Computational Uncertainty in Hybrid Atomistic-Continuum Frameworks

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Abstract Over the past decade micro and nanofluidics emerged as vital tools in the ongoing drive towards the development of nano-scale analysis and manufacturing systems. Accurate numerical modelling of the phenomena involved at these scales is essential in order to speed up the industrial design process for nanotechnology. However a parameter often ignored in hybrid simulations is the uncertainty level introduced in the numerical modelling of phenomena taking place at micro and nanoscales. The main interest of the present study is the propagation of the inherent atomistic fluctuations to the continuum solver in the case of multiscale modelling and hybrid solvers.

Keywords: Computational Uncertainty, Hybrid Atomistic-Continuum Frameworks, Molecular Dynamics

1. Introduction

Over the last years, micro and nanofluidic devices have experienced a continuously increasing applicability in a broad spectrum of disciplines spanning from biomedicine to nanoelectronics. The fluid flow through devices operating at these scales presents inhomogeneities near the solid boundaries causing the breakdown of the no-slip condition and the presence of slippage. Quantifying the slip's magnitude is essential for measuring, either experimentally or computationally, accurately flow characteristics such as shear rates and velocity profiles. The slip's strength is usually expressed through a proportionality coefficient, called as slip length Ls, representing the extrapolated distance from the wall to the point where the tangential velocity component is equal to zero (Asproulis and Drikakis, 2010). Although the parameters affecting the generation of slip at the solid-liquid interface are not explicitly known, surface roughness, fluid structure, wettability, shear rate and surface structure are considered among the main contributing factors. Experimental studies of nano-transport phenomena and slippage effects present significant challenges primarily associated with the manufacturing difficulties of controlling the atomistic roughness and the measuring accuracy of physical quantities at nano scales and therefore complimentary atomistic numerical modelling is usually embraced.

Thus, modelling phenomena at micro and nanofluidic devices presents significant difficulties due to the inaccuracy of the continuum models and inefficiency of the molecular ones. Aiming to circumvent this dilemma, multiscale frameworks, called hybrid codes, have been developed to couple the microscopic and macroscopic description of a system and to facilitate the exchange of information. Hybrid methods bridge the gap between the macroscopic and microscopic length scales and provide a unifying description of liquid flows from nanoscale to larger scales. Several hybrid methods have been proposed in the literature and can be broadly classified to the domain decomposition and embedded frameworks (Karniadakis et al., 2005). The former decompose the computational domain in two distinct regions where the first is solved by the continuum solver and the second, that needs molecular modelling, is solved by molecular dynamics. In the embedded techniques the entire domain is resolved by the macroscopic model and the microscopic solver is used as a local refinement to provide data required by the continuum description.

One of the most challenging tasks in the development of hybrid atomistic-continuum is the transfer of information from the continuum description to the atomistic system. The main difficulty lies in the disparity between the degrees of freedom modelled by the atomistic and continuum models. However, one important issue that is usually neglected is the computational uncertainty that is propagated as atomistic information is transferred to the continuum description and vice versa.

2. Error Sources and Numerical Uncertainty

In engineering simulations usually the concept of uncertainty is either ignored or found under various connotations such as variability, inaccuracy, and degree of confidence. This primarily arises from the difficulty associated with the explicit definition and evaluation of uncertainty in the context of deterministic computational modelling. Analysing uncertainty levels of either hybrid or deterministic simulations, poses significant challenges related to the inherent uncertainty of the system, named as aleatory uncertainty, and the lack of knowledge, known as epistemic uncertainty.

The techniques mainly applied for the characterisation of uncertainty can be broadly classified as probabilistic and non-probabilistic ones (Roache, 1997; Najm, 2009; Wang and Karniadakis, 2006). The probabilistic approach employs the probability theory, through the use of probability density functions (pdf), to represent the randomness of the system with the center of the probability density function to represent the most probable outcome of the simulation. In the non-probabilistic approach sensitivity analysis, fuzzy theory, and interval analysis are employed to manage the lack of knowledge of certain models and parameters.

