Multi-species Fluid Flow Simulations using a Hybrid Computational Fluid Dynamics - Molecular Dynamics Appraoch ***Jeff: Or "Polyatomic Lagrangian Dynamics Modelling for a Hybrid Computational Fluid Dynamics - Molecular Dynamics Appraoch"

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***Jeff: Before going further, check the AIAA membership on co-authors: basically Dimitris is most likely to own the membership **The constrained Lagrangian dynamics modelling in the hybrid computational fluid dynamics (CFD) - molecular dynamics (MD) approach is improved for the simulation of multi-species polyatomic fluid. Microscopic mean velocity term on the classical Lagrangian dynamics equation is replaced by the division of mean linear momentum and mean mass to account for multi-species fluid system. Also, the equation is applied on molecules instead of individual atom, to preserve the linear momentum between continuum and particle domain without encountering the unfaborable numerical break-down of molecular structure. We verify our hybrid CFD-MD simulation package by analyzing a nano-scale transient Couette flow of a single monatomic fluid. We will evaluate the multi-species polyatomic Lagrangian dynamics modelling by analyzing two different fluid models: the mixture of two monatomic fluids and a polyatomic molecular fluid under the short-range potential. These two applications will describe the effect of particle-level mass variation on the macroscopic flow evolution.**

I. Introduction

The hybrid computational fluid dynamics (CFD) - molecular dynamics (MD) approach is getting more attraction as a potential answer in describing the nano-scale flow phenomena. In this approach, the fluid system is divided into subdomains and individual subdomain is solved by either the continuum solver or the particle-based solver. Conventionally, the macroscopic flow region where the continuum hypothesis is valid is resolved by the continuum formulation and the material interface (e.g. fluid/solid or fluid/fluid) is analyzed by higher degree-of-freedom particle formulation. Compared with the classical CFD or MD methodology, this

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approach is expected to provide the high-resolution solution near the wall boundary within the acceptable computational cost.

A number of scientific studies have been published which improve the hybrid technique and/or apply this approach to various nano-scale flow fields. These researches can be categorized to constrained Lagrangian dynamics,[1,2,3,4,5,6,7,8] alternating Schwarz method,[9,10,11,12,13] and direct flux exchange,[14,15,16,17,18,19] according to the formulation of hybrid shcemes and the characteristics of variables exchanged in the overlapping region. Of these methods, the constrained Lagrangian dynamics applies the constrained Lagrangian dynamics equation to impose the hybrid MD boundary condition. Continuum and particle solvers exchange density properties (i.e., conservative variables) and they are coupled in time space. Compared to other counterparts, this method is easy to implement, is directly applicable to the unsteady flow simulation, and its molecular samples are less noisy than the flux properties.

Meanwhile, this approach is only valid for the single-species monatomic fluid flow since the constrained Lagrangian dynamics equation does not consider the mass variation of fluid particles. So, the direct aplication of the classical constrained Lagrangian dynamics equation to multi-species fluid domain results in the break-up of momentum conservation. Also, the application to the polyatomic fluid whose atomic mass are different ends up with the numerical break-up of chemical bond.

This motivates us to refine the classical constrained Lagrangian dynamics equation for the application to multi-species and polyatomic fluid flow. The equation is reformulated to provide the linear momentum conservation. Also, the equation is applied on the molecules instead of individual atoms, to prevent the numerical break-up of chemical bond. We implement this equation on our hybrid CFD-MD simulation package which has been introduced in our previous article[8] and apply it to solving multi-species and polyatomic fluid flow.

We introduce the hybrid CFD-MD approach and describe the Lagrangian dynamics equation for multi-species polyatomic fluid particles in Section II. Numerical methods and hybrid interface on individual solver will be addressed in Section III. Section IV will be dedicated to present numerical solutions of the transient Couette flow in different fluid systems. The first system consists of two monatomic fluids whose chemical properties are equivalent to the liquid argon with the variation in mass. The next one contains the polyatomic molecules whose molecular structure is equivalent to water while the long-range interaction (i.e., Coulombic force) is not considered. We will summarize our studies and propose further applications in Section V.

