will build on earlier DPD work by including processes such as growth, breaking of polymer molecules and their detachment from and /attachment to cells.

Smoothed Particle Hydrodynamics

Smoothed particle hydrodynamics (SPH) was introduced more than a quarter of a century ago to simulate astrophysical fluid dynamics [Lucy, 1977; Gingold and Monaghan 1977]. It is a fully Lagrangian particle method that has been used extensively to simulate the behavior of materials (both solids and liquids) subjected to large strains. SPH is based on the idea that a continuous field, $A(\mathbf{r})$ can be represented by a superposition of smooth bell-shaped functions, $W(|\mathbf{r}-\mathbf{r}_i|)$, (usually referred to as the smoothing function or weighting function) centered on a set of point particles, $\{\mathbf{r}_i\}$, and the gradient of the field is given by the same superposition of the gradients of the smoothing functions. A set of extensive properties, such as the particle mass, m_i, is associated with each particle, and in this case, the mass can be though of as being smoothed or smeared out by the smoothing function so that the contribution of particle i to the fluid density field, $\rho(\mathbf{r})$ is given by $\rho_i(\mathbf{r}) = m_i W(|\mathbf{r}-\mathbf{r}_i|)$. The smoothing function is normalized so that $\int W(\mathbf{r})d\mathbf{r} = 1$. Consequently, the density field is given by

$$\rho(\mathbf{r}) = \sum_i \rho_i(\mathbf{r}) = \sum_i m_i W(|\mathbf{r}-\mathbf{r}_i|). \tag{9}$$

Other intensive fields, $A(\mathbf{r})$, are given by[1]

$$A(\mathbf{r}) = \sum_i A^i(\mathbf{r}) = \sum_i a_i W(|\mathbf{r}-\mathbf{r}_i|) = \sum_i (m_i A_i / \rho_i) W(|\mathbf{r}-\mathbf{r}_i|), \tag{10}$$

and the gradient of the intensive field, ∇A, is given by

$$\nabla A(\mathbf{r}) = \sum_i (m_i A_i / \rho_i) \nabla W(|\mathbf{r}-\mathbf{r}_i|). \tag{11}$$

The SPH equations for continuous fields can also be expressed in terms of the particle number density, $n_i = \sum_i W(|\mathbf{r}-\mathbf{r}_i|)$, as

$$A(\mathbf{r}) = \sum_i A_i / n_i) W(|\mathbf{r}-\mathbf{r}_i|) \tag{12}$$

and

$$\nabla A(\mathbf{r}) = \sum_i (A_i / n_i) \nabla W(|\mathbf{r}-\mathbf{r}_i|). \tag{13}$$

The SPH equation for the flow of an inviscid fluid is based on the equation of motion

$$d\mathbf{V}/dt = -\nabla P/\rho, \tag{14}$$

where $\mathbf{V}$ is the fluid velocity and ∇P is the pressure gradient. The pressure field is obtained from the density field via the equation of state and the identity $\nabla P/\rho = \nabla(P/\rho) + (P/\rho^2)\nabla\rho$. The resulting equation of motion is

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[1] The standard SPH formulation [Gingold and Monaghan, 1977; Lacy, 1977] is based on the identity $A(\mathbf{r}) = \int A(\mathbf{r}')\delta(\mathbf{r}-\mathbf{r}')d\mathbf{r}'$, where $\mathbf{r}$ and $\mathbf{r}'$ are position vector and δ is the Kronecker delta function. If the δ function is replaced by the smoothing function $w(\mathbf{r}-\mathbf{r}')$, $A(\mathbf{r}) = \int A(\mathbf{r}')w(\mathbf{r}-\mathbf{r}')d\mathbf{r}'$, where $\int w(\mathbf{r})d\mathbf{r} = 1$, $A(\mathbf{r})$ is a smoothed version of $A(\mathbf{r}')$, and if the field A is defined in a set of points that carry masses m_i, the density field is given by $\rho(\mathbf{r}) = \sum_i m_i w(\mathbf{r}-\mathbf{r}')$ and $A(\mathbf{r}) = \sum_i (m_i / \rho_i) A_i w(\mathbf{r}-\mathbf{r}')$, which is equation (10).

