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A CONTINUUM - MOLECULAR DYNAMICS HYBRID METHOD FOR MICRO- AND NANO-FLUID FLOW

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ABSTRACT
A computer program based on a Molecular Dynamics-Continuum hybrid numerical method has been developed in which the Navier-Stokes equations are solved in the continuum region and the atomistic molecular dynamics in molecular region. The prepared algorithm and the computer code are capable of computing flows in micro and nano-scale geometries. The coupling between the continuum equations and the molecular dynamics is constructed through constrained dynamics within an overlap region where both molecular and continuum equations are solved simultaneously. An Overlap region is introduced in two directions to improve the choice of using molecular region in smaller areas. The proposed method is used to simulate steady and start-up Couette flow showing quantitative agreement with results from analytical solutions and full molecular dynamics simulations.

KEYWORDS:
Hybrid Method, Molecular Dynamics Simulation, Couette flow, Nanochannel, Microfluidics

INTRODUCTION
Continuity is the most fundamental assumption in Continuity is the most fundamental assumption in macroscopic fluid mechanics which is governed by the Navier–Stokes (NS) equations (see, for example, [1]). This assumption breaks down as the spatial scale of flows approaches the molecular mean free path [2]. In particular, the molecular mean free path may no longer be negligible and macroscopic constitutive relations and boundary conditions become inadequate. In principle, this problem can be resolved by using a fully atomistic description such as molecular dynamics (MD) simulations. However, MD simulations on current computers are typically limited to dimensions less than 100 nm and times much shorter than a microsecond ([3,4,5,6,7]). Thus they can not treat most systems of experimental interest[8]. Moreover, in most cases the breakdown of the continuum description is confined to limited domains, such as fluid– fluid or fluid–solid interfaces. Hence it is desirable to develop hybrid methods that combine continuum fluid dynamics and molecular dynamics, using the most efficient description in each region of space [9].

In recent years, a new class of hybrid approaches has been developed that combines the strengths of atomistic and continuum approaches. The first hybrid method was proposed by O'Connell and Thompson (1995) which combined MD simulation with continuum solver [10].The so-called hybrid method was widely used in simulating channel flows of larger scale but with microscopic view of interfacial phenomena. This class of approaches is a multiscale simulation combining continuum and particle (atomistic) models so as to extend the computational domain to a range beyond that in the pure atomistic simulation. Thereby the wall-fluid interaction in near-wall thin regions can be resolved by atomistic results, while the bulk flow in the core region at a sufficient distance from the wall is computed using the continuum model based on Navier–Stokes (N–S) equations. In hybrid computations, there are two kinds of interface. One is the physical interface where the fluids contact the solid walls. Another one is between the particle and the continuum regions, where an overlap region is arranged. In the overlap region, exchange of the computational results from

particle to continuum (P→C) as well as from continuum to particle (C→P) regions needs a particular care [8].

In all previous hybrid studies of flow in nanochannels, the overlap region, that combines the continuum and molecular dynamics solutions, were spread out in only one direction. Therefore, investigation of flow behavior with MD simulation over a tiny portion of wall arouses a necessity to use atomistic description for the entire region near wall. However, by considering the overlap region in two directions, selection of smaller region near the wall will be available and the simulation can focus on the interested areas.

In the current paper, a hybrid numerical method was developed. In this method interfacial regions were described by molecular dynamics simulations and bulk regions by the incompressible Navier-Stokes (NS) equations. The method was applied to a simple steady as well as start-up Couette flow and the obtained results have been shown quantitative agreement with the results from analytical solutions. The pure atomistic simulation is solved in small region near the wall. In addition, extending the overlap region to two dimensions and adding complexity to its governing equations enabled one to select longer channels and still solving the interested area with pure MD simulation.

SIMULATION METHOD

The simulation domain is divided into two spatial domains shown in figure 1, the particle (atomistic) domain is indicated by dots and the continuum domain is represented by shading. Molecular dynamics simulations are performed in the atomistic domain, and classical continuum fluid dynamics equations are solved on a grid in the continuum domain. The overlap region is constructed in molecular region and extends typically a few times the continuum grid spacing. Both molecular dynamics and continuum calculations are performed in this overlap region.