II. Hybrid CFD-MD Approach for Multi-species Polyatomic Fluid

II.A. The Hybrid CFD-MD Approach

A detailed structure of the fluid domain for the hybrid CFD-MD approach is described in Figure 1. CFD solves the flow region where the continuum hypothesis is valid while MD analyzes the complex microscopic flow feature near the solid obstacle. Overlapping region is placed sufficiently far from the solid stationary wall to prevent the direct influence of molecular-level physics.

The overlapping region is designed sufficiently large to contain five individual layers with sufficient spacing. From the bottom, we have the *particle-to-continuum* and *continuum-to-particle* (denoted as 'MDtoCFD' and 'CFDtoMD', respectively) layers where hybrid boundary condition for CFD and MD are imposed. The external force layer for particle systems is placed on the top (farthest from the material interface) of the overlapping region. These three "active" layers are separated by the buffer layer, which is designed to prevent direct interaction between particles in above "active" layers.

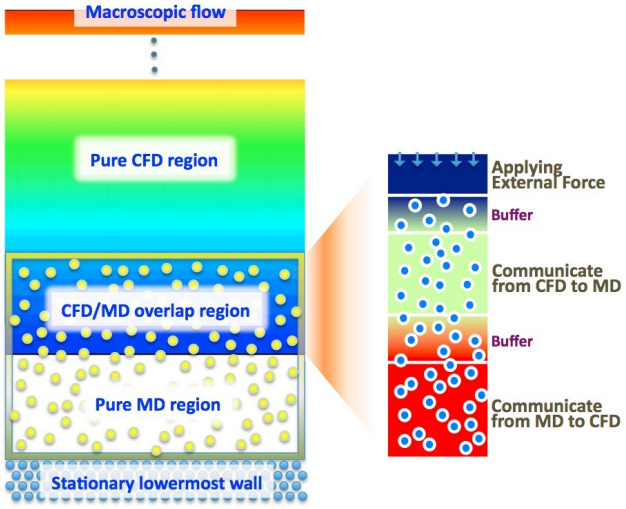

Figure 1. Schematic Diagram of the Hybrid Domain with Detailed View of Overlapping Zone: Left figure expresses the composition of hybrid simulation domain and the right figure presents individual layers in the overlapping region.

Hybrid CFD boundary condition on MDtoCFD layer is imposed by averaging molecular properties located in this layer. This boundary condition is prone

to suffer from the statistical error,[11,18] since the finite number of particles participate in sampling process in space and time. Therefore, the delicate determination of coupling parameters are very important for acquiring the noise-free hybrid solution. The distance from the material interface and the size of sampling layer determines the scale of statistical error in space; how long the molecular properties will be accumulated ("sampling duration") and how often these samples are applied to the continuum solver ("sampling interval") also affects the sampling noise in time.

The hybrid MD boundary condition is imposed by referencing the continuum solution in this layer. A single set of continuum solution is imposed to higher degree-of-freedom molecular domain through the constrained Lagrangian dynamics equation. This equation constrains particles to attain the macroscopic flow property (conservative properties) on average, while preserving their degree-of-freedom of translational motion. Details on this equation will be presented in Section II.B.

In the uppermost layer, a fictitious external force is exerted on particles to preserve the system ensembles of the particle domain. This force function is designed to be short-range so as not to influence the motion of the particles past the buffer layer in the CFDtoMD domain. The force stiffens as the particles approach outer region to prevent the particles from drifting out of the particle domain. We apply a cost-effective classical external force model by Nie *et al.*[2]

II.B. Constrained Lagrangian Dynamics for Multi-species Fluid Simulation

Once the hybrid simulation accomplished for the monatomic system, it was developing for the more realistic problem, which is multi-species polyatomic fluids including water molecules. The first step of the development was start with developing to the equation of motion in coupling CFDtoMD region. The average velocity of particles in Jth cell is equal to the velocity u_J in continuum cell.

$$u_J(t) = \frac{1}{N_J}\sum_k v_k = \frac{\sum_k m_k\ \dot{x}_k(t)}{\sum_k m_k} \tag{1}$$

where v_k is the velocity of k^{th} particle and N_J is the number of particles in the cell, and m_k is the mass of k^{th} particle. With taking Lagrangian derivative of eq. 1,

$$\frac{Du_J(t)}{Dt} = \frac{\sum_k m_k\ \frac{d\dot{x}_k(t)}{dt}}{\sum_k m_k} = \frac{\sum_k m_k\ \ddot{x}_k(t)}{\sum_k m_k} \tag{2}$$

