MONTE CARLO SIMULATION FOR SOOT DYNAMICS

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A new Monte Carlo method termed Comb-like frame Monte Carlo is developed to simulate the soot dynamics. Detailed stochastic error analysis is provided. Comb-like frame Monte Carlo is coupled with the gas phase solver Chemkin II to simulate soot formation in a 1-D premixed burner stabilized flame. The simulated soot number density, volume fraction, and particle size distribution all agree well with the measurement available in literature. The origin of the bimodal distribution of particle size distribution is revealed with quantitative proof.

Key words: *Monte Carlo simulation, soot*

Introduction

Soot formation is an important and constantly studied aspect of combustion, which has practical significance in the production of technical carbon as well as in the combustion efficiency [1] and human health. Among generally three types of numerical methods to solve soot population dynamics, *i. e.*, direct discretization of the controlling equations, various methods of moments (interpolative, quadrature/direct quadrature [2], Taylor-expansion moment [3, 4] and hybrid closures), and stochastic simulations [5, 6]. Understanding the mechanisms of soot formation is a long-standing challenge in combustion research. Stochastic simulations can provide very detailed information on soot morphology, and have the best flexibility to accommodate various physical models of soot nucleation, coagulation and surface growth. This work develops a new Monte Carlo (MC) method, and adopts special strategies to couple the method with the 1-D premixed flame code PREMIX [7] to simulate soot formation in a premix burner stabilized flame.

Monte Carlo simulation for soot

The kinetic model of soot formation with detailed chemistry from Appel *et al.* [2] is used for the present work. Soot nucleation corresponds to the formation of the first soot particle from the collision and subsequent coalescence of two polycyclic aromatic hydrocarbons (PAH) dimers. Condensation corresponds to the deposition of a PAH dimer on the surface of a soot particle. Coagulation corresponds to the collision between two soot particles to form a new particle which has volume equal to the total volume of the two colliding particles. The soot surface chemical reaction is modeled through the hydrogen abstraction – carbon addition (HACA) mechanism [8, 9]. Comb-

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like frame Monte Carlo (CFMC) uses operator splitting techniques to solve the soot particle size distribution evolution equations. Details of the scheme are discussed in [10] in detail. The coupling between the gas phase solver Chemkin II and CFMC for soot can be described as:

$$C_{\text{gas}} = \text{Chemkin}(S_{\text{soot}}), \quad S_{\text{soot}} = \text{CFMC}(C_{\text{gas}}), \tag{1}$$

where C_{gas} stands for gas phase concentration, and S_{soot} is the consumption of gas species due to soot formation. This work focuses on a burner stabilized 1-D steady flame. The PREMIX code [7] in Chemkin II is used to solve the gas phase, which uses a modified Newton's method to solve the underlying boundary value problem [11]. The Newton's method has to evaluate the Jacobian $\partial C_{gas}/\partial S_{soot}$. However, S_{soot} has stochastic fluctuation from the MC simulation. It is known [12] that Newton's method is very difficult to find the solution of a problem with strong stochasticity because the Jacobian matrix is very sensitive to the stochastic fluctuation. To overcome this difficulty, two strategies have been implemented. Firstly, S_{soot} is averaged over the values obtained by a number of independent CFMC simulations. Secondly, S_{soot} is frozen for some iteration steps when solving eq. (1) by Newton's method. This freezing strategy makes the Jacobian evaluation encounter even less stochastic error, and meanwhile saves great amount of computation, since the time consuming CFMC is called far more less frequently. This strategy also increases the stability of the Newton's method, because freezing S_{soot} greatly decreases the coupling between the two equations, which is pretty much like a relaxation scheme.

