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14. ABSTRACT

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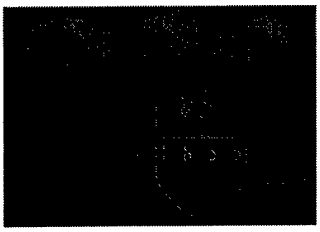

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Two topics are reported on in this interim second year summary report. The first is a continuation of the application of Molecular Dynamics/Quasi-Classical Trajectory Modeling to Direct Simulation Monte Carlo calculations (DSMC) and the second is the introduction of homogeneous condensation to the modeling of space-plumes of hydrazine thrusters. The latter research topic demonstrates the first time homogenous condensation has been modeled directly in the DSMC method.

1 MD/QCT Applied to Small Thruster Space Plume Flows

1.1 Discussion of Problem:

Last year we reported on the use of the Molecular Dynamics/Quasi-classical Trajectory method to model contamination from onboard small reaction control thrusters (RCS). Figure 1 shows a schematic of a thruster firing perpendicular to the vehicle velocity. Figures 2 and 3 show DSMC calculations of the OH number density in the flowfield formed from the combined plume thruster and vehicle at free stream conditions of 80 and 120 km altitude,

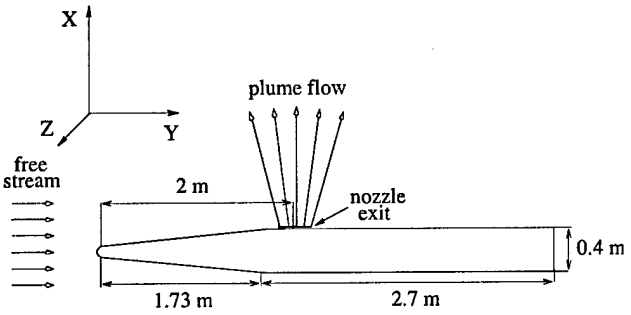

Figure 1: Schematic of a small RCS firing.

respectively. The vehicle velocity in both cases is 5 km/s. Comparisons of Figs. 2 and 3 also show that the aerodynamic features of the flowfields change significantly from 80 to 120 km. At 80 km many continuum-like features, such as a normal plume-shock, may be observed. By 120 km, however, the flow is much more rarefied and the shock structure has been replaced by a more diffuse plume-atmospheric interaction region. Both figures also show that there are chemical reactions at both altitudes and they provide a sensitive metric for flow structure changes. In order to have confidence in the flow modeling we need to understand if the chemical reaction model is adequate for hypervelocity collisions.

Table 1 lists the chemical reactions in a thruster side-jet plume - atmospheric interaction system that either produce or consume OH.[1] The first three reactions involve the dissociation of water (found in the plume) by free stream constituents N_2, O_2, and O. The ratio of these atmospheric free stream constituents changes with altitude, so that the relative importance of each of these three chemical reactions will also depend on altitude. For altitudes of 80 km and below, N_2 and O_2 are the major constituents, whereas, for altitudes above 100 km, atomic O is more abundant. The last four reactions are exchange reactions, again, between free stream and plume constituents. Of the exchange reactions, O + H is less important, because there insufficient H in the plume. The following two exchange reactions involving collisions of O with H_2 and HCl are potentially important because both H_2 and

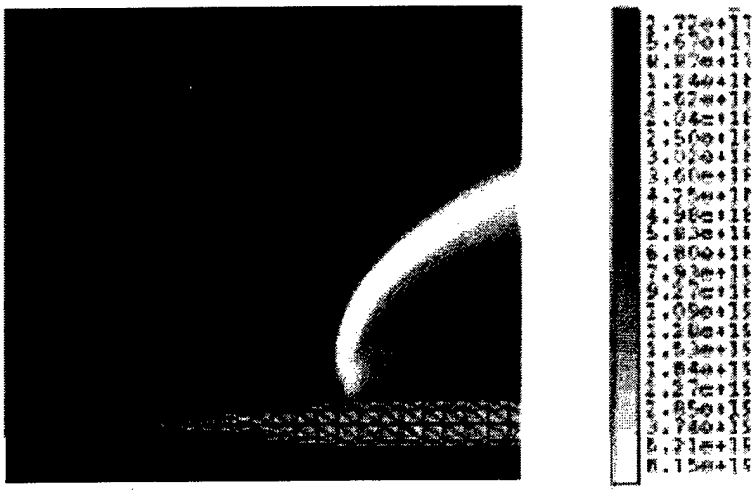

Figure 2: DSMC calculated OH number density contours at 80 km, 5 km/s.