In the scope of the hybrid computations one issue that is mainly ignored in the literature is the quantification of the uncertainty levels introduced to each description, either molecular or continuum, through the hybrid interface. Although in hybrid simulations several sources of error may be present the factor that mainly affects the uncertainty levels in the continuum region is the propagation of molecular fluctuations through the hybrid solution interface. The inherently statistical nature of the atomistic simulations and the necessity of finite sampling and averaging for calculating macroscopic fields, such as temperature or velocity, introduce additional statistical errors primarily associated with the implementation of the numerical algorithm used in the phase space. The main scope of the present study is to review the techniques employed for calculating from the molecular solver macroscopic fields and introduce an approach for automatic calculation of these properties within certain fluctuation criteria.

3. Hybrid Atomistic-Continuum Frameworks

Over the last year several multiscale approaches for simulating micro and nanoflows have been proposed in the literature (Kalweit and Drikakis, 2008a; O'connell and Thompson, 1995; Flekkoy et al., 2000; Delgado-Buscalioni and Coveney 2003; Werder et al., 2005; Nie et al., 2006; Li et al.,1999; Asproulis, Kalweit and Drikakis, 2008; Hadjiconstantinou and Patera, 1997; Asproulis and Drikakis, 2009). The domain decomposition technique has been embraced in the majority of the majority of these methods and utilizes the fact that the continuum model although it fails to provide accurate prediction of the flow phenomena in small subsets of the computational region, is valid for the rest of the computational region. The underlying idea is to decompose the domain into two regions, where the one is modelled using molecular dynamics (MD) and the other through a continuum fluid dynamics (CFD) solver (Kalweit and Drikakis, 2008a; O'connell and Thompson, 1995). The solution is computed independently in each region and the two descriptions exchange information at specific points in time through a hybrid solution interface (Kalweit and Drikakis, 2008a,b). Two main benefits

primarily arise from this approach. Firstly, regions where the continuity assumption breaks down are treated through the molecular solver and secondly the computationally intensive atomistic simulations are spatially restricted only in small regions where necessary. However, the domain decomposition the main limitation is time scale coupling between the two approaches since the overall computational time is limited to the time scales computed by the microscale solver (Asproulis, Kalweit and Drikakis, 2008; Asproulis, Shapiro, Kalweit and Drikakis, 2008).

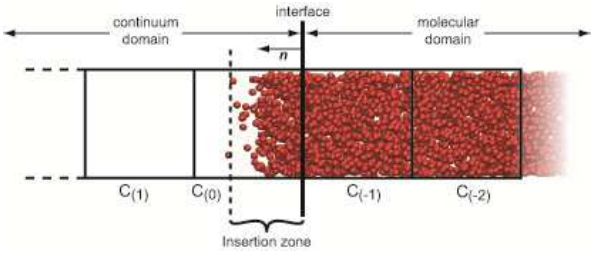

Figure 1: Domain decomposition schematic

Another approach used for coupling molecular and continuum description is the Point Wise Coupling (PWC) and in this approach the whole domain is covered with the macroscopic solver and the microscale model, which enters as a refinement, is used to obtain macroscopic properties. PWC approach couples molecular with continuum simulations by performing MD simulations around a grid point for a number of time steps at every time step of the macroscopic CFD solver. In this approach the molecular model is treated as a refinement of properties obtained by the macroscopic solver. Specifically, the MD simulations are constrained from the macroscale by the velocity gradient are performed in the beginning of each macroscopic time step. The molecular models are utilised to confront two different types of fluid flow problems (Ren,2007; Ren, 2005): a) Boundary condition problems: The microscopic simulations are engaged to calculate the appropriate boundary conditions, and b) Constitutive relations problem: The molecular simulations are utilised to calculate the constitutive relations that are needed from the continuum solver.

The two different types of problems are associated with different nature of information that is exchanged between the molecular and continuum models. In both cases the molecular simulations are constrained from the continuum solver through the strain rate. In each of the aforementioned problems different techniques are utilised for the MD simulations.

