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Implementation of an advanced fixed sectional aerosol dynamics model with soot aggregate formation in a laminar methane/air coflow diffusion flame

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An advanced fixed sectional aerosol dynamics model describing the evolution of soot particles under simultaneous nucleation, coagulation, surface growth and oxidation processes is successfully implemented to model soot formation in a two-dimensional laminar axisymmetric coflow methane/air diffusion flame. This fixed sectional model takes into account soot aggregate formation and is able to provide soot aggregate and primary particle size distributions. Soot nucleation, surface growth and oxidation steps are based on the model of Fairweather et al. Soot equations are solved simultaneously to ensure convergence. The numerically calculated flame temperature, species concentrations and soot volume fraction are in good agreement with the experimental data in the literature. The structures of soot aggregates are determined by the nucleation, coagulation, surface growth and oxidation processes. The result of the soot aggregate size distribution function shows that the aggregate number density is dominated by small aggregates while the aggregate mass density is generally dominated by aggregates of intermediate size. Parallel computation with the domain decomposition method is employed to speed up the calculation. Three different domain decomposition schemes are discussed and compared. Using 12 processors, a speed-up of almost 10 is achieved which makes it feasible to model soot formation in laminar coflow diffusion flames with detailed chemistry and detailed aerosol dynamics.

Keywords: numerical simulation; laminar coflow diffusion flame; soot aggregate formation; fixed sectional aerosol dynamics model; parallel computation

1. Introduction

Soot formation is an extremely challenging problem, because many sooting-related chemical and physical processes must be addressed when it is accounted for. These processes include nucleation to form nascent soot particles, coagulation of particles to form the fractal-like soot aggregates, surface growth and oxidation of particles [1]. In addition, these processes are strongly coupled with the gas-phase chemistry, flow field and temperature field. Nevertheless, owing to the important impacts of soot particles on the efficiency of heat transfer, air quality and human health [2], it is very important to gain fundamental understanding of the soot formation phenomenon.

The development of modelling capabilities of soot formation is an indispensable part of our overall effort to gain a fundamental understanding of various processes related to soot

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formation. Much progress has been achieved in this respect in the past years. Detailed chemical kinetic mechanisms describing the breakdown of hydrocarbon fuels and formation of soot precursors have been proposed [3–7]. In addition, different methods describing the evolution of soot particles under simultaneous nucleation, coagulation, surface growth and oxidation have also been proposed [5, 8–11]. Sectional soot aerosol dynamics models [5, 11] can provide the mean properties and the size distribution of soot particles. Conventional sectional models solve for only one variable per section such as soot mass fraction [5], which is not adequate for modelling soot aggregate formation. To model the formation and coagulation of the fractal-like soot aggregates, Park *et al.* [11] developed an advanced fixed sectional model which solves two equations (number densities of aggregates and primary particles) per section. This model has been successfully used to model soot formation in plug flow reactors [11] and shock tubes [12]. However, no attempt has been made to apply this model to multi-dimensional laminar diffusion flames.

A number of diffusion flame experimental studies [13–17] have looked at the detailed structure of soot aggregates including the primary particle diameter, primary particle number density, number of primary particles per aggregate and the fractal dimension of soot aggregates. Modelling soot aggregate nanostructure and size distribution in laminar diffusion flames requires adding to the flame code the detailed fixed sectional aerosol dynamics model [11, 12]. However, implementation of the fixed sectional soot aerosol dynamics model [11, 12], which is computationally expensive in its own, into a multi-dimensional flame code significantly increases the computational cost. Parallel computation, which divides the whole problem into smaller tasks and solves them simultaneously, offers a potential solution to the problem of large computational cost. Furthermore, as PC becomes cheaper, Beowulf-type high-performance parallel computing clusters become more accessible. This fact and the lack of detailed treatment of aerosol dynamics especially accounting for soot aggregation and size distribution in the current modelling of soot formation in laminar diffusion flames motivated the present authors to develop a parallel diffusion flame code which incorporates the fixed sectional aerosol dynamics model [11, 12] along with detailed gas-phase chemistry and can provide converged solution within a reasonable amount of time.