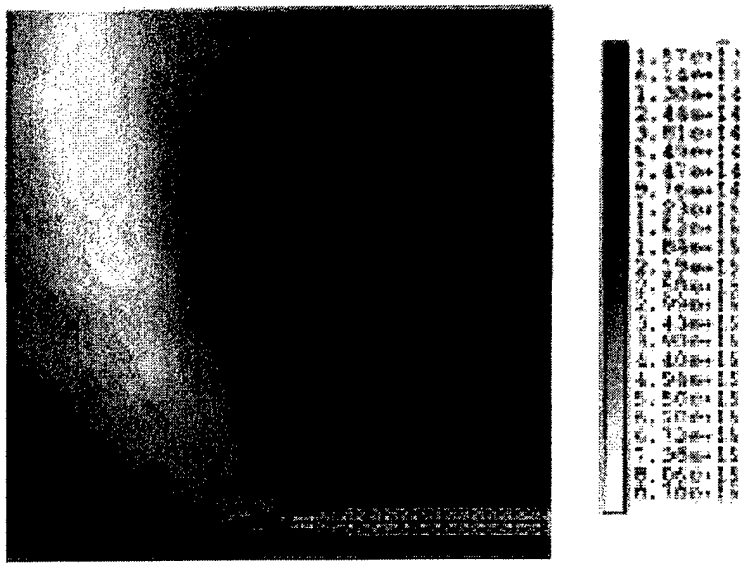

Figure 3: DSMC calculated OH number density contours at 120 km, 5 km/s.

HCl are present in relatively large mole fractions of divert solid propellant motors. However, if we compare the activation energies for these two reactions (E_r, given in Table 1) we can see that the activation energy for the HCl reaction is significantly lower, suggesting that this reaction will occur more frequently. Finally, the last reaction in the table, OH + Cl (the reverse of the O + HCl) is less likely to be important at higher altitudes where there are so few collisions in these flows that a reverse reaction requiring the presence of Cl (which would otherwise not be present) is not likely.

In previous work[2] we have studied the modeling of the first six reactions given in Table 1. The flow system studied was a hypersonic bow shock at 5 km/s for altitudes of 80 and 100 km and the Total Collisional Energy (TCE) chemical model of Bird[3] was compared with QCT/MD calculations. It was found that at 80 km the number density of OH computed using either the TCE or MD/QCT models were nearly spatially equal along the stagnation streamline. This is encouraging because the use of the TCE model to obtain reaction probabilities needed for the DSMC flow simulations is much easier than MD/QCT. However, at 100 km, it was found that the OH number density was about a factor of three lower for the MD/QCT model compared to the TCE. This discrepancy is due to the greater thermal nonequilibrium at the higher altitude and the difference in sensitivities of the two chemical models to the different temperatures. The MD/QCT model is less affected by the translational temperature, compared to the TCE model, but it is more sensitive to the reactant internal energy.

For hypervelocity collisions, such as occur in the atmospheric - jet interaction flows, the extension of the reaction rate for the O+ HCl reaction to temperatures larger than 6000-7000 K and the subsequent use of the rate in the TCE model is problematic. Since the total collision cross section, determined by the VHS model in our modeling,[1] weakly depends on temperature, the reaction rate for the O + HCl reaction is larger than the collision rate at temperatures larger than 6000-7000 K. As a result, the reaction probability becomes larger than one for the TCE chemistry model used here. Note that this could be a problem for any other chemistry reaction model that is based on the use of the Arrhenius form for the experimental rate. Since the reaction probability cannot be greater than one,

Table 1: Freestream-plume species reactions that produce OH

Reagents	Products	A	B	E_r
N_2+H_2O	N_2+OH+H	$5.81 \cdot 10^{-15}$	0.0	$7.314 \cdot 10^{-19}$
O_2+H_2O	O_2+OH+H	$1.13 \cdot 10^{-7}$	-1.31	$8.197 \cdot 10^{-19}$
O +H_2O	O +OH+H	$1.13 \cdot 10^{-7}$	-1.31	$8.197 \cdot 10^{-19}$
O+H_2O	2OH	$3.8 \cdot 10^{-21}$	1.3	$1.275 \cdot 10^{-19}$
H + O_2	OH + O	$1.66 \cdot 10^{-16}$	0.00	$1.061 \cdot 10^{-19}$
H_2+O	OH + H	$3.12 \cdot 10^{-16}$	0.00	$9.518 \cdot 10^{-20}$
O+HCl	OH+Cl	$5.6 \cdot 10^{-27}$	2.87	$2 \cdot 10^{-20}$
OH+Cl	O+HCl	$3.1 \cdot 10^{-27}$	2.91	$7 \cdot 10^{-21}$