$$d\mathbf{V}_i / dt = -\sum_j \left(\frac{P_i}{\rho_i^2} + \frac{P_j}{\rho_j^2} \right) \nabla W(|\mathbf{r}_i - \mathbf{r}_j|) \tag{15}$$

This is one of many possible SPH formulations of the Euler equation for inviscid fluid flow. The point masses used in SPH simulations can be thought of in terms of a moving (Lagrangian) disordered grid, and there are many ways of solving differential equations using SPH, just as there are many possible ways of formulating fluid flow equations using regular grids. In reality, the SPH equations cannot be used to simulate the Euler equation because the particles in an SPH simulation move between regions with different velocities thus creating a viscosity in the same way that a viscosity is created in molecular dynamics simulations[2].

If a body force, such as the effects of gravity acting on the fluid density is added, the equation of motion becomes

$$d\mathbf{V}_i / dt = -\sum_j \left(\frac{P_i}{\rho_i^2} + \frac{P_j}{\rho_j^2} \right) \nabla W(|\mathbf{r}_i - \mathbf{r}_j|) + \mathbf{f}_i, \tag{16}$$

and for gravity driven flows $\mathbf{f}_i = m_i\mathbf{g}$, where $\mathbf{g}$ is the gravitational acceleration.

Since the first applications of SPH to astrophysical fluid dynamics, where viscous forces do not play a significant role, it was almost 20 years until the effects of viscosity were included in SPH simulations (Takeda *et al*, 1994, Posch *et al*., 1995), apart from the addition of "artificial" viscosity to improve numerical stability. In SPH simulations of fluid mechanics, it is usual to add an SPH expression for the effects of viscosity on the fluid flow, and the equation of motion becomes [Zhu *et al*., 1977]

$$\frac{d\mathbf{V}_i}{dt} = -\sum_j m_j \left(\frac{P_i}{\rho_i^2} + \frac{P_j}{\rho_j^2} \right) \nabla W(|\mathbf{r}_i - \mathbf{r}_j|) + \sum_j \frac{m_j(\mu_i + \mu_j)(\mathbf{v}_i - \mathbf{v}_j)}{\rho_i \rho_j |\mathbf{r}_i - \mathbf{r}_j|^2}(\mathbf{r}_i - \mathbf{r}_j) \bullet \nabla_i W(|\mathbf{r}_i - \mathbf{r}_j|) + \mathbf{g}.$$

(17)

Here $\mathbf{v}_i$ is the velocity of particle i and μ_i is the fluid viscosity at particle i, (the viscosity can vary spatially in multiphase and/or multicomponent systems). The corresponding equation of motion based on the particle number density is

$$\frac{d\mathbf{V}_i}{dt} = -\sum_j \left(\frac{P_i}{n_i^2} + \frac{P_j}{n_j^2} \right) \nabla W(|\mathbf{r}_i - \mathbf{r}_j|) + \sum_j \frac{(\mu_i + \mu_j)(\mathbf{v}_i - \mathbf{v}_j)}{n_i n_j |\mathbf{r}_i - \mathbf{r}_j|^2}(\mathbf{r}_i - \mathbf{r}_j) \bullet \nabla_i W(|\mathbf{r}_i - \mathbf{r}_j|) / m_i + \mathbf{g}. \tag{18}$$

In theoretical work on SPH it is often convenient to use a Gaussian form for the smoothing function. However, in numerical investigations a variety of spline functions with a finite range, h, have been used, and the smoothing function, $W(|\mathbf{r}|)$, in the above equations may be replaced by $W(|\mathbf{r}|, h)$ to emphasize this.

To simulate fluid flow in confined systems such a porous media and fractured porous media it is convenient to use stationary particles to represent the confining solid phase with a combination of short range repulsive interaction and (relatively) long range interactions between the liquid particles and the solid particles. Bounce back boundary conditions (reversal of particle velocities) can be used to return to the fluid particles that penetrate to far into the solid, and if

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[2] SPH is isomorphic with molecular dynamics using an embedded atom potential [Hoover, 1998]. In SPH simulations, viscosity is generated in the same manner as it is in atomic fluids (with no thermally accessible internal degrees of freedom and no viscous particle-particle interactions) and the corresponding molecular dynamics models,.

