Hybrid molecular dynamics and Navier-Stokes method in complex nanoflow geometries

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ABSTRACT

We present a new hybrid molecular dynamics and continuum solver for modelling nanoflows in arbitrary domain geometries. The method makes use of a domain decomposition, in which the molecular sub-domain is applied adjacent to interfaces or walls, and is coupled to a continuum sub-domain in the rest of the flow field in order to reduce the computational cost of full MD simulations. Coupling between disparate sub-domains employs an alternating time framework, and consists of mutual two-direction (i.e. continuum-to-molecular and molecular-to-continuum) boundary conditions. We demonstrate the hybrid technique in shear flow configurations.

Keywords: Nanofluidics, Molecular Dynamics, Hybrid Continuum-Molecular, Boundary Conditions

1 INTRODUCTION

Nanofluidics is an emerging technology of the 21st century, and molecular dynamics (MD) is perhaps the most important numerical method available to capture liquid phenomena at small scales. MD is, however, far too computationally intensive, limiting it to only small systems and relatively short time-scales. Hybrid methods for liquids [1]–[4] have recently been proposed in order to alleviate the overwhelming computational cost of full MD simulations. This is achieved by applying MD solely in small regions of the domain where it is required, and coupling it with a computationally-fast continuum formulation for the bulk of the flow field. In the literature, only relatively simple systems in cubic domains have been treated using hybrid methods, and periodic boundary conditions in MD still pose many limitations to the development of new coupling strategies. In order to study nanoflows in more engineering-related problems both MD and hybrid MD-continuum models need to be applicable in arbitrary geometries, treat boundaries as non-periodic entities, and handle parallel processing [5]–[7].

2 METHOD

We consider a domain of arbitrary geometry, defined by an unstructured polyhedral mesh. The domain is segmented into sub-meshes assigned to Computational Fluid Dynamics (CFD) and MD solvers[1], allowing an overlap region at all CFD-MD interfaces, in which coupling between disparate sub-domains can occur at regular time intervals of the simulation. An example of the hybrid domain decomposition methodology for a shear flow with a complex coupling region is portrayed in Figure 1. The MD fluid occupies its sub-domain and consists of "molecules" that interact through a pair-wise potential $U(r_{ij})$, where $r_{ij} = |\mathbf{r}_{ij}|$ and $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ is the separation vector between a pair of molecules (i, j).

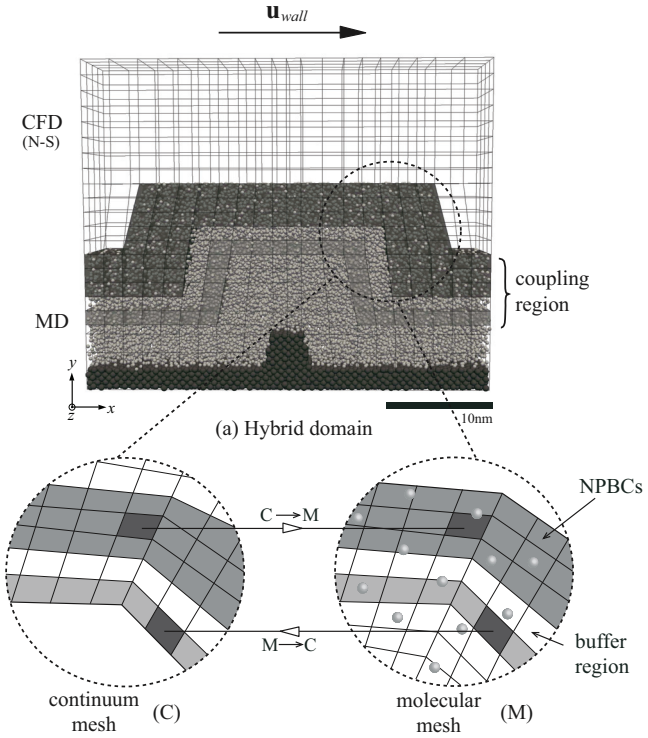

Figure 1: (a) Hybrid domain decomposition of a shear flow with a complex fixed-wall topology. (b) Schematic of the coupling region between continuum and molecular sub-domain meshes.

[1]The continuum and molecular solvers are both implemented in OpenFOAM, an open-source toolbox of C++ libraries for computational physics. It may be downloaded freely from [8].

The standard shifted Lennard-Jones (LJ) 12-6 potential [9] is used in this paper. The positions $\mathbf{r}_i$ and velocities $\mathbf{v}_i$ of molecules evolve according to standard Newtonian dynamics, using a molecular time-integration step $\Delta t_m = 10$ fs.