As shown in figure 2, the geometry is a 3D channel surrounded by solid planar wall. The top and bottom walls are constructed in y-x plane and the side walls are in z-x plane. Periodic boundary condition is applied on the inlet and outlet of the channel in y-z plane and x-direction. The MD region is a slit shaped domain, constructed on the center of the bottom wall and spread from inlet to the outlet of the channel, and measures 4Δy and 5Δy in y and z directions respectively. The bottom wall behaves as atomistic wall when MD equations are solved and acts as a barrier with no-slip condition when NS equations are solved.

In the MD simulation, fluid atoms are allowed to interact via Lennard-Jones 6-12 potential. The molecular interaction potential between both wall-fluid and fluid-fluid is given by a shifted Lennard–Jones potential as follows:

$$U^{LJ}(r)=\begin{cases}4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12}-\left(\frac{\sigma}{r}\right)^{6}-\left(\frac{\sigma}{r_c}\right)^{12}+\left(\frac{\sigma}{r_c}\right)^{6}\right] & r\le r_c\\ 0 & r>r_c\end{cases} \qquad (1)$$

where σ is molecular diameter and ε is the well depth of potential function.

Using Newton's second law the molecular dynamics equation of motion is found from:

$$m_i\ddot{\boldsymbol{r}}_i=\boldsymbol{f}_i \qquad \boldsymbol{f}_i=-\frac{\partial}{\partial \boldsymbol{r}_i}U^{LJ} \qquad (2)$$

where U^{LJ} is established from (1).

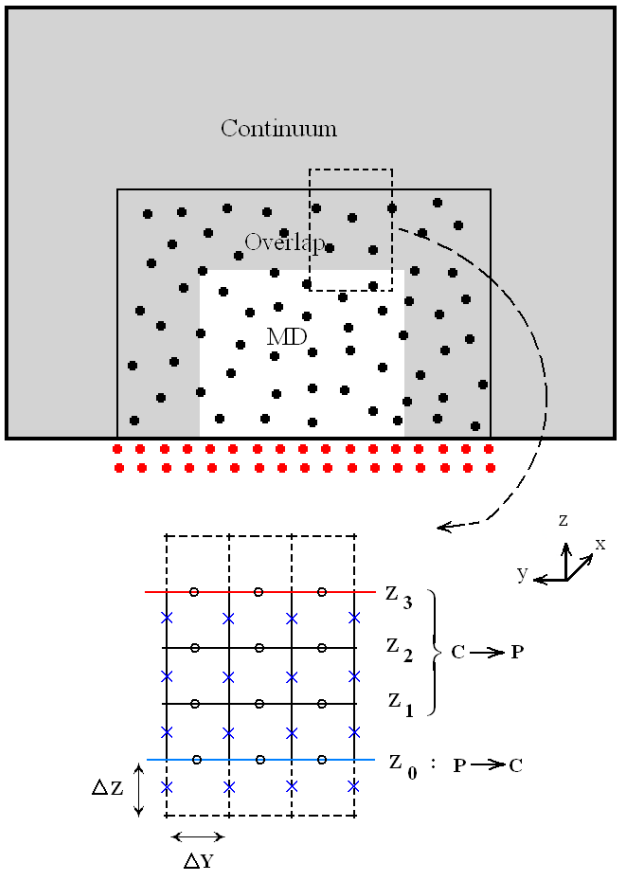

Figure 1- locations of Overlap, Continuum and MD in the simulation geometry and the discretized computational space

To reduce the computational cost, the interaction is set to zero when atoms are distanced more than the cut-off length r_c = 2.5σ. Mass of atoms were set to m and the density is set to ρ= 0.81mσ^{-3}, to allow comparison between previous results [8,9,11]. The atomic wall consists of atoms, with mass and density the same as fluid and forming two (001) planes of an FCC crystal. Each wall atoms is attached to its lattice site by a harmonic spring of constant k=400ε/σ^2 to maintain a well-defined solid structure with a minimum number of solid atoms. The spring constant k controls the thermal roughness of the wall as well as its responsiveness to the fluid and is adjusted so that the mean squared displacement of each atom about its lattice site is less than the Lindemann [12] criterion for melting. For the results shown $<\delta u_2>/d_2$=0.01 is used.