$k_r = A\,T^B\,exp(-\frac{E_r}{kT})$, A is in m^3/s, and E_r is in J.

the specific implementation used in Ref. [1] was to model a single reaction in all cases, if the reaction probability was larger than one. This essentially causes a much smaller number of reactions to occur in the simulation compared to that governed by the Arrhenius equation. The situation is even more complicated by the fact that flow is in thermal non-equilibrium, which means that the chemical rate may be different from the experimental one obtained under conditions close to equilibrium.

To-date, a careful study and comparison of the TCE versus a more fundamental approach, such as MD/QCT of the chemical reaction probability for the formation of the hydroxyl radical by the reaction of free stream O exchange reactions with HCl has not been performed. Let us consider the impact that the present use of the TCE model for this reaction may have on the side-jet - atmospheric interaction problem. The experimental rate coefficient used in our previous side-jet modeling[1] was measured for temperatures between 350 and 1490 K,[4] which are much lower than the temperatures observed in the plume-shock interaction. The more recent work of Lin *et al*[5] extended the rate measurements up to temperatures of 3200 K which were found to be consistent with the earlier work. However, the high temperature exponent for both sets of measurements (2.87 for the older work and 3.67 for the more recent

one) indicate that the rate coefficient significantly increases at high temperatures as the pre-exponential term in the Arrhenius equation becomes dominant. In addition, the recent quantum calculations for the rate constant of Xie *et al*[6] suggest that these experiments are in error.

1.2 Work in Progress

From the above discussion it is clear that we have to carefully reevaluate the adequacy of the present model for the O+HCl reaction. We need to consider two factors in this reevaluation: (1) the adequacy of the reaction probability and (2) the possibility that other reaction productions are possible.

To accomplish the first task, we need to use a chemical reaction model that is based on the reaction cross section or reaction probability, rather than a chemical rate *for hypervelocity collisions.* Since the chemical reactions occur between high velocity collision partners, we can use a semi-classical (QCT) instead of a fully quantum mechanical method to calculate the chemical reaction cross section, if an adequate potential surface (PES) can be obtained. Note that the recent quantum calculations of the chemical rate of Xie *et al*[6] were performed for gas temperatures of 3,000 K. The quantum calculations were compared with less exact, transition state methods, and were found to agree within 22%. Since our translational temperatures are 30,000 K, in as much as temperature is relevant to the description of a hypervelocity collision, we expect a semi-classical approach to give reasonable results.[1] Recent work in the chemical physics community suggests that there are a number of options available for obtaining a reliable PES.

The second factor in the O + HCl chemical model that has to be considered is that for hypervelocity collisions there is sufficient energy available such that higher energetic channels that are usually ignored should now be included. In other words, the reaction of O + HCl

[1]The work of Xie *et al*[7] also compared quantum and semi-classical calculations, but these were performed for temperatures on the order of 300 K.

actually has two possible outcomes,

$$O(^3P) + HCl(^1\Sigma^+) \rightarrow OH(^2\Pi) + Cl(^2P) \quad (1)$$

$$\rightarrow ClO(^2\Pi) + H(^2S) \quad (2)$$

The second channel forms the ClO system, a stable radical, but it is usually ignored because it plays an insignificant role for the highest temperatures studied to date, $T = 3,000$ K. Its large activation barrier of $\sim$ 53 Kcal/mole justifies that assumption for "normal" temperatures, but not hypervelocity collisions.[2] Therefore, we need to add this reaction to the reaction set shown in Table 1. This requires that for this second reaction, we obtain a chemical rate that could be used in the TCE model or a PES to be used in a MD/QCT calculation.

To improve the chemical model the following research tasks are in progress:

1. The probability of reaction from the TCE model is being recomputed using the improved reaction rate, Eq. (5) of Ref. [6] for Eq. 1, the lower energy process.