The continuum sub-domain uses a finite-volume discretisation of the compressible, isothermal Navier-Stokes equations, $\partial\phi/\partial t = -\nabla \cdot \mathbf{J}_\phi$, where $\phi = \{\rho, \rho\mathbf{u}\}$ are the densities, while $\mathbf{J}_\phi = \{\rho\mathbf{u}, \rho\mathbf{u}+\mathbf{\Pi}\}$ are the flux-densities; $\mathbf{\Pi} = p\mathbf{1} + \tau$ is the stress tensor, including the normal pressure and the viscous shear-stress. The continuum time-integration step is given by $\Delta t_c = 0.2$ ps.

The coupling procedure applies M→C and C→M boundary conditions via an arbitrary pair of coupled cells within the overlapping region, as shown in the two highlighted regions of Figure 1(b). C→M coupling is achieved through non-periodic boundary conditions (NPBCs): mainly the imposition of feedback loop controllers [7] that operate to converge the continuum-derived state properties (e.g., density, velocity and temperature). M→C coupling is achieved by coarse-graining of properties from cell-occupant molecules, and replacing the corresponding boundary values on the continuum fields. Properties transferred between continuum and molecular subdomains employ a relaxation technique in order to improve stability of the coupling scheme. For example, the target MD fields for an arbitrary state property ϕ is:

$$\phi_m^{(n)} = \theta\phi_m^{(n-1)} + (1-\theta)\phi_c^{(n)}, \qquad (1)$$

where θ $(0 < \theta < 1)$ is the relaxation parameter, subscripts (m, c) denote molecular and continuum fields respectively, and n denotes the time index of coupling.

A coupling time-framework is used to advance the MD and CFD in a sequential manner by a common coupling time-interval $\Delta t_{coupling} = \tau_C \Delta t_c = \tau_M \Delta t_m = 40$ ps:

1. Advance continuum solution $t \rightarrow t + \Delta t_{coupling}$ by $\tau_C = 200$ time-steps. MD waits.
2. Apply C→M BCs.
3. Advance MD $t \rightarrow t + \Delta t_{coupling}$ by $\tau_M = 4000$ time-steps. CFD waits.
4. Apply M→C BCs.
5. Repeat the dual time-marching scheme until the end of the simulation.

3 RESULTS

In order to test the hybrid method we investigate flows of liquid argon molecules, which have a characteristic length scale of $\sigma = 0.34$ nm, a characteristic energy of $\epsilon = 120k_b = 1.65678 \times 10^{-21}$ J, where k_b is the Boltzmann constant, and a mass $m = 6.69 \times 10^{-26}$ kg. A cut-off radius of $r_{cut} = 2.5\sigma = 0.85$ nm is used. We present our results in reduced units: time, $t^* = t\sqrt{\epsilon/m\sigma^2}$, number density $\rho^* = \rho\sigma^3$, temperature $T^* = T(k_b/\epsilon)$ and velocity $\mathbf{u}^* = \mathbf{u}\sqrt{m/\epsilon}$.

3.1 Simple shear flows

We validate the hybrid method by simulating a start-up Couette flow configuration and a simple oscillating flow. For start-up Couette flow, we consider a domain of dimensions $(L_x = 188\sigma, L_y = 20\sigma, L_z = 20\sigma)$ with two moving molecular walls at the extremities of the domain. We apply MD at the two wall regions (50σ thickness), and the compressible Navier-Stokes equations in the central part of the domain. Two independent coupling regions are therefore required at the continuum-molecular interfaces, while periodic boundary conditions are applied in the $y-$ and $z-$directions. The shear viscosity in the entire continuum field is chosen to match closely that in the coupling region of the MD domain, i.e., $\eta = 0.899(\sqrt{\epsilon m}/\sigma^2)$, where the bulk state of the fluid is $\rho^* = 0.6$, and $T^* = 2.4$ for number density and temperature, respectively. The walls (A, B) are accelerated from rest using a ramp velocity function over a time-period $\Delta t^* = 20$, until a maximum velocity $\mathbf{u}_A^* = (0.0, -2.5, 0.0)$ and $\mathbf{u}_B^* = (0.0, 2.5, 0.0)$ are reached. The walls are then maintained at uniform velocity. The transient velocity profiles for both hybrid and full MD simulations are shown in Figure 2, where generally good agreement is observed.

