Coupling a Fluctuating Fluid with Suspended Structures
Part I. Particle-Continuum Hybrid

Aleksandar Donev
Courant Institute, New York University
&
Berni J. Alder, Lawrence Livermore National Laboratory
Alejandro L. Garcia, San Jose State University
John B. Bell, Lawrence Berkeley National Laboratory

1This work performed in part under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

CIMS BioMathematics Seminar
December 11, 2010
Outline

1. Introduction
2. Particle Methods
3. Coarse Graining
4. Fluctuating Hydrodynamics
5. Hybrid Particle-Continuum Method
   - Brownian Bead
   - Adiabatic Piston
Flows of fluids (gases and liquids) through micro- (µm) and nano-scale (nm) structures has become technologically important, e.g., micro-fluidics, microelectromechanical systems (MEMS).

Biologically-relevant flows also occur at micro- and nano- scales.

The flows of interest often include suspended particles: colloids, polymers (e.g., DNA), blood cells, bacteria: complex fluids.

Essential distinguishing feature from “ordinary” CFD: thermal fluctuations!
Example: DNA Filtering

Fu et al., Nature Nanotechnology 2 (2007)

Consider modeling of a polymer chain in a flowing solution, for example, DNA in a micro-array.

The detailed structure of the polymer chain is usually coarse-grained to a model of spherical beads.

E.g., Kuhn segments of the chain are represented as spherical beads connected by non-linear elastic springs (FENE, worm-like, etc.)

The issue: How to coarse grain the fluid (solvent) and couple it to the suspended structures?
Introduction

Our approach: Particle/Continuum Hybrid

Figure: Hybrid method for a polymer chain.
The most direct and accurate way to simulate the interaction between the \textbf{solvent} (fluid) and \textbf{solute} (beads, chain) is to use a particle scheme for both: \textbf{Molecular Dynamics (MD)}

\[ m \ddot{r}_i = \sum_j f_{ij}(r_{ij}) \]

The stiff repulsion among beads demands small time steps, and chain-chain crossings are a problem.

Most of the computation is “wasted” on the \textit{unimportant solvent particles}!

Over longer times it is \textbf{hydrodynamics} (\textit{local momentum} and energy \textit{conservation}) and \textbf{fluctuations} (Brownian motion) that matter.

We need to coarse grain the fluid model further: Replace \textit{deterministic interactions with stochastic ones}. 
Stochastic conservative collisions of randomly chosen nearby solvent particles, as in DSMC (also related to MPCD/SRD).

Solute particles still interact with both solvent and other solute particles as hard or soft spheres [2].

No fluid structure: Viscous ideal gas.

One can introduce biased collision models to give the fluids consistent structure and a non-ideal equation of state. [3, 4].
In order to examine the time-scales involved, we focus on a fundamental problem:

A single bead of size $a$ and density $\rho'$ suspended in a stationary fluid with density $\rho$ and viscosity $\eta$ (Brownian walker).

By increasing the size of the bead obviously the number of solvent particles increases as $N \sim a^3$. But this is not the biggest problem (we have large supercomputers).

The real issue is that a wide separation of timescales occurs: The gap between the timescales of microscopic and macroscopic processes widens as the bead becomes much bigger than the solvent particles (water molecules).

Typical bead sizes are $nm$ (nano-colloids, short polymers) or $\mu m$ (colloids, DNA), while typical atomistic sizes are $1\,\text{Å} = 0.1\,\text{nm}$. 
Coarse Graining

Brownian Bead

- Classical picture for the following dissipation process: **Push a sphere suspended in a liquid with initial velocity** $V_{th} \approx \sqrt{kT/M}$, $M \approx \rho' a^3$, **and watch how the velocity decays:**

- **Sound waves** are generated from the sudden compression of the fluid and they take away a fraction of the kinetic energy during a **sonic time** $t_{sonic} \approx a/c$, where $c$ is the (adiabatic) sound speed.

- **Viscous dissipation** then takes over and slows the particle non-exponentially over a **viscous time** $t_{visc} \approx \rho a^2/\eta$, where $\eta$ is the shear viscosity. Note that the classical **Langevin time** scale $t_{Lang} \approx m/\eta a$ applies only to unrealistically dense beads!