this is done only a thin layer of solid particles near the solid-fluid interface is needed. A similar approach can be used to simulate multiphase fluid flow by labeling the particles to indicate which fluid component they represent and using different interaction potentials ($U_{11}, U_{12}, U_{22} \ldots$, etc. for multi-component fluids). Figure 2 illustrates a two-dimensional simulation carried out in this manner. In addition, a similar approach can be used to simulate unconfined liquid drops (single component, two phase fluids).
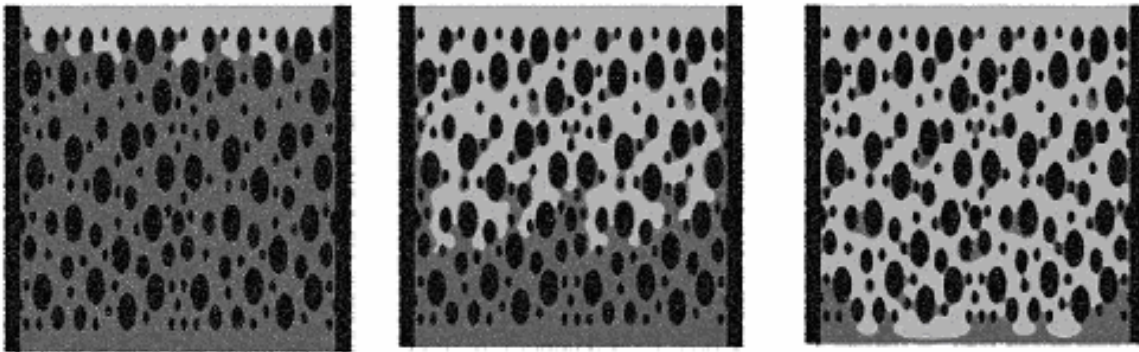

Figure 2: Three stages in a 2-dimensional simulation of the gravity driven penetration of a dense non-wetting fluid into an anisotropic porous medium

Particle methods have a number of advantages: they rigorously conserve mass; they very accurately conserve momentum; they are isotropic; they are Galilean invariant; and relatively little code development effort is required to add additional physics to the model. In addition, they exhibit qualitatively, but not generally quantitatively, correct contact line/contact angle behavior. However, particle models must be calibrated (the model interfacial energies and the fluid viscosities cannot be calculated theoretically), and their computational efficiency is low (sometimes very low) compared with continuum grid-based methods.

   The development of multiscale methods for the particle simulation of fluids is very much in its infancy [Kitsionas and Whitworth, 2002, Lastiwka *et al.*, 2005, Alimi *et al.*, 2003]. In the case of the smoothed particle hydrodynamics model, the particles can be thought of as the nodes of an unstructured Lagrangian grid, and it is natural to think of particle refinement at a direct analog to mesh refinement.

   In many applications, adaptive particle refinement will be needed, and this will involve particle fragmentation (or insertion) where increased resolution is needed and particle coalescence (or removal) where improved efficiency can be achieved without sacrificing accuracy. Particle smoothing lengths can also be changed. Clearly, particle refinement/coarsening should conform to the conservation principles and symmetries discussed above. A variety of particle – continuum hybrid methods have been developed [Flekkoy *et al.*, 2000, Nie *et al.*, 2004]. In general, there is an overlap region in which the solutions of the hybrid and continuum methods are matched. Hybrid particle methods such as MD/SPH, MD/DPD/SPH and DPD/SPH could also be used to improve computational efficiency for processes that depend on microscopic details near surfaces and interfaces.

   Fragmentation will increase the number of degrees of freedom, and it is likely that the positions and velocities of the new particles will not be uniquely defined. A similar problem is encountered in hybrid molecular-continuum models where the number of degrees of freedom

associated with the particles in a grid volume of the continuum model in the overlap region where the particle and continuum models are matched is much larger than the degrees of freedom associated with the continuum fluid in the same grid volume.

Summary

Although particle methods are less computationally efficient than grid-based continuum equation solvers, they have a number of important advantages for subsurface multiphase fluid flow and biogeochemical processes. These include rigorous mass conservation, momentum conservation, Galilean invariance and isotropy. In addition there is no need for interface tracking, realistic, but not necessarily accurate, dynamic contact angle behavior is obtained, and additional physical, chemical and biological processes can be added with relatively little code development effort. The methods developed for molecular dynamics can be used to efficiently implement other particle methods on large parallel computing systems.

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