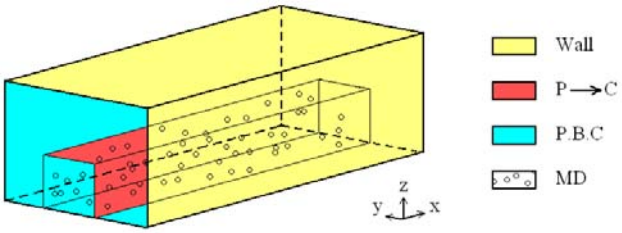

Figure 2- Simulation Geometry and boundary conditions schematic

Fluid and solid atoms also interact with a Lennard- Jones potential, but with a reduced binding energy ε_{wf} = 4.0ε which leads to a no-slip boundary condition. By implementing thermal wall model, momentum and energy exchanges between fluid and the wall take place properly. Therefore, excessive heat in the fluid is transferred to the walls and then, the heat is dissipated through the thermostat applied on the walls. Nose-Hoover chain of second order [13] was applied to the separate layers of the wall atoms to hold the wall temperature constant at $1.1\varepsilon/k_B$. The equations of motion are integrated using the Gear predictor-corrector scheme [14] of fifth order with time step $\Delta t_{MD} = 0.005\tau$, where $\tau \equiv (m\sigma^2/\varepsilon)^{1/2}$ is the characteristic time of MD equations of motion.

In the continuum region, the following three-dimensional incompressible Navier–Stokes equations are solved.

$$\nabla . \boldsymbol{u} = 0$$

$$\frac{\partial \boldsymbol{u}}{\partial t} + \boldsymbol{u}.\nabla \boldsymbol{u} = -\frac{1}{\rho}\nabla P + \frac{\mu}{\rho}\nabla^2 \boldsymbol{u} \qquad (3)$$

*where **u** is the fluid velocity, μ is the dynamic viscosity and p is the pressure. The transport coefficients are determined through preliminary MD simulations to be consistent with the atomistic potential in order to ensure continuity in the overlap region and therefore the dynamic viscosity is selected as $\mu = 2.14\varepsilon\tau\sigma^{-3}$. The above equations* ***Error! Reference source not found.*** *are solved numerically using the SIMPLE [15] method. The Navier-Stokes equations are integrated with mesh size of $\Delta x = \Delta y = 5.21\sigma$ and $\Delta z = 4.82\sigma$ on staggered grid [16] as shown in figure 1. Pressure is defined at the centers of cells and x and y components of the velocity are marked out at the middle of the vertical (crosses) and horizontal (circles) edges of the cells, respectively. The time scale on which atoms sample different kinetic energies is of order the velocity autocorrelation time t_{vv}= 0.14s [17]. Therefore, the simulations used $\Delta t_{FD} = 50\Delta t_{MD} = 0.25\tau$, where Δt_{FD} and Δt_{MD} are the time steps for continuum and atomistic regions respectively.*