The objectives of this paper are to present the implementation strategy of the fixed sectional aerosol dynamics model [11, 12] in a laminar axisymmetric coflow diffusion flame and the development strategy of the parallel flame code. The paper is organised as follows. The problem formulation, numerical method and parallelisation methodology are first presented. Then, the performance of the parallel code is demonstrated through its application to the modelling of a laminar coflow methane/air diffusion flame [5] with the results analysed and compared to available experimental data from the literature. Finally, the efficiency of the parallel code is analysed and conclusions are drawn.

2. Problem formulation

2.1. Physical problem

A schematic of the diffusion flame burner and the resultant flame is shown in Figure 1. Gaseous fuel is delivered upwards through the central vertical fuel tube and air is delivered through the co-annular region. After ignition and stabilisation, a laminar coflow diffusion flame is established. We only consider the over-ventilated flame in this study. The shaded rectangular area in Figure 1 shows schematically the computational domain. Owing to the axisymmetric property of this flame, it is modeled in the two-dimensional cylindrical coordinates to save central processing unit (CPU) time.



Figure 1. Schematic of the burner and flame.

The fully coupled elliptic conservation equations for mass, momentum, gaseous species mass fractions, sectional soot aggregate and primary particle number densities, and energy in the twodimensional axisymmetric cylindrical coordinate system are solved. The sectional soot equations are introduced below while the detailed forms of the other governing equations can be found in the previous publication [18]. It is noted that the interactions between soot formation and gas-phase chemistry through soot nucleation, surface growth and oxidation processes and the interaction between soot formation and flame temperature through radiative heat transfer are accounted for in the governing equations.

2.2. Soot nucleation, growth and oxidation sub-models

Initially, the detailed soot nucleation, surface growth and oxidation sub-models developed by Appel *et al.* [6] were implemented to model the soot formation in the laminar coflow methane/air diffusion flame [5], which is the target flame of the current study. These detailed soot sub-models include polycyclic aromatic hydrocarbon (PAH) chemistry and the hydrogen-abstraction/carbon-addition (HACA) mechanism responsible for the growth of PAHs and soot particles. Unfortunately, the peak soot volume fraction was under-predicted by almost two orders of magnitude. This is not unexpected as Roesler *et al.* [19] states that methane has other PAH growth pathways which involve odd-carbon-numbered species in addition to the HACA mechanism in [6] and that these pathways are currently not well understood.

We found that satisfactory results could be obtained with the acetylene-based soot sub-models of Fairweather *et al.* [20]. In this study, the soot model of Fairweather *et al.* [20] is chosen to represent the soot nucleation, growth and oxidation reactions. We think this is sufficient for the current study based on the following considerations. (1) Despite the fact that the soot model of Fairweather *et al.* [20] is less fundamental as compared to the PAH-based soot model in [6], this model seems to be able to capture the major characteristics of the soot formation in

this methane/air flame [5]. (2) The scope of this study is not to investigate certain chemical aspects of the soot formation problem. Rather, the main focus of this study is to present the implementation strategy of the fixed sectional model [11, 12] into a two-dimensional laminar diffusion flame and the development strategy of the parallel flame code. As such, the choice of the simplified soot model of Fairweather *et al.* [20] does not undermine the scope of this study.

In the soot model of Fairweather *et al.*, nucleation of soot particles is assumed to take place by the reaction of acetylene, an important intermediate sooting species

$$C_2H_2 \rightarrow 2 C_{(s)} + H_2 \tag{R1}$$

with the rate given as

$$r_1 = k_1(T)[C_2H_2]$$
(1)

In equation (1), T and $[C_2H_2]$ are temperature and C_2H_2 mole concentration respectively. It is emphasised that reaction R1 is a highly simplified situation since it has been generally agreed that growth of PAHs leads to nucleation of soot particles [21].