- **Thermal fluctuations** get similarly dissipated, but their constant presence pushes the particle diffusively over a **diffusion time** $t_{diff} \approx a^2/D$, where $D \sim kT/(a\eta)$. 
The mean collision time is $t_{coll} \approx \frac{\lambda}{v_{th}} \sim \frac{\eta}{(\rho c^2)}$, where the thermal velocity is $v_{th} \approx \sqrt{\frac{kT}{m}}$, for water

$$t_{coll} \sim 10^{-15} \text{s} = 1 \text{fs}$$

The sound time

$$t_{sonic} \sim \begin{cases} 1 \text{ns for } a \sim \mu \text{m} \\ 1 \text{ps for } a \sim \text{nm} \end{cases}$$

with gap $\frac{t_{sonic}}{t_{coll}} \sim \frac{a}{\lambda} \sim 10^2 - 10^5$
Viscous time estimates

\[ t_{\text{visc}} \sim \begin{cases} 
1 \mu s & \text{for } a \sim \mu m \\
1 ps & \text{for } a \sim nm 
\end{cases} \]

, with gap \( \frac{t_{\text{visc}}}{t_{\text{sonic}}} \sim \sqrt{C} \frac{a}{\lambda} \sim 1 - 10^3 \)

Finally, the diffusion time can be estimated to be

\[ t_{\text{diff}} \sim \begin{cases} 
1 s & \text{for } a \sim \mu m \\
1 ns & \text{for } a \sim nm 
\end{cases} \]

, with gap \( \frac{t_{\text{diff}}}{t_{\text{visc}}} \sim \frac{a}{\phi R} \sim 10^3 - 10^6 \)

which can now reach macroscopic timescales!
Levels of Coarse-Graining

Figure: From Pep Español, “Statistical Mechanics of Coarse-Graining”
Formally, we consider the continuum field of conserved quantities

\[ \mathbf{U}(\mathbf{r}, t) = \begin{bmatrix} \rho \\ \mathbf{j} \\ e \end{bmatrix} \approx \tilde{\mathbf{U}}(\mathbf{r}, t) = \sum_i \begin{bmatrix} m_i \\ m_i \mathbf{v}_i \\ m_i \mathbf{v}_i^2 / 2 \end{bmatrix} \delta [\mathbf{r} - \mathbf{r}_i(t)], \]

where the symbol \( \approx \) means that \( \mathbf{U}(\mathbf{r}, t) \) approximates the true atomistic configuration \( \tilde{\mathbf{U}}(\mathbf{r}, t) \) over long length and time scales.

Formal coarse-graining of the microscopic dynamics has been performed to derive an approximate closure for the macroscopic dynamics [5].

This leads to SPDEs of Langevin type formed by postulating a random flux term in the usual Navier-Stokes-Fourier equations with magnitude determined from the fluctuation-dissipation balance condition, following Landau and Lifshitz.
Due to the *microscopic conservation* of mass, momentum and energy,

$$\partial_t U = -\nabla \cdot [F(U) - \mathcal{Z}] = -\nabla \cdot [F_H(U) - F_D(\nabla U) - B\mathcal{W}],$$

where the flux is broken into a *hyperbolic*, *diffusive*, and a *stochastic flux*.

Here $\mathcal{W}$ is spatio-temporal *white noise*, i.e., a Gaussian random field with covariance

$$\langle \mathcal{W}_i(r, t)\mathcal{W}_j^*(r, t') \rangle = (\delta_{ij}) \delta(t - t')\delta(r - r').$$

Adding stochastic fluxes to the *non-linear* NS equations produces *ill-behaved stochastic PDEs* (solution is too irregular), but we will ignore that for now...
\[ D_t \rho = - \rho \nabla \cdot \mathbf{v} \]
\[ \rho (D_t \mathbf{v}) = - \nabla P + \nabla \cdot (\eta \nabla \mathbf{v} + \Sigma) \]
\[ \rho c_p (D_t T) = D_t P + \nabla \cdot (\mu \nabla T + \Xi) + (\eta \nabla \mathbf{v} + \Sigma) : \nabla \mathbf{v}, \]

where the variables are the \textbf{density} \( \rho \), \textbf{velocity} \( \mathbf{v} \), and \textbf{temperature} \( T \) fields,

\[ D_t \square = \partial_t \square + \mathbf{v} \cdot \nabla (\square) \]
\[ \nabla \mathbf{v} = (\nabla \mathbf{v} + \nabla \mathbf{v}^T) - 2 (\nabla \cdot \mathbf{v}) \mathbf{I} / 3 \]

and capital Greek letters denote stochastic fluxes:

\[ \Sigma = \sqrt{2 \eta k_B T} \mathbf{W} \]
\[ \langle \mathcal{W}_{ij}(\mathbf{r}, t) \mathcal{W}_{kl}^*(\mathbf{r}', t') \rangle = (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - 2 \delta_{ij} \delta_{kl} / 3) \delta(t - t') \delta(\mathbf{r} - \mathbf{r}') \]
Ignoring density and temperature fluctuations, we obtain the incompressible approximation:

\[ \rho D_t \mathbf{v} = \eta \nabla^2 \mathbf{v} - \nabla \pi + \sqrt{2\eta k_B T} (\nabla \cdot \mathbf{W}), \]
\[ \nabla \cdot \mathbf{v} = 0 \]

where the stochastic stress tensor \( \mathbf{W} \) is a white-noise random Gaussian tensor field with covariance

\[ \langle \mathbf{W}_{ij}(r, t) \mathbf{W}_{kl}^*(r', t') \rangle = (\delta_{ik} \delta_{jl}) \delta(t - t') \delta(r - r'). \]

We have algorithms and codes to solve the compressible equations, and we are now working on the incompressible ones.

