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Modelling Unsteady Processes with the Direct Simulation Monte Carlo Technique

H.M. Cave[1], K.-C. Tseng[2], J.-S. Wu[3], M.C. Jermy[1], Y.-Y Lian[2], S.P. Krumdieck[1], T.-C. Kuo[2] and M.-Z. Wu[2].

[1]Department of Mechanical Engineering
University of Canterbury, Christchurch 8140, NEW ZEALAND.

[2]National Space Organisation
Hsinchu Science Park, Hsinchu 30050, TAIWAN.

[3]Department of Mechanical Engineering
National Chiao-Tung University, Hsinchu 30050, TAIWAN.

Abstract

Over the past 40 years, the Direct Simulation Monte Carlo (DSMC) technique has been developed into a flexible and effective solver for flow problems in the rarefied to near continuum regime. However, even with modern parallelised code, the efficient computation of unsteady near-continuum flows, which are important in processes such as Pulsed Pressure Chemical Vapour Deposition (PP-CVD), remains a challenge. We have developed an unsteady parallel DSMC code (PDSC) utilising advanced features such as transient adaptive sub-cells to ensure nearest neighbour collisions and a temporal-variable time step to reduce computation time. This technique is combined with a unique post-processor called the DMSC Rapid Ensemble Averaging Method (DREAM) which reduces the statistical scatter in the data sets produced by PDSC. The combined method results in a significant memory and computational reduction over ensemble averaging DSMC, while maintaining low statistical scatter in the results. The unsteady code has been validated by simulation of shock-tube flow and unsteady Couette flow, and a number of test cases have been demonstrated including shock impingement on wedges. The technique is currently being used to model the development of an under-expanded jet in a PP-CVD reactor.

Background

The Direct Simulation Monte Carlo (DSMC) method has become a widely used computational tool for the simulation of rarefied gas flows where effects at the molecular scale become significant [1]. DSMC is a flexible and effective particle based approach in which the movement and collisional behaviour of a large number of representative particles within the flow field are decoupled over a time step which is a small fraction of the local mean collision time. The flow field itself is divided into cells which serve as regions to obtain the local macroscopic properties and for the selection of intermolecular collision partners.

The advent of parallel computing has greatly increased the capability of DSMC to model flows in the near-continuum range with acceptable computational times. One implementation of the DSMC algorithm on parallel systems has been carried out by Wu and Tseng [2]. The code, called PDSC, can be run on two-dimensional, axisymmetric or three-dimensional structured or unstructured meshes and includes many sophisticated features such as dynamic domain decomposition, adaptive mesh refinement, a spatial variable time step scheme and a conservative weighting scheme for the simulation of flows which include trace species.

Unsteady rarefied flows, in which the flow structure changes significantly with time, are interesting flow problems with a number of applications such as the development of under-expanded jets from sonic nozzles during the start up of rocket engines and during the injection phase of the Pulsed Pressure Chemical Vapour Deposition (PP-CVD) process [3]. Unsteady DSMC simulations have been greatly neglected in the literature, primarily because sampling over a small time interval requires either a very large number of simulated molecules or the average of a large number of separate simulations (usually termed "ensemble-averaging"). The associated high computational expense and large memory requirements mean that investigations in the literature tend to be restricted to one-dimensional problems, such as shock tube flow or the shock waves generated by moving one-dimensional pistons. Two dimensional unsteady problems have been attempted, and one method of decreasing the statistical scatter of the results is to use statistical smoothing procedures. Bird's two-dimensional axisymmetric code DS2V incorporates unsteady sampling techniques, however this single processor code is unable to model processes such as PP-CVD with acceptable run-times [4]. The increased computational capacities of parallel-DSMC techniques have the potential to enable the simulation of time-dependent flow problems at the near-continuum regime.

Unsteady Sampling for PDSC

Unsteady sampling procedures for PDSC have been implemented are discussed extensively in the paper by Cave *et al.* [5]. In this section the basics of the procedures will be outlined.