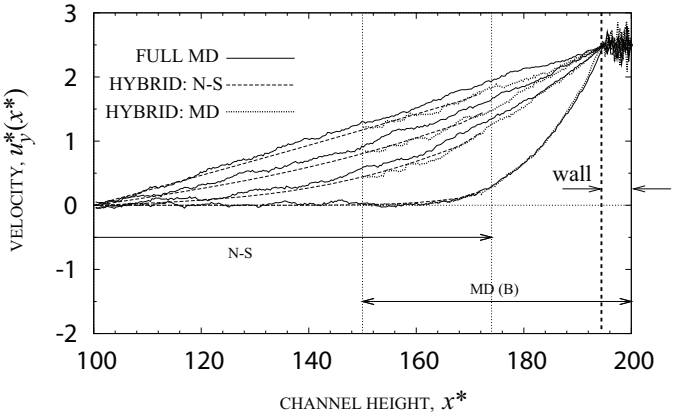

Figure 2: Transient half-channel velocity profiles for the start-up Couette flow problem, with relaxation $\theta = 0.6$. Profiles are shown for times: $t^* = \{100, 500, 1000, 4000.\}$. Hybrid solutions are compared with the full molecular solution.

A hybrid simulation of an oscillating shear flow problem is then carried out to demonstrate that the hybrid

algorithm may also be applied to unsteady cases. The hybrid domain decomposition consists of an MD subdomain encompassing the oscillating molecular wall and adjacent layer of molecular liquid, coupled to a Navier-Stokes continuum solver with no-slip boundary conditions applied at the other end of the domain (i.e. at $L_x = 100\sigma$). The molecular wall moves with an oscillating motion $\mathbf{u}_{wall}(t) = \mathbf{u}_{max}\sin(2\pi t)/\tau_T$, where $\mathbf{u}^*_{max} = (0, 10.0, 0)$ is the maximum velocity, and $\tau^*_T = 200$ is the time-period of the oscillatory wave. In Figure 3 we show velocity profiles at different times in one oscillatory cycle, and see that good agreement is achieved between the hybrid and a full-MD case. For both start-up and oscillating shear flow cases, a computational speed-up of approximately two times is achieved using the hybrid method, although the MD component still takes up the majority of the cost of the entire simulation.

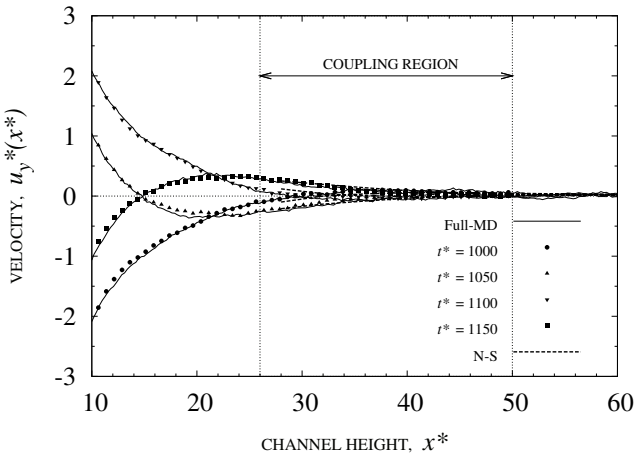

Figure 3: Velocity profiles shown only in the coupling region for the simple oscillating shear flow case. Results are only displayed for one cycle ($\tau^*_T = 200$) of the oscillatory motion. Comparisons are made with a full MD simulation.

3.2 Complex coupling region

We now demonstrate the capability of our hybrid method by applying it to a Couette flow problem with a complex-shaped coupling region, as shown in Figure 1. The bottom stationary wall and adjacent layer of liquid is modelled by MD, while the rest of the domain and upper moving wall is simulated by CFD. A moving wall velocity of $\mathbf{u}^*_{wall} = (0.5, 0, 0)$, with a no-slip boundary condition, is applied to the top boundary of the domain, and cyclic boundary conditions are applied in the other two directions. At the molecular-continuum interface, a 3D overlap region is present for velocity and density coupling between the CFD and MD formulations.