2. The probability of reaction from the TCE model will be recomputed using an estimate of the reaction rate for the high energy channel, Eq. 2. Reference [6] suggests that the reaction rate for this channel may be evaluated by calculating the cumulative distribution function as,

$$N(E) = \sum_{\nu} \sum_{j} (2j+1)\, \theta\left(E_{tot} - E_{\theta} - E_{\nu j}^{ClO}\right) \quad (3)$$

and then substituting that result in to the equation below for the reaction rate,

$$k(T) = \frac{1}{hq_r} \int_0^{\infty} N(\varepsilon) \exp\left(-\frac{\varepsilon}{k_B T}\right) d\varepsilon \quad (4)$$

where $E_0 = 1.65$ eV, $N(\varepsilon)$ is the cumulative distribution function, θ is the heavyside function, $k(T)$ is the rate, h is Planck's constant, and q_r is the total reaction molecular partition function. The energy levels of the ClO radical required to evaluate Eq. 3 may be obtained from standard spectroscopic references such as Herzberg. Finally, recent modeling of the OCl+H $\rightarrow$ HCl + O reaction (the reverse of Eq. 2) indicates

[2]For reference, 60 Kcal/mole is approximately 2.5 eV or a temperature of about 30,000 K ($2.5 = k_B T$).

that there are no barriers to this reaction. Therefore, the rate for the desired reaction, Eq. 2, could be obtained by microscopic reversibility. This rate could then be used and compared with the reaction probability obtained from the TCE model.

3. The above rate calculations of Xie *et al*[6] are based on the new PES of Ramachandran and Peterson[8]. The PES for Eq. 1 consists of contributions from two reactions paths through the $^3A''$ and $^3A'$ electronic transition states. The former is a bent geometry and is a lower energy barrier state, almost thermoneutral, with a barrier height of 8.83 kcal/mole. The latter is the higher energy barrier state, with a barrier height of 11.97 kcal/mole (0.5 eV) with a linear geometry. This second state could contribute to the rate at higher temperatures; in fact, Xie *et al*[6] found that at $T = 3200$ K this surface contributed approximately 30% to the rate. The papers to not report the details of the PES, but through communications with Professor Ramachandran and Bowman the functional fit for the PES has been made available for our use in MD/QCT calculations.

4. In the DSMC modeling we seek chemical models sufficiently accurate to represent flow averaged conditions but accurate on a single collision-by-collision basis. The question may well be asked as to whether less accurate, and more readily available PES may also be used. One such possibility is the commonly used London-Eyring-Polanyi-Sato (LEPS) energy surfaces. The use of LEPS surfaces in MD/QCT calculations was discussed by Muckerman[9] and was applied by Persky and Broida[10] to Eq. 1. This approach will be used to obtain the LEPS surface for the high energy channel process. The sensitivity of the flow modeling to the different reaction probabilities obtained by using the LEPS surface versus the surface of Ramachandran[8] will be studied.

2 Modelling of homogenous nucleation in a supersonic plume

2.1 Introduction and Highlights of Research

When a rocket engine exhaust plume expands into a vacuum or a rarefied background gas, the water vapor in the exhaust flow may become saturated or even supersaturated. In the absence of foreign nuclei, homogenous condensation may occur if super-critical embroys form due to a large degree of supersaturation. Once the condensation process begins, it transfers mass from and adds energy to the gas phase, resulting in changes of the flowfield. The changes that occur in the flowfield may result in increased likelihood of plume contamination of space surfaces as well as affect the signature of high altitude rocket plumes. If we consider the MIR thruster hydrazine firings, the water mole fraction at the nozzle exit is about 20%. This is a significant amount and the affect that condensation might have on the DSMC flow as well as molecular and particulate radiation modeling should not be ignored.[11] Therefore, it was felt by the PI that an investigation of the role of condensation into these types of plumes was the next logical step in the general modeling of space plumes for surveillance applications. Our modeling only considers homogenous condensation, and clearly heterogenous condensation must be considered in the future as well.

During this past year we have made very solid progress on this difficult problem. Three conference papers[12, 13, 14] have been given and one will be given at Reno 05. Of these three papers, the first has been submitted to a journal[15] and two others are in preparation. The material will also result in a Ph.D thesis and be continued by a second, new Ph.D student. The highlights of the research that these papers discuss are given below:

- The implementation of classical nucleation theory (CNT) to DSMC which has never been done before and a validation of the numerical method is given. CNT has been previously applied to continuum flow calculations.[16]

- Nucleation is inherently a kinetic process, hence the coupling with a kinetic model for flows, DSMC, is an important step. Prior to this *all* homogenous condensation

modeling for rarefied conditions has been performed for a homogenous, uniform gas. However, we recognize that the use of CNT to model condensation, while challengingly feasible, is not completely consistent with rarefied flows. The main inconsistency is related to the assumption of microscopic reversibility (through the concept of cluster surface tension) used to relate evaporation and condensation rates for different cluster sizes.