PDSC uses the "time-averaging" method of unsteady sampling illustrated in figure 1. Here, a number of time steps are averaged over an interval just before the sampling time. This method only requires one simulation run, however it suffers a potential disadvantage in that the results will be "smeared" over the time over which samples are taken. Hence the sample time must be sufficiently short to minimize time "smearing" and yet long enough to obtain a good statistical sample. A further disadvantage of the method is that particles may not collide between samples, which will introduce a statistical bias into the results. This method of time averaging has been used previously by Auld to model shock tube flow [6] and is the method used in Bird's DS2V code.

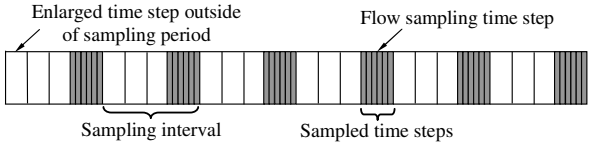

Figure 1. Schematic of the unsteady "time-averaging" sampling method and temporal variable time-step (TVTS) method implemented in PDSC.

The unsteady sampling method in PDSC also incorporates a temporal variable time step (TVTS) algorithm. Here the time step outside of the sampling period is enlarged to speed up computation, without violating the recommended time step size for DSMC [1].

Transient Adaptive Sub-Cells

The use of sub-cells is well established within DSMC research. These cells help improve the "quality" of the intermolecular collisions during the simulation by ensuring collisions occur between "nearest-neighbours" within the sampling cells. Bird has pointed out that sub-cells also help maintain the vorticity within a cell [1]. A measure of the collision quality promoted by Bird is the mean collision spacing to mean free path ratio (mcs/mfp) [7]. Where mcs/mfp << 1, the collision occurs between particles separated by much less than a mean free path, thus maximising the physical realism of the collision process.

As mentioned, PDSC is able to utilise unstructured grids which expedites integration with hybrid solvers and simplifies creating a mesh for complex geometries. Previously it was thought that implementing sub-cells on unstructured grids would be overly complicated as these sub-cells would themselves have to be unstructured. One method of overcoming this problem is to use rectangular transient adaptive sub-cells as illustrated in figure 2.

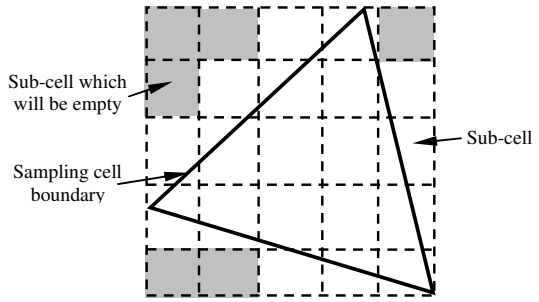

Figure 2. Transient adaptive sub-cells used in PDSC.

The sub-cells are generated for each cell individually during the DSMC collision step based on the number of particles within the sampling cell. Particles are selected from within these sub-cells ensuring nearest neighbour collisions and that collisions cannot occur between the same two collision partners successively. The method requires negligible computer memory expenditure, as the cells are only generated for each cell individually during the collision phase. The additional computational expense depends on the density of particles within the cell, and the authors propose to implement the "virtual sub-cells" algorithm developed by LeBeau *et al.* [8] and promoted by Bird [7] to reduce the computational expense for sampling cells containing low numbers of particles.

The implementation and verification of transient adaptive sub-cells in PDSC will be the subject of a future paper by the authors.

DREAM Post-Processing Method

Despite the efficient implementation of unsteady sampling procedures on parallel computers, simulating denser flows in reasonable computational times requires somewhat of a compromise on the statistical scatter in the results. This is because reducing the statistical scatter significantly in time-averaged data necessitates a very large number of simulation particles with consequent large computational times. Other researchers have attempted to use data smoothing to prepare their results for presentation [9], however ultimately this removes data which may have physical significance.

The approach we have developed is known as the DSMC Rapid Ensemble Averaging Method (DREAM) and is outlined in figure 3. Here we select a raw data set *X-n* produced by PDSC *n* sampling intervals prior to the sampling interval of interest *X*. New particle data is generated from the macroscopic properties in data set *X-n* by assuming a Maxwellian distribution of velocities. The standard PDSC algorithm is then used to simulate forward in time until the sampling period of interest *X* is reached. The time steps close to the sampling point are time-averaged in the same way as in PDSC and this process is repeated a number of times, thus building up a combination of ensemble- and time-averaged data without having to simulate from zero flow time for each run. This process decreases the statistical scatter in the results by adding to the number of particles in the sample, rather than by some artificial smoothing process. Because only a short period of the flow is processed in this way, the scheme has significant memory and computational advantages over both the ensemble- and time-averaging schemes, while allowing a reduction in the number of simulated particles during the initial DSMC run.