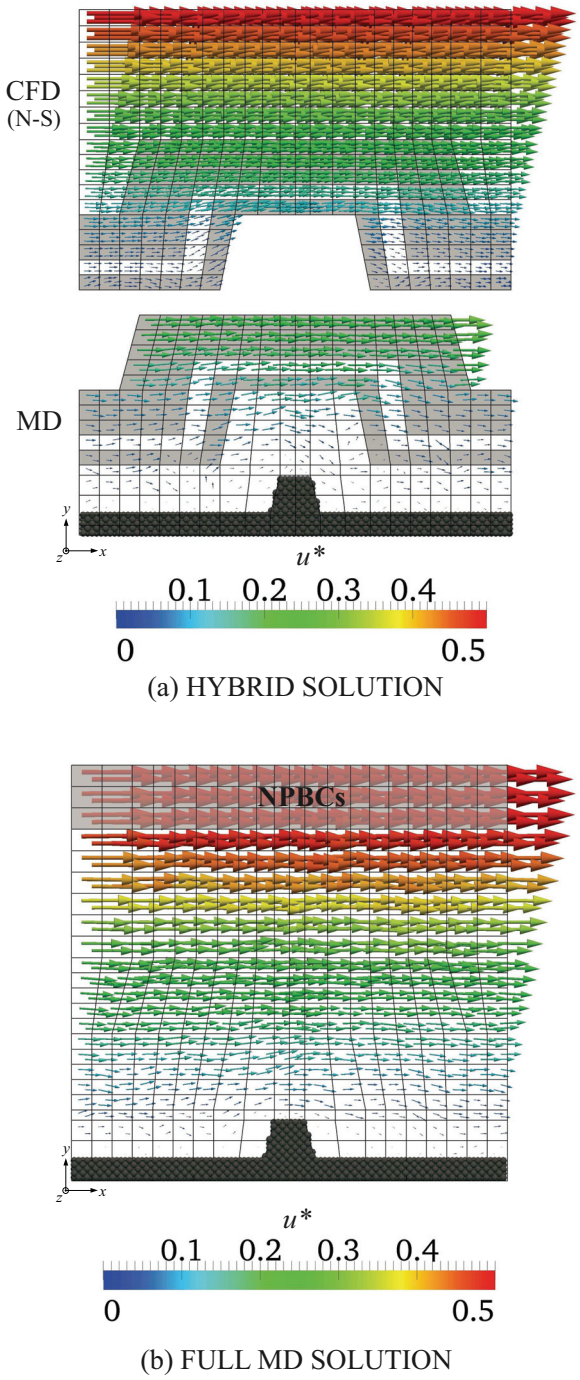

Figure 4: Velocity vector fields: (a) hybrid (top – N-S solution; bottom – MD solution) and (b) full-MD simulations of the same complex Couette flow case.

Results are shown in Figures 4 and 5, where general agreement of the velocity field is observed between the full-MD and hybrid MD-CFD simulations. However, the hybrid simulation is approximately two times faster than the full-MD simulation. Speed-up may also be estimated from the term $N_{mol}^{\mathrm{FM}}/N_{mol}^{\mathrm{H}}$, where $N_{mol}^{\mathrm{H}} = 34,394$ and $N_{mol}^{\mathrm{FM}} = 68,944$ are the average number of molecules in the hybrid MD sub-domain and the full-MD domain respectively.

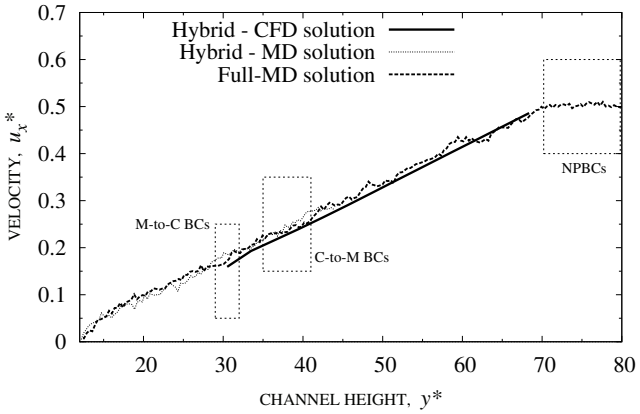

Figure 5: Comparisons of the velocity profile in the y-direction starting from the wall protrusion seen in Figure 4.

4 CONCLUSIONS

We have presented a new hybrid MD-CFD method based on spatial domain decomposition and an alternating time-coupling framework with relaxation field coupling. Our method is tested for shear-flow applications, and shows generally good agreement between the hybrid and full-MD solutions, with more than twice the speed-up. Our hybrid method is parallelised and generally applicable to any complex domain configuration, and so may serve as a very useful tool in engineering design and simulation of nano-scale applications.

5 ACKNOWLEDGMENTS

This work is funded in the UK by the James Weir Foundation (MKB), the Institution of Mechanical Engineers (WDN) and the Royal Society of Edinburgh / Scottish Government (JMR).

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