The Classical MD equation of motion can be generalized to obtain the constraint by adopting the fluctuation in the acceleration of each particles, ζ_k

$$\frac{F_k}{m_k} = \ddot{x_k}(t) = \frac{Du_J(t)}{Dt} + \zeta_k = \frac{\sum_k m_k\ \ddot{x}_k(t)}{\sum_k m_k} + \zeta_k = \frac{\sum_k F_k(t)}{\sum_k m_k} + \zeta_k \tag{3}$$

$$\zeta_k = \ddot{x_k}(t) - \frac{\sum_k F_k(t)}{\sum_k m_k} = \frac{F_k(t)}{m_k} - \frac{\sum_k F_k(t)}{\sum_k m_k} \tag{4}$$

where $m_k = m$ only for monatomic system,

Finally, constrained particle dynamics equation was modified with the conventional equation of motion considering multi-mass to simulate polyatomic CFD-MD coupling hybrid simulation, it can be written as:

$$\ddot{x_k}(t) = \frac{F_k}{m_k} - \frac{1}{m_k}\frac{\sum_{i=1}^{N_J} F_i}{\sum_{i=1}^{N_J} m_i} - \frac{m_k}{\Delta t_{MD}}\{\frac{\sum_{i=1}^{N_J} m_i\dot{x}_i}{\sum_{i=1}^{N_J} m_i} - u_J(t+\Delta t_{MD})\} \tag{5}$$

The acceleration of the cell is now constrained instead of the average acceleration of the particle in the cells in this equation. Again, these two quantities are the same when all of the masses of atoms are the same.

III. Development of a Hybrid CFD-MD Simulation Package

We address the numerical schemes and hybrid interface of individual CFD and MD solvers in brief: full details can be found in Ref. 8.

III.A. Continuum Incompressible Flow Solver

The current in-house continuum hydrodynamics code solves the unsteady incompressible Navier-Stokes equations. In this work, the pseudo-compressibility method[20] is adopted to form a hyperbolic system of equations which can be marched in pseudo-time. For time-accurate unsteady simulation, a dual time stepping method is adopted and it is combined with the LU-SGS (Lower-Upper Symmetric Gauss-Seidel) scheme[21] for the implicit time integration. The inviscid fluxes are upwind-differenced using Osher's flux-difference splitting scheme.[22] For higher-order spatial accuracy, the MUSCL (Monotone Upstream-centered Schemes for Conservation Laws)[23] approach is used on the inviscid flux calculation. Viscous fluxes are calculated using conventional second-order central differencing.

III.B. Particle Dynamics Solver

Newton's conservation of momentum is employed at the atomic level to propagate the system's motion through time evolution. In this work the most commonly used Lennard-Jones (12-6) intermolecular force potential model[24] is employed to calculate pair-wise interactions of particles in the system. A cut-off distance is introduced to reduce the computational cost and is set to be 2.2 magnitude of atomic characteristic length.[25] The most common velocity Verlet algorithm[24] is employed for time integration of the equations of motion of the interacting particles and to compute molecular trajectories in the simulation. In this work, the MD simulations were performed by using an appropriately modified version of the Large Atomic Molecular Massively Parallel Simulator (LAMMPS). It is a classical molecular dynamics open-source code written in C++ and developed by Sandia National Labs.[26]

III.C. Implementation of Hybrid Schemes and Interface

The file-based communicator is implemented on each solver to exchange conservative properties at every sampling interval. CFD solver stores the instataneous solution at that time instance, while MD solver produces the backward sample over the sampling duration. Thus, the extrapolation/interpolation mechanism for time-accurate hybrid simulation[8] is also incorporated to avoid time-lagging pattern. CFD solver directly applies these extrapolated/interpolated properties as the hybrid boundary condition. MD solver inputs these values on employed constrained Lagrangian dynamics formulation. MD solver also applies the external force function in the uppermost layer.