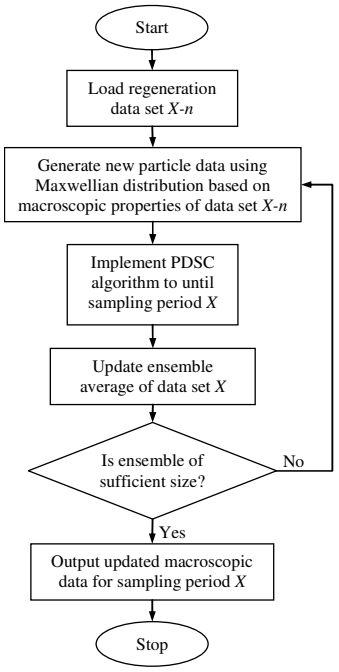

Figure 3. Simplified flow chart of the post-processing technique for unsteady DSMC sampling, called the DSMC Rapid Ensemble Averaging Method (DREAM)

The DREAM method has been extensively tested by Cave *et al.* [5] who have developed rules for the selection of regeneration data sets such that the velocity distribution will "relax" sufficiently quickly towards the true distribution in any non-Maxwellian regions and so that the macroscopic properties at the regeneration data set would not overly constrain the data at the sampling time step.

The method has been shown to reduce statistical scatter in unsteady PDSC data by 2.5-3.3 times with an acceptably low computational expense.

Method Validation

The unsteady sampling procedures and DREAM post-processing method were validated by the simulation of a shock tube and the

development of Couette flow. These simulations are described in more detail in reference 5.

Figure 4 shows the typical flow structure in a shock tube, in which a shock wave is created by bursting a diaphragm between a high-pressure and low-pressure gas. The Riemann incompressible continuum solution for a shock tube allows the properties of the flow structure to be determined at any given time [10].

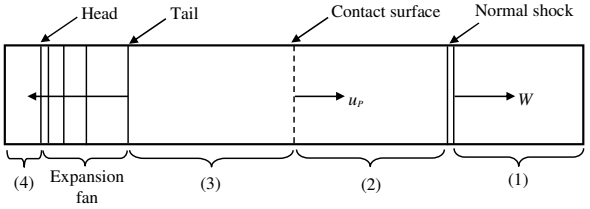

Figure 4. Flow structure in a shock tube. The flow regions include (1) the undisturbed low pressure gas, (2) the constant velocity gas behind the shock front, (3) the gas behind the contact surface between the driving and driven gases and (4) the undisturbed high pressure gas.

To validate the PDSC code, simulations were conducted on a quasi one-dimensional shock tube of length 0.1m and width 0.0125m with argon as the working gas (the VHS molecular model was used). The upper and lower walls were implemented as specular walls to preserve the one-dimensional nature of the flow, while the end walls were simulated as diffusive walls at 300K. The initial conditions in the high pressure and low pressure ends of the shock tube are p_4 = 100Pa and p_1 = 10Pa respectively, however the temperatures at both ends of the tube are the same such that $T_4 = T_1$ = 300K. The solution was first computed using Bird's DS2V code (version 3.7.03) using its standard settings and then a run was conducted using PDSC using a similar number of cells and particles. The DS2V run were conducted on a single processor Pentium IV 3.2GHz (hyper-threading enabled), requiring a run time of ~24,150 hours per second of flow, while the PDSC run was conducted on a PC cluster system of ten Athlon XP2100s, requiring ~2,449 hrs/sec with TVTS disabled. The conditions for the PDSC run were selected to attempt to preserve the simulation conditions from the DS2V run.

Figure 5 shows the temperature profile after solution after 27.45μs. Both methods capture the flow profile accurately with the positions and general structure of each flow feature comparing well with the continuum solution. As would be expected the sharp continuum solution is not followed exactly since it does not include viscous effects.

Figure 6 shows the effect of DREAM on reducing the scatter in the shock profile. Here the amount of statistical scatter in the undisturbed region is approximately halved while maintaining the correct profile throughout the flow.