- Therefore, analogous to our efforts to improve chemical reaction models in DSMC, we are in the process of using molecular dynamics (MD) to calculate the fundamental probability of condensation and evaporation. In these calculations, we do not need sophisticated or complex PES since the potential energy functions are mostly simpler pair-wise potentials.

- A DSMC calculation in the densest part of the plume would be unnecessarily computationally expensive since condensation does not begin until the pressure has dropped. Therefore in our present implementation we have utilized the concept of a starting surface, whereby we perform a CFD calculation from the nozzle exit plane to approximately two nozzle diameters downstream. Then taking the CFD steady state solution, we construct a starting surface and begin our DSMC calculation at about 1.5 nozzle diameters down stream. However, the DSMC method assumes that all collisions must be binary in nature. We have performed the first MD calculations of an expanding supersonic jet from an orifice and find that in fact tertiary conditions are indeed important as the initiating cluster formation process. Taking this result, and implementing this into DSMC will be attempted in the Reno 05 paper.

- Cluster formation in supersonic jets is not a new phenomena, but there are some important novel aspects about our research. First, the modeling of coupled condensation in supersonic flows requires expertise in multiple computational-flow techniques. Second, the Rayleigh scattering data set of Williams *et al*[17], which we are using, is unique in that it provides cluster **distributions** in supersonic jets. These data have been less visible to the scientific community since most have been published in Arnold Engi-

neering Development Center reports and only portions have been published in journals read primarily by the aerospace community. Moreover, at the time that these works were published, the state-of-the-art in flow modeling and simulation fell short of what would have been necessary to simulate these data.

- Finally, in the course of this research it was found that although the condensation of water is our goal, important and reliable condensation measurements have been made for Ar. Also, the potential for Ar is much simpler than it is for water. Although results are presented for water, we have emphasized the validation of our numerical and physical models by comparison with condensation for Ar.

2.2 Examples of Results to date

Figure 4 shows the results of a calculation we performed that incorporates a CNT description of cluster evaporation, nucleation, and condensation into a DSMC simulation.[12] The figure shows the stagnation pressures and orifice diameters necessary to obtain an average cluster size of $\sim$ 500 as determined by simulation (circles). These are compared with the initial conditions predicted by the well - known empirical scaling laws for condensation in argon supersonic jets (dashed line).[18] The figure shows that this modeling and simulation approach is able to confirm the scaling law. However, our prediction of the cluster size distributions[14] significantly deviated from experimental data.[17] This is consistent with the work of Ohkubo *et al*[19] who also found that the correct prediction of the cluster size **distribution** along with internal and kinetic energy distributions is beyond the capability of the classic approach.

The reasons for the discrepancy between the CNT-based distributions and experimental data are due to problems inherent with CNT and the flow environment of an expanding jet. The former problems include the ambiguous definition of surface energy of small clusters,[20] the negligence of the rotational and translational degrees of freedom of freshly nucleated clusters,[21] and the unrealistic description of vapor-cluster and cluster-cluster interactions.[12] The latter problems are due to the main assumptions underlying the derivation of the nucleation rate which may be violated in rapidly expanding supersonic jets. The

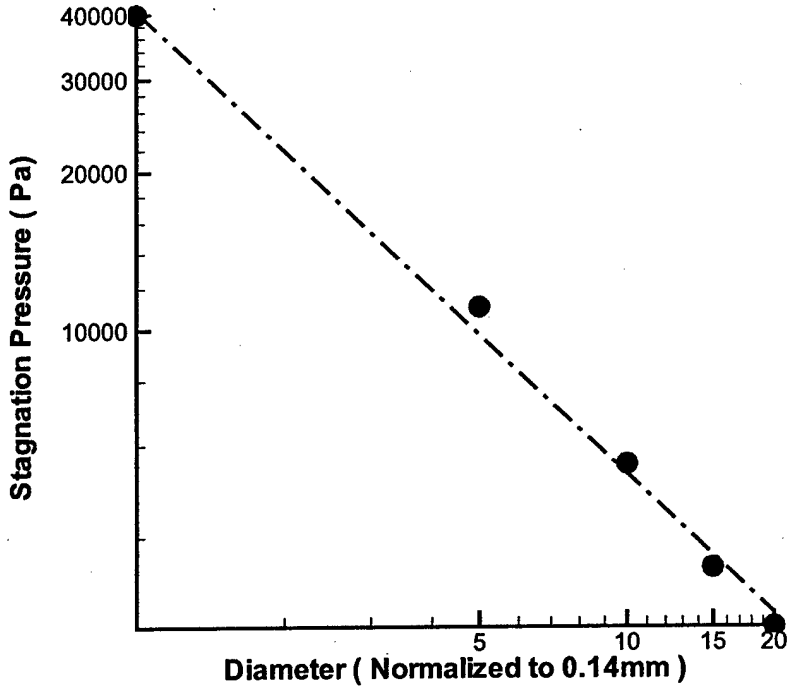

Figure 4: Comparison of the simulation results[12] with the empirical scaling laws for Argon cluster formation in a supersonic jet.[18]