The unit conversion function is also incorporated in CFD solver. This function changes the non-dimensional CFD solution into non-dimensional MD unit and vice versa. Considering the current formulation of CFD solver, the artificial pressure property needs to be converted to an equivalent molecular mass density. We acquire the relation between artificial compressibility P and density ρ by comparing the mass conservation equation and artificial compressibility formulation:

$$\frac{\partial P}{\partial t} = a^2 \frac{1}{\rho} \frac{\partial \rho}{\partial t} \tag{6}$$

This relation, written in discretized form, provides the hybrid number density and/or aritificial pressure boundary conditions. CFD solutions in the hybrid MD boudnary layer is rewritten in density form as,

$$\rho_{N+1} = \frac{\rho_N}{1 - \frac{P_{N+1} - P_N}{a^2}} \tag{7}$$

Likewise, sampled molecular density is converted to equivalent artificial pressure at the hybrid CFD boundary layer as,

$$P_{N+1} = P_N + a^2(1 - \frac{\rho_N}{\rho_{N+1}}) \tag{8}$$

From above equations, a denotes the artificial speed of sound and N, $N+1$ stands for the time step.

Coupling parameters, such as location and size of hybrid layers in space and sampling interval and sampling duration in time, are delicately determined by the reference to previous studies[2,6,7,10,12,14,16] and preliminary measurement of noise level on pure molecular dynamic domain. We also apply the replica sampling approach[27] in case the individual hybrid solution suffers from the excessive sampling noise. This approach averages multiple independent simulations from the different initial Maxwell-Boltzmann distribution to find the non-fluctuating solution. The solution from N replicas provide the same order of accuracy as an individual solution from N times larger domain: this approach (running multiple small tasks instead of one big task) is computationally more effective in view of scheduling on many supercomputing resources.

IV. Numerical Results

IV.A. Problem Description and Validation

The application problem is a transient Couette flow problem, which is widely used for the verification of the hybrid CFD-MD solver. Fluid model is the liquid argon, whose characteristic length is $\sigma = 3.405\times10^{-10}$ meter and the time scale is $\tau = 2.2\times10^{-12}$ second. The number density is $0.81m\sigma^{-3}$. The channel is 52 σ in height and the solid wall has artificial properties that are the same as those of liquid argon. The slip ratio between the fluid and the wall material is set at 0.6 to satisfy the linear velocity gradient along a vertical direction.[2]

The computational domain for the hybrid simulation is depicted in Figure 2. The pure MD region is specified as 10 σ, which was reported to be sufficient to prevent the excessive sampling noise on the hybrid CFD boundary condition.[6] Individual hybrid layer has 2 σ in height. Hybrid CFD boundary region consists of two consecutive layers to provide the accurate boundary condition for this collocated mesh system. The width of the MD domain along the periodic direction is determined at 140 σ, after a number of numerical experiments. Both the sampling interval and the sampling duration are set to be 10 τ, considering the characteristics of our deterministic application targets.

Figure 3 presents a transient Couette flow profile by the hybrid simulation which is compared to the analytic solution. The hybrid solution succeeds in describing the same flow physics as the analytic solution under the level of molecular dynamic sampling noise. This evaluates the accuracy of the current hybrid simulation package as a tool for solving the nano-scale flow field. This sampling noise is even diminished further through the sampling of multiple replicas.[8]

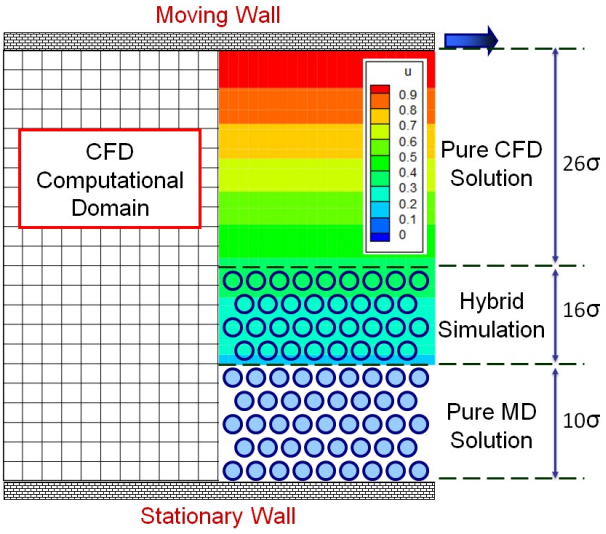

Figure 2. Computational Domain of the Couette Flow Simulation: The height of the fluid domain is 52σ (≈177Å). CFD mesh size is 71×27 and CFD cells at the pure MD region are treated as holes. MD domain size is about 140σ along the horizontal direction and around 26σ along the vertical direction, including the bottom wall. Periodic boundary condition is applied on the principal flow direction.