The key element in hybrid methods is the scheme used for coupling macroscopic and microscopic solutions to ensure the consistency of mass, energy and momentum in the overlap region and the continuity of fluxes across it. In transferring information from particle region to the continuum, a (Particle to Continuum) $P \rightarrow C$ condition is introduced in which continuum velocities are extracted from the coarse-grained MD velocity. On the other hand, the (Continuum to Particle) $C \rightarrow P$ condition is more complicated because the information about macroscopic quantities does not uniquely specify the microscopic state of particles and thus, a sum of various actions builds this condition. The goal of continuum to particle coupling is to constrain the mean particle behavior without introducing unphysical artifacts associated with the constraint or the termination of the particle domain. The overlap region helps to minimize these effects. For construction of $P \rightarrow C$ condition, MD solution obtains velocity values by averaging the velocities of all MD particles within a volume of dimensions $\Delta x \times \Delta y \times \Delta z$ that is centered on the point of interest. The average is also performed over a time interval $\Delta t_{FD} = 0.25\tau$ that is centered on the time for the continuum equations. This means that in transferring the data from the continuum region to the MD region, information is only available for times $\Delta t_{FD}/2$ earlier than the current MD time. In the overlap region, a constrained dynamics is implemented for atoms equations of motion in order to achieve the mass and momentum consistencies between the molecular and continuum descriptions. The momentum consistence implies that the local mean of the particle momentum is equal to the instantaneous macroscopic momentum for arbitrary cells in the overlap region:

$$\frac{1}{N_J}\sum_{i}^{N_J} \boldsymbol{v}_i = \boldsymbol{u}_J(t) \qquad (4)$$

Where N_J is the number of particles in cell J. Therefore, the mean particle velocity in each cell J is constrained to follow the continuum solution u_J. Equation (4) introduces a nonholonomic constraint to equations of motion of the particles in the overlap region. There are various ways to modify the MD equations of motion which has its own advantage and disadvantages. Kane`s method, Lagrange multiplier method and Gauss's principle of least constraint are the methods that can be utilized to implement a nonholonomic constrain to equations of motion. By implementing Lagrange multiplier and Kane's methods a very accurate formulation, which can predict the true trajectories of the particles can be derived [18]. But the shortcoming of both methods is the complexity of the derived relations which made it almost impractical to implement on MD equations of motion. In this paper, according to [8,9,19] the following modified equation for the ith particle is found by using Gauss's principle of least constraint:

$$\ddot{\boldsymbol{x}}_i = \frac{\boldsymbol{F}_i}{m} + \xi\left(-\frac{1}{N_J m}\sum_{k=1}^{N_J}\boldsymbol{F}_k + \frac{D\boldsymbol{u}_J(t)}{Dt}\right) \qquad (5)$$

where $D\boldsymbol{u}_J/Dt$ indicates the material derivative. The last two terms ensure that the mean particle velocity tracks the continuum solution by subtracting the sum of all forces on particles in the cell and adding the material derivative of the continuum solution [8]. ξ is a characteristic time constant which relaxes the continuum equation on MD equation. The relaxation is found necessary for the current 3D geometry to decrease the unsatisfactory fluctuation induced by the MD simulation. Various methods have been developed recently to

find an appropriate relation for modifying the relaxation factor [11,20]. In this paper, an arithmetic progression is chosen to find a relation for increasing the relaxation factor so that the atomistic solution relaxes on continuum solution with a finite time lag in the very beginning of the simulation. By using the proposed relation, the relaxation factors starts from 0.1 and tends towards unity as the simulation progresses. The relaxation factor relation is shown in the figure 3.

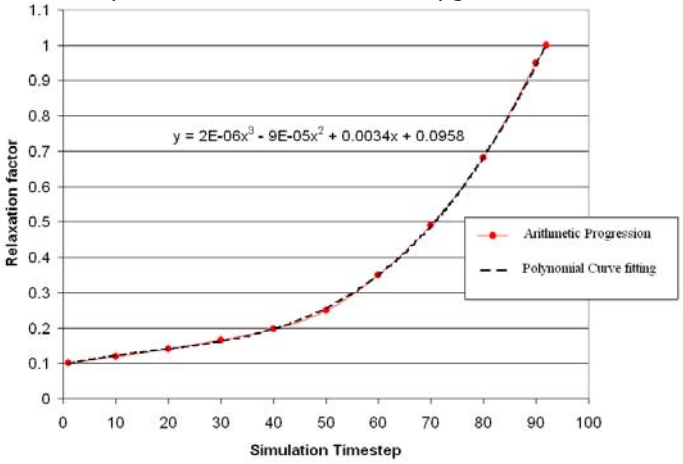

Figure 3- Variation of the relaxation factor with simulation timestep. The relaxation factor increases from 0.1 to 1 following the above curve. A polynomial of order 3 is fitted to the mentioned progression which is shown in the figure