Solving them numerically requires paying attention to discrete fluctuation-dissipation balance, in addition to the usual deterministic difficulties [6].
Split the domain into a particle and a continuum (hydro) subdomains, with timesteps $\Delta t_H = K \Delta t_P$.

Hydro solver is a simple explicit (fluctuating) compressible LLNS code and is not aware of particle patch.

The method is based on Adaptive Mesh and Algorithm Refinement (AMAR) methodology for conservation laws and ensures strict conservation of mass, momentum, and energy.
Hybrid Particle-Continuum Method

Continuum-Particle Coupling

- Each macro (hydro) cell is either particle or continuum. There is also a reservoir region surrounding the particle subdomain.
- The coupling is roughly of the state-flux form:
  - The continuum solver provides state boundary conditions for the particle subdomain via reservoir particles.
  - The particle subdomain provides flux boundary conditions for the continuum subdomain.
- The fluctuating hydro solver is oblivious to the particle region: Any conservative explicit finite-volume scheme can trivially be substituted.
- The coupling is greatly simplified because the particle fluid is ideal (no internal structure): No overlap region.

Steps of the coupling algorithm [7]:

1. The hydro solution is computed everywhere, including the **particle patch**, giving an estimated total flux $\Phi_H$.

2. **Reservoir particles** are *inserted* at the boundary of the particle patch based on *Chapman-Enskog distribution* from kinetic theory, accounting for *both* collisional and kinetic viscosities.

3. Reservoir particles are *propagated* by $\Delta t$ and *collisions* are processed (including virtual particles!), giving the total particle flux $\Phi_p$.

4. The hydro solution is overwritten in the particle patch based on the particle state $u_p$.

5. The hydro solution is corrected based on the more accurate flux, $u_H \leftarrow u_H - \Phi_H + \Phi_p$. 

We investigate the **velocity autocorrelation function** (VACF) for a Brownian bead

\[ C(t) = \langle \mathbf{v}(t_0) \cdot \mathbf{v}(t_0 + t) \rangle \]

From equipartition theorem \( C(0) = kT/M \).

For a **neutrally-boyant** particle, \( \rho' = \rho \), incompressible hydrodynamic theory gives \( C(0) = 2kT/3M \) because the momentum correlations decay instantly due to sound waves.

Hydrodynamic persistence (conservation) gives a **long-time power-law tail** \( C(t) \sim (kT/M)(t/t_{\text{visc}})^{-3/2} \) not reproduced in Brownian dynamics.
Small Bead (~10 particles)

\[ \frac{M C(t)}{k_B T} \]

- Stoch. hybrid (L=1)
- Det. hybrid (L=1)
- Stoch. hybrid (L=2)
- Det. hybrid (L=2)
- Particle (L=1)
- Theory

\[ \frac{t c_s}{R} \]

\[ \frac{t}{t_{visc}} \]

\[ t_{L=1} \]
Hybrid Particle-Continuum Method

Large Bead (~1000 particles)

\[ \frac{MC(t)}{k_B T} \]

- Stoch. hybrid (L=2)
- Det. hybrid (L=2)
- Stoch. hybrid (L=3)
- Det. hybrid (L=3)
- Particle (L=2)
- Theory

AX. Donev (CIMS)

Dec 2010 28 / 32
The adiabatic piston problem
Figure: Massive rigid piston ($M/m = 4000$) not in mechanical equilibrium: The deterministic hybrid gives the wrong answer!
Figure: The VACF for a rigid piston of mass $M/m = 1000$ at thermal equilibrium: Increasing the width of the particle region does not help: One must include the thermal fluctuations in the continuum solver!
Tethered DNA Dynamics in Shear Flow.

Stochastic Event-Driven Molecular Dynamics.

Stochastic Hard-Sphere Dynamics for Hydrodynamics of Non-Ideal Fluids.

A Thermodynamically-Consistent Non-Ideal Stochastic Hard-Sphere Fluid.

P. Español.
Stochastic differential equations for non-linear hydrodynamics.

On the Accuracy of Explicit Finite-Volume Schemes for Fluctuating Hydrodynamics.

A hybrid particle-continuum method for hydrodynamics of complex fluids.