IV.B. Transient Couette Flow Profiles in Multi-species and Polyatomic Fluids

We currently apply our simulation package to solving two fluid systems. The first experiment solves a liquid compound which consists of two monatonic molecule species differing in mass. Chemical properties of each artificially designed molecule are set to be the same as that of the liquid argon. This numerical experiment will aims to verify the accuracy of the improved constrained Lagrangian dynamics equation, by comparing its result with the pure molecular dynamic solution.

The next experiment will solve a polyatomic molecular fluid whose molecular structure is equivalent to that of water. This experiment focuses on proving that imposing the constrained equation on the center of mass of each molecule is advantegeous in preserving the molecular structure of polyatomic fluid, compared with the traditional imposition of constrained equation on individual atom. We sense that the hybrid CFD-MD approach is incomplete in considering the long-range force field since outside the hybrid region is treated as the vacuum in particle domain. Thus, we apply the short-range Lennard-Jones potential for describing the molecular motion and compare our hybrid solution with the pure molecular dynamic simulation on the same flow field.

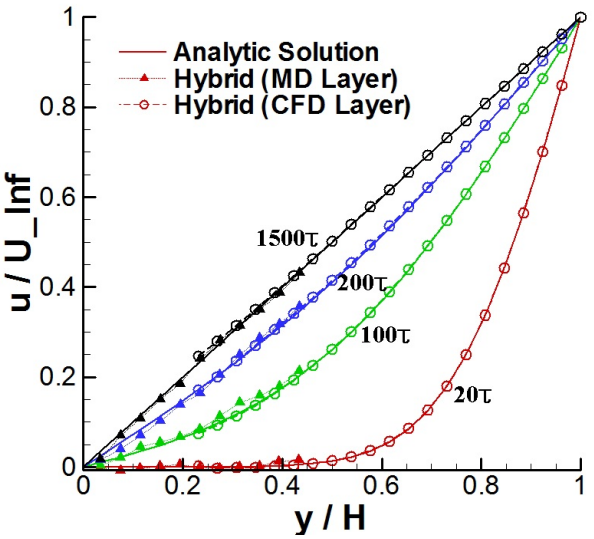

Figure 3. A Time-accurate Couette Flow Profile: The evolution of velocity field along the vertical direction is presented. CFD solution is the instantaneous profile at specified time and MD solution is spatially averaged over 2 σ in height and temporally averaged for 1 sampling durations (=10τ). Hybrid solution presents the same flow physics as the analytic solution with the slight variation due to the sampling noise.

V. Conclusion and Future Works

We have proposed the improved constrained Lagrangian dynamics model for a hybrid CFD-MD simulation in this paper. It is designed to satisfy the momentum conservation of the fluid solution and avoid the numerical unstability in applying the equation to polyatomic molecules. The classical constrained Lagrangian dynamics equation is reformulated to satisfy the linear momentum conservation of the multi-species fluid system and the equation is applied on the center of mass of each molecule to preserve the chemical bond of a polyatomic molecule. We have implemented this model on our hybrid CFD-MD simulation package which consists of a pseudo-compressibility incompressible Navier-Stokes solver and a molecular dynamics solver solving the Lennard-Jones potential. The simulation package has been verified by solving the transient Couette flow profile of a single-species monatomic fluid system. It is under the application to two fluid systems who contains different atomic species in the particle domain. Simulation of the monatomic fluid solution with different mass will verify the accuracy of the improved equation. Simulation of a polyatomic fluid will provide the numerical validity of imposing the constrained Lagrangian dynamics equation on molecular level. These numerical experiments, in turn, will express the potential to apply the hybrid CFD-MD approach to any fluid systems.

The future work will be dedicated to solving the long-range force field (e.g., Coulombic force) as well as the short-range Lennard-Jones potential on the particle domain. Hybrid simulation so far does not consider long-range potentials since outside the hybrid region is modelled as the vacuum state in view of particle domain. Mathematically modelling the long-range force field or putting the virtual slab on the border of hybrid region could be ones of potential answers.

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Content

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