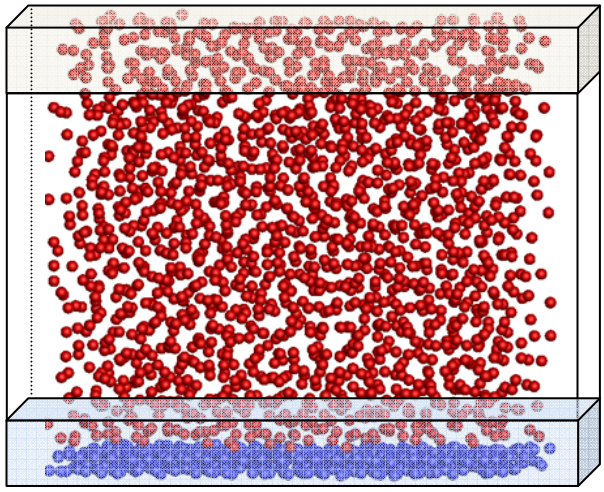

Figure 2: Schematic of PWC, with the upper region to represent the boundary transfer domain and lower one to represent the solid wall (blue particles)

4. Calculation of Macroscopic Properties

The statistical effects are quantified through confidence intervals that can be either estimated a priori or in the case of hybrid simulations calculated during the simulation process by quantifying the variance of averaging quantities. Standard deviation of physical substance $\langle A \rangle_\tau$ is usually employed to quantify its fluctuations strength (Hadjiconstantinou et al., 2003). Basically, it gives the average deviation of any computed value $\langle A \rangle_\tau$ from the true value of A. Assuming that the individual quantities $A(\tau)$ are independent from each other, it is defined by

$$\sigma(< A >_t) = \sqrt{\sigma^2(< A >_t)} \qquad (1)$$

with $\sigma^2(< A >_t)$ being the variance in $\langle A \rangle_\tau$ that is simple given by:

$$\sigma^2(< A >_t) = \frac{\sigma^2(A)}{M} \tag{2}$$

For a time averaged quantity the average fluctuation strength can be defined as the fractional error, which is the standard deviation of $\langle A \rangle_\tau$ over its true value:

$$F_{\langle A \rangle_t} = \frac{\sigma(\langle A \rangle_t)}{|A|} = \frac{\sqrt{\sigma^2(\langle A \rangle_t)}}{|A|} \tag{3}$$

An important parameter that one may need to calculate is the number of time steps M that the averaging has to be performed in order to reduce the fluctuations level below an acceptable value. In that sense, Equation 3 can be re-arranged to give the minimum value for M

$$M = \frac{\sigma^2(A)}{F^2 A_0^{\,2}} \tag{4}$$

where A_0 is the true value, or the limit of the average $A_0 = \lim_{t\to\infty} \langle A \rangle_t$.

The predictions can be given for: density, velocity in one dimension, temperature, and pressure are:

$$\sigma(\rho) = \frac{m\sqrt{N}}{VAc} \tag{5}$$

$$\sigma(u_\alpha) = \sqrt{\frac{k_B T}{mN}} \tag{6}$$

$$\sigma(T) = \sqrt{\frac{k_B T^2}{c_v N}} \tag{7}$$

$$\sigma(P) = \frac{k_B T Ac\sqrt{\gamma N}}{V} \tag{8}$$

The most interesting question is: Over how many time steps M should one average to reduce the fraction fluctuation below an acceptable level, for instance $F = 5\%$