transient time needed for a system to reach the steady state in terms of the unimolecular cluster reactions may be such that the jet macroparameters will significantly change during that time.

The modeling and simulation of expanding supersonic flows covers both transitional and rarefied simulation regions. Figure 5 shows the flow Knudsen number defined with respect to the nozzle diameter as a function of normalized distance from the nozzle exit for an expanding supersonic jet. Since the flow closest to the nozzle is continuum with a typical Knudsen number less than 10^{-3}, the use of DSMC inside this region would be impractical.[12, 15] Also, since the flow is supersonic and information does not propagate upstream, the usual technique for modeling expanding transitional flows is to use a **continuum Navier-Stokes (NS) calculation** to model the dense region of the flow closest to the nozzle.[1] When the

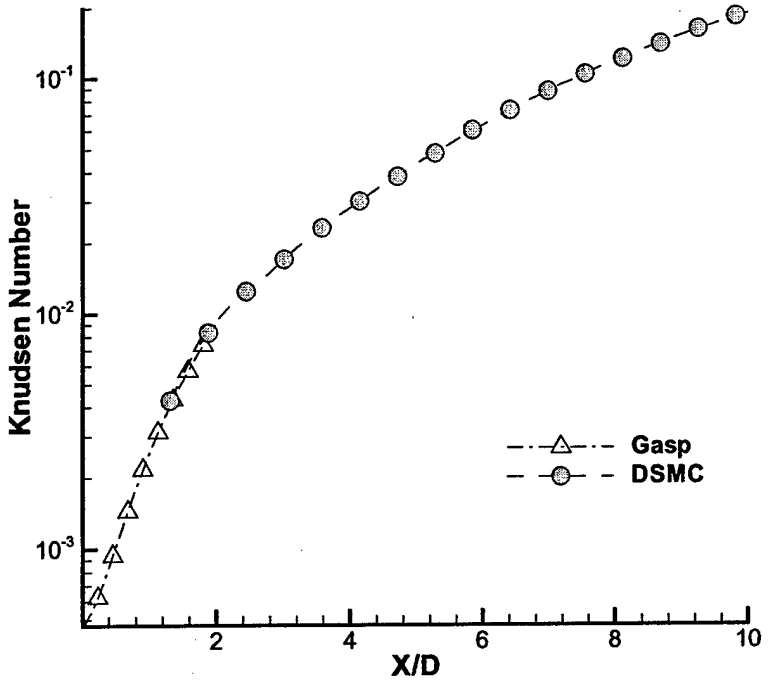

Figure 5: Flow Knudsen number as a function of distance normalized by nozzle diameter, D, from the nozzle exit for an expanding supersonic jet into a vacuum background.

flow has expanded sufficiently down stream (on the order of a few nozzle radii), the Navier Stokes solution can be used to generate a starting surface of temperature, velocity, and species concentration macroparameters to begin the **DSMC calculation**. The numerical solution of the Navier-Stokes equations for viscous gas expansion can be obtained with a computational tool such as the General Aerodynamics Simulation Program (GASP) which uses a finite spatial discretization.[22]

Let us consider the present simulation implementation for an expanding Ar jet with stagnation conditions of 6400 Pa and 170 K, expanding through an orifice with a diameter of 1.4 mm into a vacuum. This corresponds to one of the NS/DSMC simulation cases used to test the semi-empirical scaling law predictions for terminal cluster size shown earlier in Fig. 4. Figures 6 and 7 show the Ar gas plume mass density and temperature contours obtained by

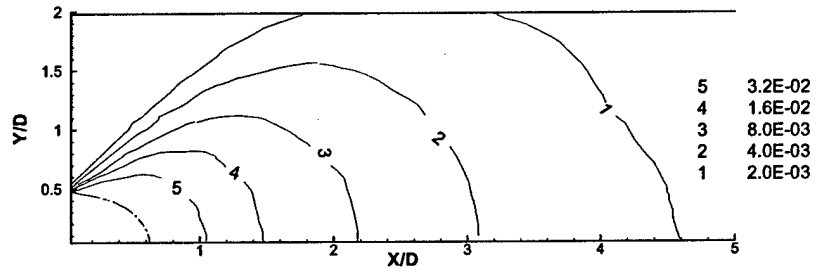

Figure 6: Mass density contours (kg/m^3) for an Argon gas jet expansion obtained from a solution of the DSMC simulation. The orifice is at the origin.