Another method used to validate both the unsteady sampling techniques in PDSC and DREAM was the simulation of the development of Couette flow. Here argon gas is initially at rest between two parallel diffuse plates at the same uniform temperature as the gas, in this case 300K. At time t=0 the upper plate begins moving instantaneously at speed U_∞=96.6 m/s. These conditions correspond to a Mach 0.3 flow with a Knudsen number of 0.02, based on the initial mean free path and the distance between the walls. Although this problem is one-dimensional, a 100 x 100 cell two-dimensional grid was used to help validate the code. This grid spacing was chosen to be half of the mean free path in the undisturbed gas.

Figure 7 shows that the PDSC/DREAM solution lags the incompressible continuum solution obtained from the Navier-Stokes equations [11]. This is because of compressible effects and because the high level of rarefaction effectively results in slip between gas particles and the walls. The PDSC/DREAM solution also exhibits the expected phenomenon of velocity slip at the walls.

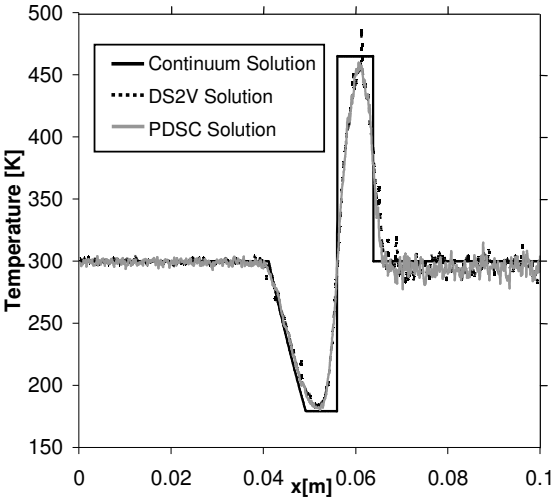

Figure 5. Shock tube flow field profile of temperature at 27.45μs generated using PDSC and DS2V compared to the Riemann solution.

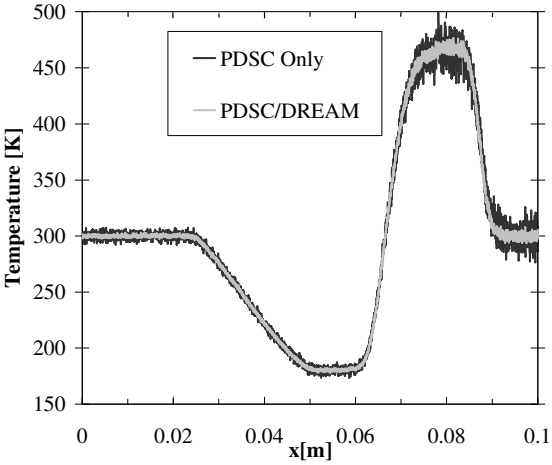

Figure 6. Comparison of shock tube temperature profiles at 76.9μs as predicted by PDSC and after processing by DREAM (ten ensembled samples).

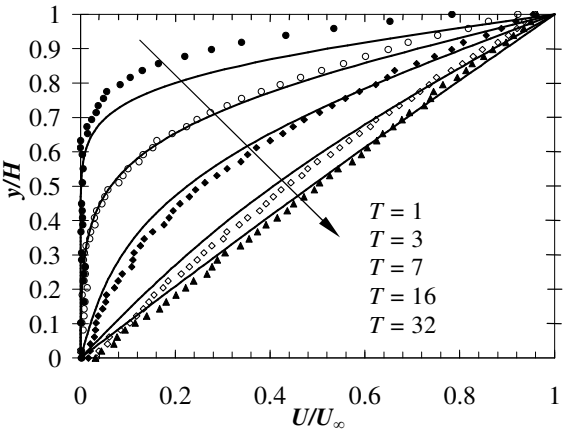

Figure 7. Comparison of Couette flow development predicted by unsteady PDSC/DREAM (symbols) with the exact incompressible Navier-Stokes solution (lines). All times are normalized as $T = tU_\infty/H$.