Numerical results and discussion

Figure 1 shows simulation results of the mean and standard deviation (std) of pyrene consumption rate owing to soot formation in a premixed flame (see next section for the details of the flame). The mean rates using 1 *k* and 10 *k* particles (both over 20 independent simulations) are almost identical, while the rate using 0.1 *k* particles (over 100 simulations) deviates slightly. The ratio of std between 1 *k* and 10 *k* is very close to the theoretical value 3.162, which says the stochastic error of MC simulation is proportional to the inverse of the square root of the particles number. However, the std ratio between 0.1 *k* and 1 *k* is nearly two times of the theoretical value 3.162. It is believed that the additional stochastic error comes from the re-sampling in CFMC. When using larger number of particles, the stochastic error in re-sampling becomes negligible. So, using 1 *k* particles is an optimal choice, which needs only around 3 percent computation of 10 *k* case to reach the same precision for the mean value. Abid *et al*. [13] measured the soot PSD in a series of premixed ethylene flames with very high fidelity. The measurement is a good benchmark to test the ability of CFMC's prediction of soot formation in a flame. The setting of the simulation is the same as the C2 flame in [13]. Figure 2 shows the profile of soot volume faction. The solution using the method of moments with interpolative closure [8] is also added for comparison. Both simulations give comparable results with the measurement. The difference between simulations and measurement is within a factor of 3, which adds additional proofs to the high precision of the mechanism used [8].

At the early stage, nucleation dominates and the PSD is of uni-modal. Then the PSD gradually evolves to be of bi-modal. This phenomenon can be explained qualitatively. As revealed by the CFMC simulations, soot particle number density should keep increasing along the flame owing to nucleation. At a specific height, the number density reaches the maximum, since after then nucleation rate decreases owing to condensation consumes more and more PAH. Condensation and surface reaction are usually monotonic function of soot size, which means that all soot particles of difference size will grow in a similar way. Under this condi-

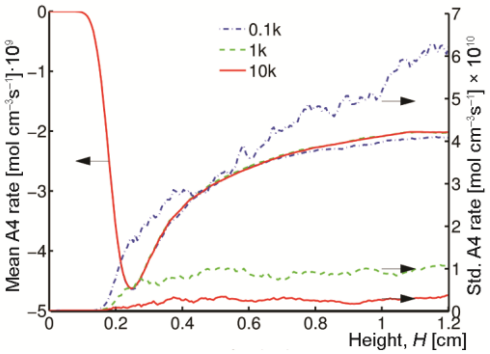

Figure 1. The simulated mean and std of pyrene consumption rate owing to soot formation

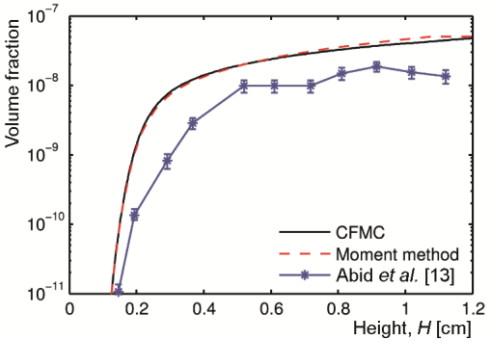

Figure 2. Profiles of soot volume fraction

tion, PSD is of uni-modal, and the peak shifts towards larger size direction as the height increases. On the other hand, coagulation reduces the number density of soot particles, and the coagulation rate between two particles increases as the size ratio (big to small particles) increases. It implies that small particles will be removed more quickly because they coagulate with very big particles. Remind that nucleation constantly generates the smallest particles, then the number of particles slightly larger than the nucleated ones will decrease most sharply. That is why a trough in PSD appears around 3 nm.

On the other hand, the difference between CFMC and moment method is negligible. Despite the fact that moment method uses averaged values of soot size and surface area to model the soot dynamics, it seems to be able to predict well on the volume fraction, compared to the more delicate CFMC, which takes account of individual soot size and surface area. Figure 3 shows the soot particle number density. Both simulations over predict the number density but catch the trend well. CFMC gives closer result compared to the measurement than the method of moments. One great virtue of MC methods over methods of moments is that MC can provide the soot PSD. Figure 4 compares the PSD from CFMC and the measurement. CFMC correctly predicts the bi-modal PSD at height 0.65 cm. The PSD at other heights also compares well with the measurement (not shown here).

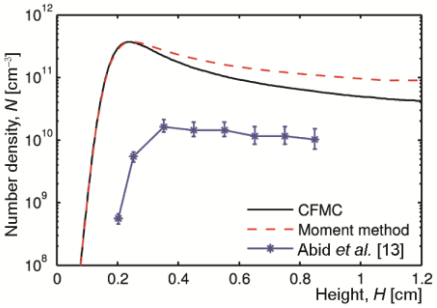

Figure 3. Profiles of soot number density

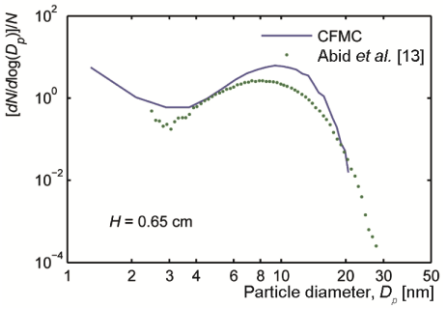

Figure 4. PSD at height H = 0.65 cm

Conclusions

A new MC method termed CFMC is developed to simulate soot dynamics. CFMC uses an operator splitting technique. To achieve high computational efficiency and low sto-