To prevent molecules from freely drifting away from the MD simulation domain, an external force is applied to particles in the last layer of the overlap region. A similar force is also applied in the y direction. For the particles between Z_2 and Z_3 this equation is as follow: [9]

$$F_Z = -\alpha p\sigma \frac{(z - z_2)}{1 - \frac{z - z_2}{z_3 - z_2}} \qquad (6)$$

where p_0 is the equilibrium pressure corresponding to the fluid density and α is a constant of order one. In the current simulation α was set to unit and the hybrid simulation is insensitive to the magnitude of α when it is of unit order.

Mass continuity can be achieved by moving particles across the boundary of the overlap region. The number of particles in each cell is changed by the net flux in an interval Δt_{FD}. The mass flux is evaluated by the NS equations and the number of particles that should be inserted or removed across the boundary is evaluated from:

$$n = -A_\perp \rho u_\perp \Delta t_{FD} / m \qquad (7)$$

where $A_\perp$ is the area of the cell perpendicular to the interface. If n is negative, the n particles closest to the outer layer of the overlap region are removed. If n is positive, particles are inserted at regular intervals over the subsequent Δt_{FD} at positions near the outer layer of the overlap region and randomly distributed along x and y direction in each cell. To prevent the distance between the inserted particles and the previous ones from being too small, the above process is repeated. At each interval Δt_{FD}, the nearest integer of n is taken and the remaining particle fraction is included at the next time step. [11]

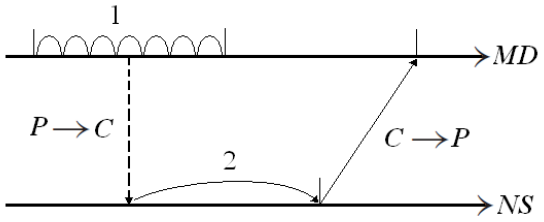

Figure 4- Time coupling in the Hybrid method showing the C→P and P→C condition. MD is the molecular region timestep and NS is the continuum region timestep

The time coupling scheme in the overlap region is shown in Figure 4. First, 50 time steps in the MD are conducted, then the average is performed over the time interval in a box of Δx×Δy×Δy around the interested point and the averaged velocities are transferred to the NS equations using P→C condition at the center point of the interval; then, the NS equations are advanced for one time step and the information is sent to the MD using the C→P condition, where the extrapolation of velocity field is needed. The MD and NS equations are shown in the figure with 1 and 2, respectively.

RESULT AND DISCUSSION

Steady Couette flow with no-slip boundary condition is first simulated in a nanochannel using presented hybrid model. Argon atoms are confined between four parallel walls in a 3D geometry as shown in figure 2. Channel measures 53.6σ, 36σ and 23.04σ in x, y and z direction respectively. The MD region is in the middle of the channel adjacent to the lower wall and is comprised of 2780 fluid atom and 1950 wall atoms. The overlap region is spread from Z_0 plane to Z_3 plane in the Z direction and includes three layers of computational cells as shown in figure 1.

The temperature of the fluid is kept constant at $T = 1.1\varepsilon/k_B$, by using second order Nose-Hoover chain thermostat and at these given T, the fluid is in a well-defined liquid phase with viscosity $\mu = 2.14\ \varepsilon\tau\sigma^{-3}$. Top wall is moving in the x direction with $u_w = 10\sigma/\tau$ while the lower wall is kept still. The computational domain is divided into bins of the same dimensions as computational grid. To observe the accuracy of the presented model velocity profile is obtained in the x-direction and compared to the results obtained from simulating the whole domain with pure atomistic molecular dynamic simulation. As illustrated in figure 5, the stream direction velocity profile is computed for different layers of fluid from the lower layer (layer 1), adjacent to the still wall; to the upper one (layer 16) adjacent to the moving wall. The hybrid and the MD solutions are shown with continuous line and discrete dots respectively. The results showed a good agreement between the predicted velocity profile from both hybrid and MD simulations in a nanochannel steady flow.