Shock Impingement on Wedges

A further test problem used as a demonstration is the impingement of a planar Mach 1.3 shock over a two dimensional wedge in a channel. This problem was first studied in the classic experiment by Schardin who used high speed cinematography to study the flow [12]. The flow results in a complex evolution of interacting shock and vortex structures. Numerous authors have also studied similar problems numerically including Huang who used a Navier-Stokes solver [13]. Unsteady PDSC allows the study of the flow under rarefied conditions.

The computational domain for the problem is shown in figure 8. Here the channel wall and wedge surfaces were implemented as specular walls, and the inlet boundary was set to the same conditions as the behind the moving shock. Argon gas was used as the working gas, and the Knudsen number based on the width of wedge normal to the flow and the high density flow to the left of the shock was 0.012.

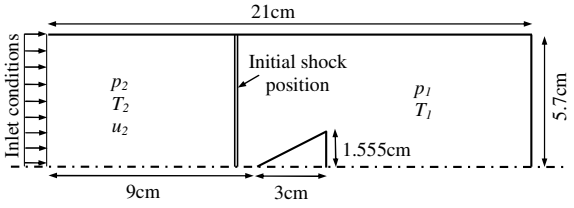

Figure 8. Computational domain for the shock structure passing over a wedge in a channel.

The computational domain was divided into 147,011 unstructured elements, with the element size in the region of the wedge being approximately equal to the equilibrium mean free path of the conditions to the left of the shock. A sampling time step of 2×10^{-8}s was set such that no particle could traverse more than approximately one third of the mean free path over the sampling period, which consisted of 50 time steps. The TVTS scheme was employed, enabling the time step to be increased by a factor of ten when sampling was not occurring. The number of particles within the simulation domain peaked at approximately 21 million, and the total simulation required approximately 3.5 hours of computational time on a PC cluster system of ten Athlon XP2100s (a run without TVTS required approximately 24 hours). The data sets at the points of interest generated by PDSC were then post-processed using DREAM resulting in a reduction of the standard deviation of the density in the undisturbed region of flow from 9.27% in the original data to 2.82% in the processed data and in better resolution of the flow structure.

Figure 9 shows a series of density contours at different times as the shock passes over the wedge. The results reveal some very interesting flow field features, which are qualitatively consistent with the experimental and numerical results of other authors, although exhibit greater levels of rarefaction. As the incident shock passes over the wedge, the reflected cylindrical shock and Mach stem become clearly visible. At the end of the wedge, the Mach stem diffracts around the corner forming a further cylindrical shock and an expansion fan which moves in the opposite direction. The vortex structure formed behind the wedge then begins to move downstream. The cylindrical shocks from the upper and lower corner of the wedge cross each other (or, in the case of the simulation, reflect from the line of symmetry), which can be faintly seen in the figure. As the reflected shock from the front of the wedge grows larger, it reflects from the channel wall and then interacts with the shock structure below it.