DSMC, respectively. The temperature and mass density contours show the general properties of a rapidly expanding gas into a near vacuum background environment. A free jet forms when a gas expands from a plenum chamber into a vacuum or a low-pressure background gas through a small orifice.[23] The flow velocity rapidly increases with the distance from the orifice, reaching a terminal value at a distance of several nozzle diameters. At the same time, the translational temperature in the jet rapidly decreases with distance which creates an environment for the condensation of clusters.[24] The nucleation region is usually in the transitional to rarefied regime, which occurs at a distance of a few nozzle diameters from the nozzle exit.[17]

The DSMC numerical parameters are $F_{num} = 8 \times 10^6$, a cluster weighting factor of $W = 5.0 \times 10^{-5}$, and the number of simulated molecules and clusters of $N_m = 0.48 \times 10^6$ and $N_c = 0.34 \times 10^6$, respectively. Similar contours may also be obtained for the near-exit fields generated by the NS solutions. The NS and DSMC calculations are performed on grids that spatially overlap so that a starting surface (shown as dashed lines in Figs. 6 and 7) may be generated. Figure 7 shows that there is a smooth connection between the Ar temperature contours obtained with NS/CFD (red) and DSMC (black). This procedure ensures the DSMC solution is independent of the specific starting surface location. The starting surface, typically composed of ~ 500 segments, is constructed from the NS solution for a typical Mach number ≤ 2.5.

In addition to the usual collision processes among the monomers, the processes of cluster

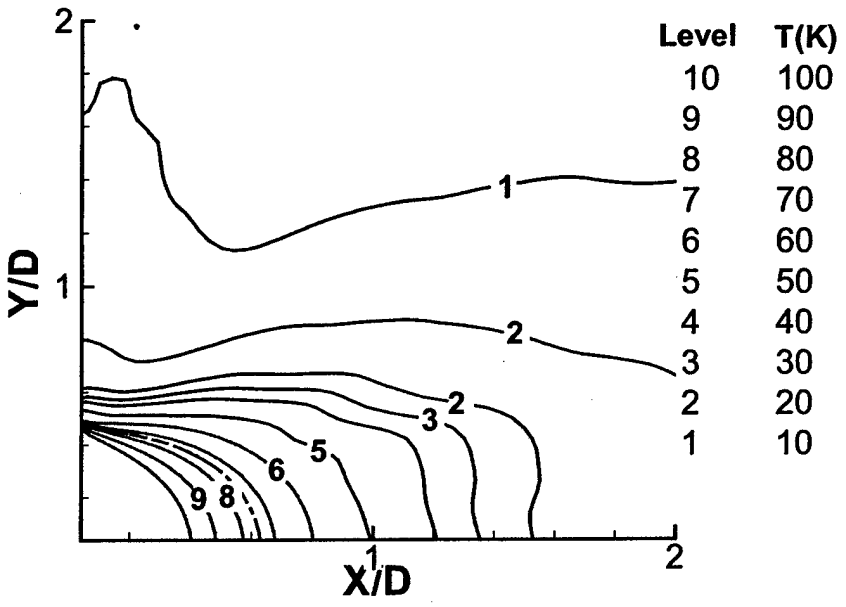

Figure 7: Argon gas temperature contours (K) for an Argon gas jet expansion obtained from a DSMC simulation. The orifice is at the origin. See text for further explanation.