Because the current geometry is three dimensional and the overlap region is spread in two directions, for reducing the unsatisfactory fluctuations induced by the MD solution, choosing a proper relaxation factor becomes very important. Therefore, unsteady sudden start-up Couette flow is investigated in the above geometry as another example. The velocity profiles are obtained for two different times and are compared with the analytical solution in figure 6. According to [21] velocity profile at the outlet plane for a Couette flow can be found from:

$$\frac{u}{U_0}=\left(1-\frac{y}{h}\right)-\frac{2}{\pi}\sum_{n=1}^{\infty}\frac{1}{n}exp\left(-n^2\pi^2t^*\right)sin\left(\frac{n\pi y}{h}\right) \qquad (8)$$

Where h is the channel height and U_0 is the wall velocity.

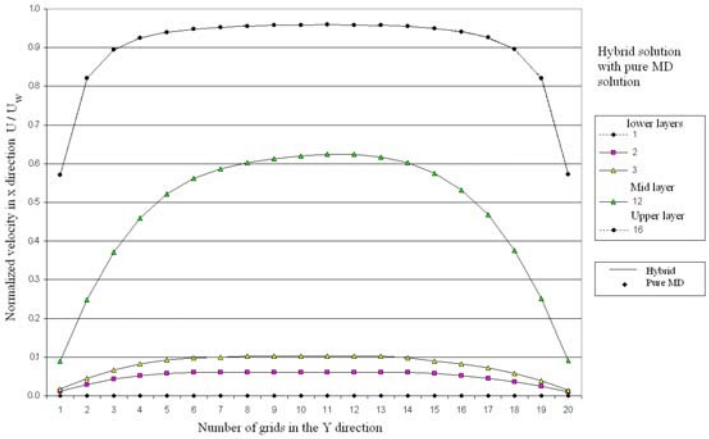

Figure 5- different layers of fluid undergoing Couette flow with hybrid solution and pure MD solution

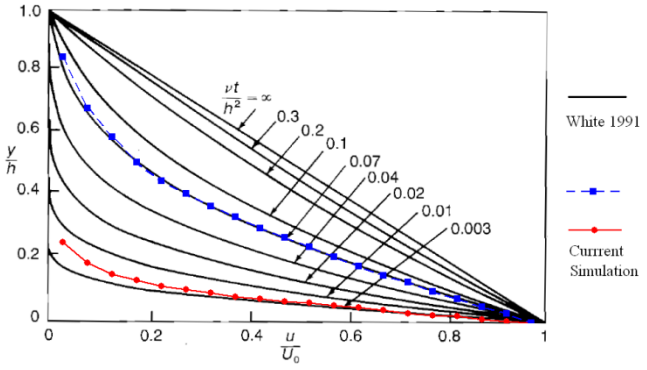

Figure 6- Comparison of obtained results with analytical results for sudden start-up Couette flow

CONCLUSION

A computer program based on a Molecular Dynamics-Continuum hybrid numerical method has been developed. The prepared algorithm and the computer code are capable of computing flows in micro and nano-scale geometries. MD simulations are used in interfacial regions where the discreteness of the fluid is important and the Navier–Stokes equations are solved in regions where a continuum description is accurate. The two descriptions are coupled by imposing continuity of fluxes at the boundaries of an overlap region. The mean particle velocities provide boundary conditions for the NS solution at one side of the overlap region (P→C). A constrained dynamics algorithm forces the instantaneous mean particle velocity to equal the continuum solution at the other boundary (C→P). Flux across the overlap region is maintained by adding or removing a number of particles that is consistent with the continuum flux.

Simulations of steady and sudden start-up Couette flow are investigated in a 3D nanochannel and the obtained results are compared to pure MD simulation and analytical solutions. As the overlap region expands to two dimensions the unsatisfactory fluctuations become necessary and to cope with current phenomena gradually increasing relaxation coefficient is implemented.

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