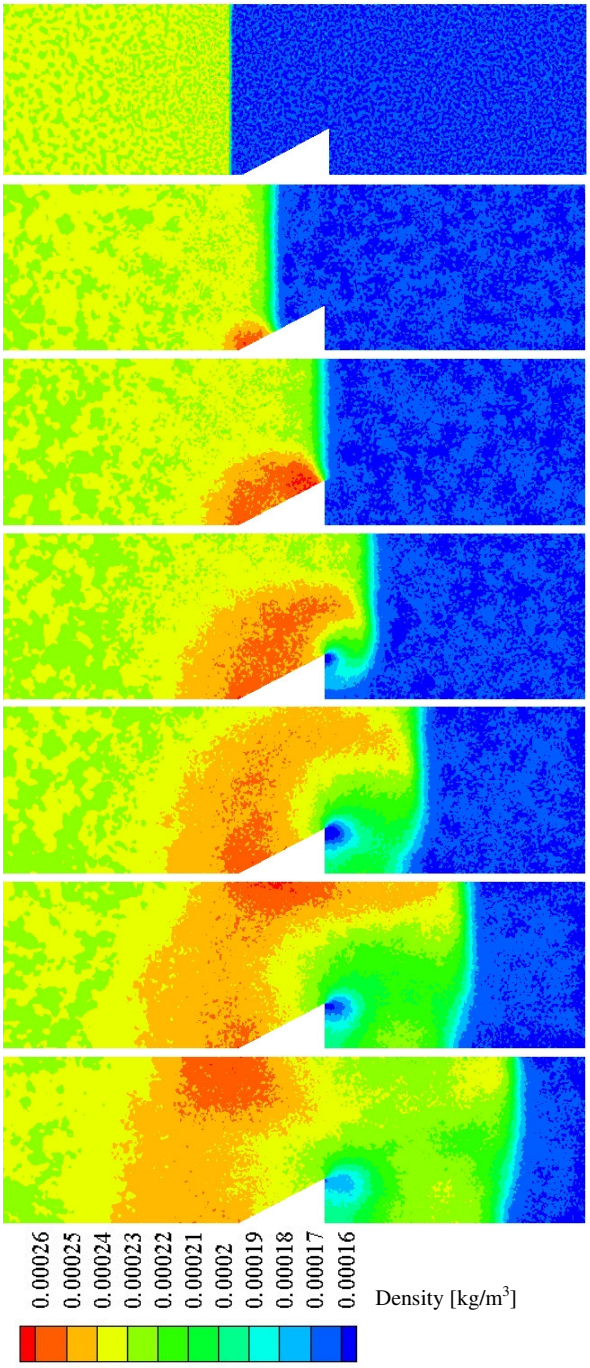

Figure 9. Contours of density for shock impingement on a wedge in a channel after processing by DREAM (Kn = 0.012). Each image is separated by 40μs.

PP-CVD Reactor Simulation

The Pulsed Pressure Chemical Vapour Deposition (PP-CVD) reactor was initially developed by Versteeg *et al.* [14] and has since been extensively investigated by the group led by Krumdieck (see, for example, reference 15). Like other CVD processes, deposition occurs via the thermal decomposition of a precursor on a heated substrate, however unlike other processes the precursor is delivered in an unsteady manner whereby timed pulses of precursor are released into a continuously evacuated reactor volume. The technique can be used to deposit films from

liquid precursors delivered via an ultrasonic nozzle, or from gaseous precursors delivered from a high pressure source volume through an orifice. The films deposited by the technique have been observed to have a highly uniform thickness across the substrate [15].

Figure 10 shows a schematic of a gas-supplied PP-CVD reactor along with a plot of the reactor pressure during operation showing the unsteady nature of the flow.

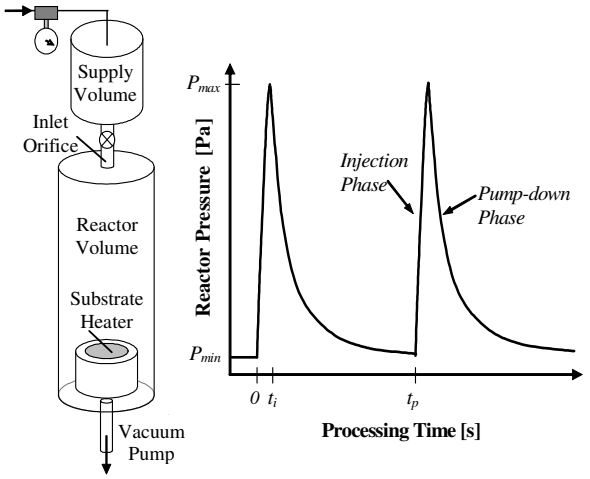

Figure 10. Functional schematic of a Pulsed Pressure Chemical Vapour Deposition (PP-CVD) reactor with a plot of the reactor pressure during operation.

Previous work by Krumdieck *et al.* [3] has shown that there are large regions of the PP-CVD flow field which undergo time-dependent continuum breakdown due to the rapid expansion of the precursor during the injection phase. A precursor particle tracking method was developed by Cave *et al.* [16] and this, along with Bird's DS2V DSMC code, was used to investigate the flow phenomenon in low supply pressure PP-CVD systems [17].

The unsteady nature of the flow, the extremely large density gradient between the inlet and the reactor volume, and the relatively low precursor concentration means the computational expense of modelling experimental PP-CVD reactors using single processor code is unfeasibly high. For these reasons PDSC has been used to model the flow.

Table 1 shows the parameters used in the simulation which was conducted on an axisymmetric mixed grid using radial-weighting. Transient adaptive sub-cells and TVTS were employed. The particle weighting was redistributed periodically during the simulation, to ensure that the total simulated particles did not become too high and thus slow the simulation down.