chastic error, CFMC requires to re-sample the PSD to keep the number of simulation particles in an appropriate range. CFMC is coupled with the gas phase solver Chemkin II to simulation soot formation in 1D burner stabilized premixed flame. The coupling is implemented by averaging over a number of independent MC simulations to obtain smooth gas consumption due to soot formation, and by freezing the consumption for some iterations in the Newton's method when solving the gas phase equations.

The simulated soot number density, volume fraction, and PSD all agrees well with the measurement available in literature. The reason for the formation of the bi-modal PSD is also discussed.

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References

[1] Yu, M. Z., Lin, J. Z., Chan, T. L., Numerical Simulation of Nanoparticle Synthesis in Diffusion Flame reactor, *Powder Technology*, *181* (2008), 1, pp. 9-20
[2] Yu, M. Z., Lin, J. Z., Chan, T. L., Effect of Precursor Loading on Non-Spherical TiO_2 Nanoparticle Synthesis in a Diffusion Flame Reactor, *Chemical Engineering Science*, *63* (2008), 9, pp. 2317-2329
[3] Yu, M. Z., Lin, J. Z., Chan, T. L., A New Moment Method for Solving the Coagulation Equation for Particles in Brownian Motion, *Aerosol Science and Technology*, *42* (2008), 9, pp. 705-713
[4] Yu, M. Z., Lin, J. Z., Taylor-Expansion Moment Method for Agglomerate Coagulation due to Brownian Motion in the Entire Size Regime, *Journal of Aerosol Science*, *40* (2009), 6, pp. 549-562
[5] Nie, D. M., Lin, J. Z., A Fluctuating Lattice Boltzmann Model for Direct Numerical Simulation of Particle Brownian Motion, *Particuology*, *7* (2009), 6, pp. 501-506
[6] Nie, D. M., Lin, J. Z., A Lattice Boltzmann-direct Forcing/Sictitious Domain Model for Brownian Particles in Fluctuating Fluids, *Communications in Computational in Physics*, *9* (2011), 4, pp. 959-973
[7] Kee, R. J., Grcar, J. F., Smooke, M. D., PREMIX: A Fortran Program for Modeling Steady Laminar One-Dimensional Premixed Flame, *Technical Report SAND85-8240*, Sandia National Laboratories, Albuquerque, N. Mex., USA, 1985
[8] Appel, J., Bockhorn, H., Frenklach, M., Kinetic Modeling of Soot Formation with Detailed Chemistry and Physics: Laminar Premixed Flames of C2 Hydrocarbons, *Combustion and Flame*, *121* (2000), 1-2, pp. 122-136
[9] Frenklach, M., Wang, H., Detailed Modeling of Soot Particle Nucleation and Growth, Proceedings of the Combustion Institute, 23 (1990), pp. 1559-1566
[10] Zhou, K., Bisetti, F., Operator Splitting Monte Carlo: A Highly Efficient Simulator for Aerosol Dynamics, *Journal of Computational Physics*, (2012), (in progress)
[11] Grcar, J. F., The Twopnt Program for Boundary Value Problems, *Technical Report SAND91-8230*, Sandia National Laboratories, Albuquerque, N. Mex., USA, 1992
[12] Spall, J. C., *Introduction to Stochastic Search and Optimization: Estimation, Simulation, and Control*, John Wiley & Sons, Inc. 2003
[13] Abid, A. D., Heinz, N., Tolmachoff, E. D., On Evolution of Particle Size Distribution Functions of Incipient Soot in Premixed Ethylene-Oxygen-Argon Flames, *Combustion and Flame*, *34* (2008), 4, pp. 775-788
[14] Frenklach, M., Harris, S. J., Aerosol Dynamics Modeling Using the Method of Moments, *Journal of Colloid and Interface Science*, *118* (1987), pp. 252-261

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