$$M_\rho = \frac{1}{F^2}\frac{1}{NAc^2} \tag{9}$$

$$M_u = \frac{1}{F^2}\frac{1}{NAc^2 Ma^2 \gamma} \tag{10}$$

$$M_{e_{int}} = \frac{1}{F^2}\frac{k_B T^2 c_v N}{(e_{i0})^2 V^2} \tag{11}$$

$$M_T = \frac{1}{F^2}\frac{k_B}{Nc_v} \tag{12}$$

$$M_P = \frac{1}{F^2}\frac{k_B{}^2 T^2 Ac^2 N^2 \gamma}{P_0 V} \tag{13}$$

For calculating a quantity, it is necessary to know about the accuracy of the calculation. This is true for experiments as well as for simulations. For equilibrium MD simulations, the accuracy can be increased arbitrarily, within the computational resources, to a desired level by running the simulation longer and averaging over more calculated values. A practical problem one faces is that the variance $\sigma(A)$ and correlation time τ_c are usually not known in advance. Hence, the required minimum number of values $N_{m,min}$ for achieving the required accuracy is unknown and thus the minimum number of time steps which need to be performed as well. Thus, one needs to run the simulation for a period of time which seems appropriate and to calculate the mean, correlation time, variance, and confidence level from the values obtained during this run. If the confidence level is too low, the simulation has to be extended to the estimated minimum number of integration time steps with a new analysis being carried out. This procedure is repeated until the confidence level has reached the required level. Several iterations may be necessary, since the estimates variance, correlation time and confidence level change as the number of calculated values increases.

Executing the described procedure manually, by using statistical software tools for the analysis is elaborate and time consuming. Therefore, to confront this problem and formulate an automatic procedure that can be implemented within the hybrid atomistic-continuum framework an *automatic calculation simulation mode* (ACSM) can be implemented (Kalweit, 2008c). The basic principle is to extend the MD simulation automatically until the mean value of the calculated variable satisfies the chosen statistical confidence criteria. As input parameters, the procedure requires:

1. the calculation frequency τ_{mes}
2. the calculation period $N_{\tau,mp}$
3. the threshold of the time correlation coefficient ρ_{max}
4. the confidence interval δl_{ci}
5. the confidence level P_{cl}

The present automatic calculation is directly utilized in the PWC where the time scale decoupling is achieved. On the contrary in the geometrical coupling this technique cannot be directly applied in the present form since the number of time step that the atomistic solver will execute until the macroscopic quantities are calculated is predefined based on the continuum time step.

A Lennard-Jones fluid has been selected as the first test case employed for the automatic calculation procedure. A cut-off distance $r_c = 2.5\sigma$ has been employed with an integration time step $\delta t = 0.005$ in LJ units. The temperature is controlled through a weak velocity rescaling every 1000 time steps with scaling factor $f_s = 0.1$. A total of 2000 particles have been generated based on an fcc lattice and have been initalised through a Maxwell-Boltzmann distribution. The calculation frequency employed is $\tau_{mes} = 10$ time steps over intervals of $N_{\tau,mp} = 10^3$. In Table 1 the calculated values for the potential energy and pressure are provided.

ρ	T	Variable	μ	σ	δl_{ci}	$N_{\tau,\min}$
0.1	2	e_p	-0.05	10^{-6}	0.1%	960240
0.1	2	P_s	0.19	0.04	1%	676940
0.8	1	e_p	-3.57	0.01	0.01%	181720
0.8	1	P_s	1.63	0.86	1%	215620
1.2	1	e_p	-6.65	0.02	0.01%	85250
1.2	1	P_s	17.25	51.4	1%	18800

Table 1: Calculated state variables for the used LJ-material including the statistical accuracy and minimum time steps

From the data it is obvious that for all states energies can be calculated with much higher precision than the pressure with comparable computational effort. When comparing the number of required integration time steps, one needs to bear in mind that the computational effort per time step is less for lower densities than for higher ones, because of the lower number of interactions that need to be computed

5. Conclusions

In hybrid simulations various uncertainties and sources of error exist linked to the assumptions of the physical models employed, and the inherent statistical fluctuations of the numerical procedure due to the effects of boundary conditions, cut-off radius and computational time step. Despite the several origins that contribute to the total uncertainty level the complexity of the multiscale solvers and the fact that their behaviour is not completely understood prevents the scientific community from performing thorough uncertainty analysis for this kind of problems. The first step towards the reduction of statistical fluctuations introduced to the continuum solver is to monitor the averaged information travelled from the atomistic to the continuum side and through proper averaging imposing confidence limits.

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