Supply Pressure, P	100 kPa
Supply Temperature, T_S	293 K
Pulse Range $P_{min} \rightarrow P_{max}$	10→ ~ 1000 Pa
Inlet Orifice Diameter, d_i	1 mm
Substrate Temperature, T_{sub}	800 K
Wall Temperature, T_W	300 K
Injection Time, t_i	0.1 s
Sampling Cells	74,903 (1,090 unstructured)
Molecular Model	VHS Helium
Simulation Particles	Maintained at ~ 1 x 10^6

Table 1. PDSC Simulation Parameters for PP-CVD

To carry out the simulation, a Navier-Stokes solution was used near the nozzle inlet and this provides the inlet boundary condition to the PDSC simulation. The flow in this region is very dense, however it becomes steady very rapidly (within 20μs) and so the use of a Navier-Stokes solution alleviates the computational expense of PDSC. Figure 11 shows the computational domain used in the simulation along with an enlarged region near the nozzle showing the Navier-Stokes region, the grid near the nozzle and the contours of density after 20μs of simulation.

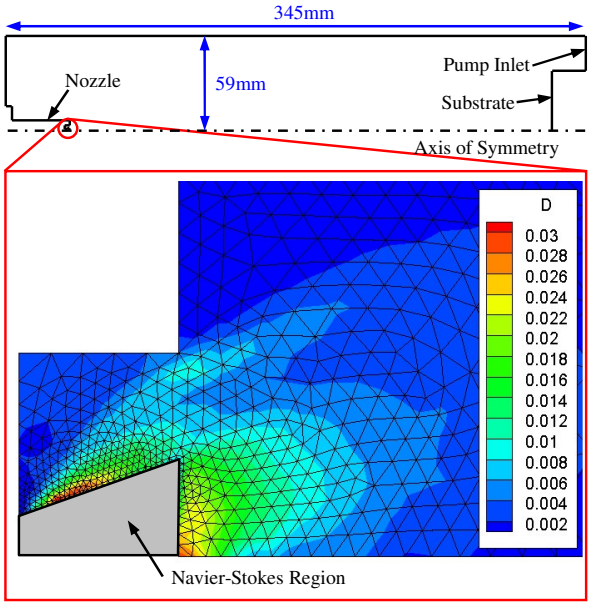

Figure 11. Computational domain for PDSC simulation of PDSC and (insert) nozzle region showing the Navier-Stokes region, mesh and contours of density after 20μs

Figure 12 shows the contours of density during the first 5ms of flow. Note that the scale is selected to show the flow structure within the body of the reactor, rather than near the inlet and that the linear scale varies for each figure.

The computational expense of the simulations remains high, and the simulations remain on-going, so only initial results are presented here. It should be noted that the scatter in the results is high, as time constraints prevented the post-processing with DREAM being completed, however the results give a good qualitative understanding of the PP-CVD flow structure.

The initial expansion is very rapid, and a shock propagates along the length of the reactor, reflecting from the substrate at approximately 0.3ms. After approximately 0.5ms the features of an under-expanded jet become distinguishable including the Mach disc and barrel shock, however the shock reflection from the reactor walls and from the substrate disrupts this structure. Some interesting features include dual barrel shocks formed by the expansion fan from the orifice and the lip of the orifice holder, and the vortex structure visible at 1.5 ms.

As the expansion continues, the highly transient structure begins to die down and a quasi-steady under-expanded jet begins to form. As the pressure in the reactor rises this jet begins to contract, and this process is expected to continue until the jet shuts off and collapses.