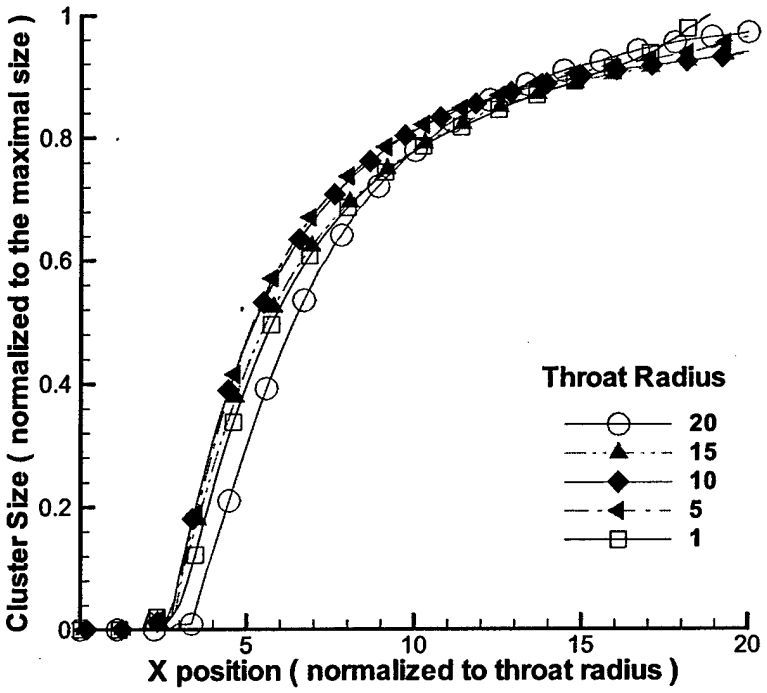

Figure 8: Argon cluster growth along the centerline of the DSMC simulation for an orifice diameter of 0.14 mm.

formation, growth and decay were incorporated into our DSMC calculations in a manner similar to chemical reactions. The reaction rates used are the CNT rates of cluster nucleation, condensation, and evaporation. Figure 8 shows the average cluster growth along the plume centerline for stagnation conditions of 6400 Pa and various temperatures.[12] It can be seen that cluster growth weakly depends on the initial conditions, which is consistent with Hagena's derivation of the scaling laws.[18] The small differences between the curves may be attributed to the nonlinear effects of the coupled evaporation and condensation processes accompanying the growth of clusters.

Finally, Fig. 9 shows a comparison of Mach number contours of a hydrazine rocket plume computed with and without water condensation.[12, 15] The horizontal axis represents the distance (m) from the nozzle along the plume axis. To model water condensation, a weighting

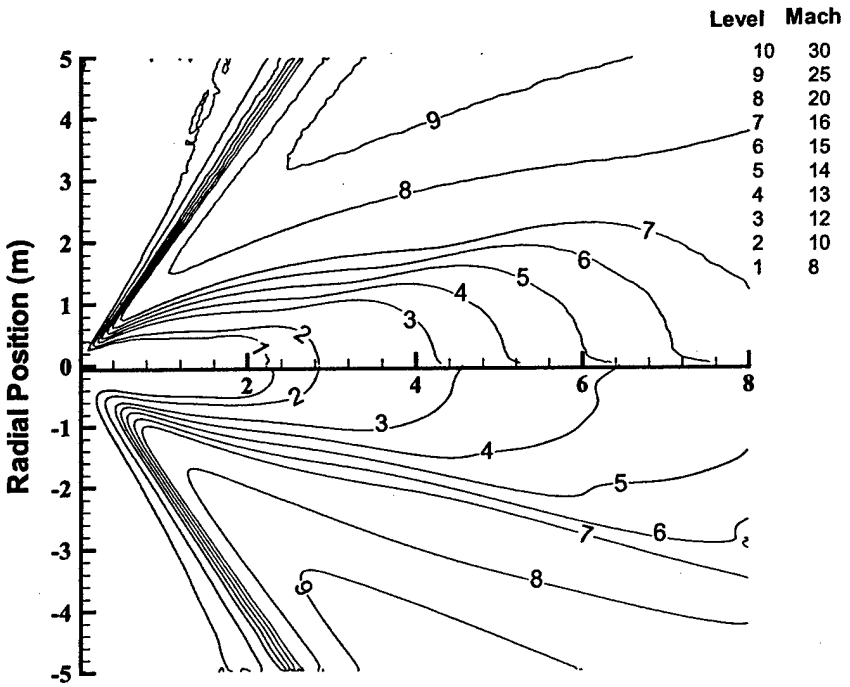

Figure 9: The Mach contours of the rocket plume with water condensation neglected (upper part) and modeled with CNT by a weighing-scheme DSMC (lower part).

factor for water clusters seven orders of magnitude higher than the weighting factor for the main plume species was used. Figure 9 shows that the DSMC method without trace species (cluster) weighting would have not revealed any difference in the Mach contours. Moreover, despite the significant difference in the concentrations, the presence of the trace species may affect the main flow, as is shown on Fig. 9.

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