The flow density within the reactor is clearly non-uniform during the injection phase. After 5ms a quasi-steady jet actually impinges on the substrate, which will result in non-uniform deposition during the injection phase. The experimental

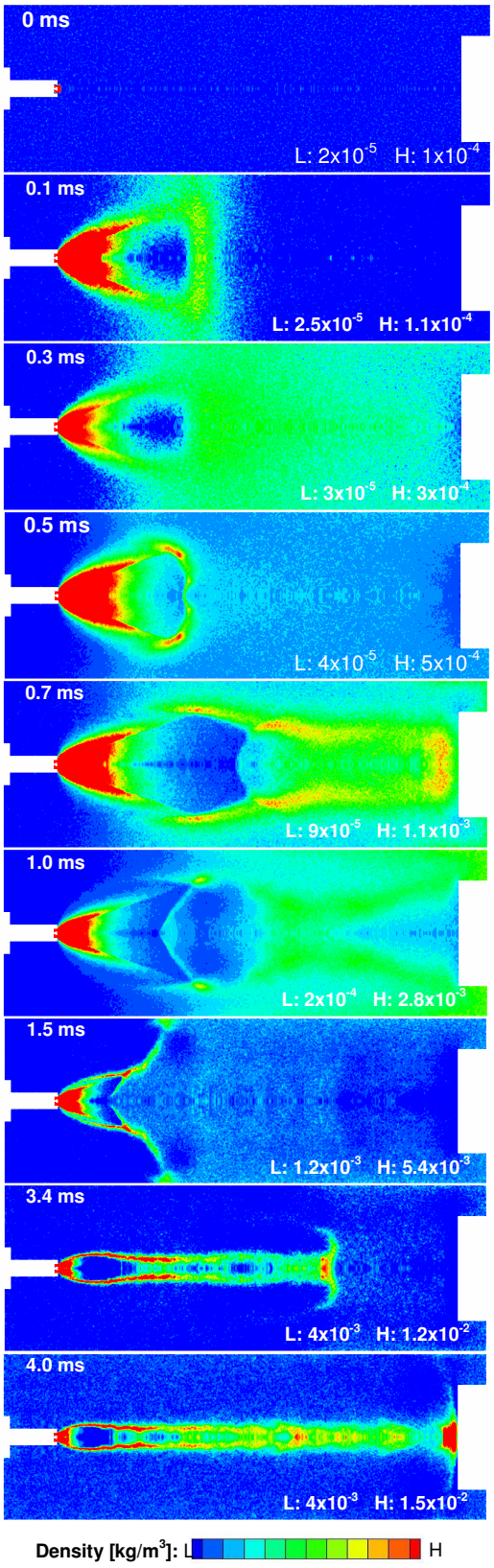

Figure 12. Contours of density [kg/m^3] for a PP-CVD reactor. Note that the scale is linear and varies between plots. The values on the figures give the range of density contours in that figure.

deposition results which show high deposition uniformity seem to contrast with this result, however the following points should be kept in mind:

a. The injection phase is relatively short (~0.1s) compared to the pump-down phase (~20s). At the conclusion of the injection phase, the collapse of the jet structure will result in a uniform distribution of precursor throughout the reactor volume. During the pump-down phase, the negligible convective effects will mean that the precursor will travel to the surface by diffusion alone, resulting in highly uniform deposition, as shown in reference 17.

b. Many of the experimental results for deposition uniformity have been obtained from liquid-supplied reactors for which the flow structure will differ from gas-supplied reactors. Naphthalene sublimation experiments for measuring flow field uniformity carried out in gas-supplied reactors are consistent with the simulation results, as they show extended injection times which result in a reduction in overall flow field uniformity over a pulse duration.

c. As the reactor pressure continues to rise, the quasi-steady jet will continue to retract and reduce in size and therefore will probably not impinge directly on the substrate during the entire injection phase.

Conclusions

Unsteady parallel Direct Simulation Monte Carlo (DSMC) code including efficient sampling techniques and transient-adaptive sub-cells has been developed. The code has been verified using several test cases including shock tube flow and unsteady Couette flow and shows good agreement with analytical and alternative numerical solutions. Furthermore, a post processing method called the DSMC Rapid Ensemble Averaging Method (DREAM) has been developed to reduce the statistical scatter in the results.

Several test cases have been conducted, including the simulation of a shock impingement on a 2D wedge and the expansion of an axisymmetric jet in a Pulsed Pressure Chemical Vapour Deposition (PP-CVD) reactor. The simulations show excellent qualitative results and capture the significant features of the flow.

Future work modelling the PP-CVD is proposed. Additional levels of complexity will be added to the model to enable the full simulation of gas-supplied reactors of arbitrary configuration. However, the high computational expense of simulating these reactors means further developmental simulations will be conducted on lower supply pressure reactors (~ 0.1 atm).

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