Soot surface growth is assumed to proceed by the addition of acetylene to the surface of soot particles, which again can be regarded as a simplification of the generally accepted HACA soot growth model [9], via the following step

$$C_2H_2 + nC \rightarrow (n+2)C + H_2 \tag{R2}$$

The reaction rate for surface growth is written as

$$r_2 = k_2(T)f(A_s)[C_2H_2]$$
(2)

where $f(A_s)$ denotes the functional dependence on A_s which is the total soot surface area summed over all sections per unit volume of gaseous mixture. Linear function of $f(A_s)$ is assumed, i.e. $f(A_s) = A_s$.

Oxidation of soot particles by O_2 is included. It is assumed that soot oxidation by O_2 proceeds as

$$0.5 \text{ O}_2 + \text{C} \to \text{CO} \tag{R3}$$

with the rate expression given as

$$r_3 = k_3(T)A_s[O_2] \tag{3}$$

Neoh *et al.* [22] investigated the soot oxidation process in flames and found that the oxidation owing to both O_2 and OH is important, depending on the local equivalence ratio. Therefore, soot oxidation by OH is included in the soot oxidation mechanism. In this study, soot oxidation owing to OH is accounted for by the Fenimore and Jones model [23, 24] with the reaction path and rate

<i>k</i> _i	А	b	E	Ref.
k_1 k_2	1.35E+06 5.00E+02	0 0	4.10E+04 2.40E+04	[20] [20]
$k_3 k_4$	1.78E+04 1.27E+03	-0.5	0.00E+00	[20] [23, 24]

Table 1. Reaction rates in the form of $k_i = AT^b e^{-E/RT}$ (units in kg, m, s, kmol, kcal and K).

as

$$OH + C \rightarrow CO + H$$
 (R4)

$$r_4 = \varphi_{\rm OH} k_4(T) A_s P_{\rm OH} / M W_{\rm C} \tag{4}$$

where $\varphi_{\text{OH}} = 0.1$ is the collision efficiency; P_{OH} is the partial pressure of OH in atm and is equal to its mole fraction X_{OH} for the atmospheric flame studied here; MW_{C} is the molecular weight of carbon.

The reaction rates of nucleation, surface growth and O_2 oxidation steps are kept the same as those of Fairweather *et al.* [20] and all the reaction rates are provided in Table 1.

2.3. Fixed sectional aerosol dynamics model

The fixed sectional model [11, 12] implemented in this paper uses the classical sectional description of the aerosol dynamics equations based on the fixed pivot approach [25]. The mass range of the fractal-like solid soot aggregates is divided into a number of discrete sections. Each section has a fixed, i.e. prescribed representative mass. According to their mass, soot aggregates are assigned to individual sections. Each section has a transport equation, or population balance equation, for the number density of soot aggregates. The nucleation step connects the gas-phase incipient species with the solid soot phase. The incipient soot particles are assumed to be spherical and belong to the first section. By coagulation or surface growth, lower section particles move to higher sections. On the other hand, higher section particles move to lower sections or become gaseous products by oxidation.

One major advantage of the current fixed sectional model [11, 12] is its capability to model the formation of soot aggregates. As has been observed, the fractal-like soot aggregate is comprised of almost equally sized primary particles firmly connecting each other [13, 26]. The fractal dimension of flame-generated soot aggregates does not vary significantly from 1.8 [14, 15, 27]. In the current fixed sectional model, each aggregate is assumed to be comprised of equally sized spherical primary particles and a constant fractal dimension of 1.8 is assumed for the soot aggregates. A constant fractal dimension has been used extensively in modelling the simultaneously occurring particle nucleation, coagulation, and surface growth processes. To model the soot aggregate formation, one additional variable-the number density of primary particles-is solved for in each section. In each section, all the soot aggregates are assumed to be identical. By knowing the number densities of aggregates and primary particles in one section, the average number of primary particles per aggregate in that section is known. Since the mass of a single aggregate in the section under consideration is prescribed and known, the volume of the aggregate, the diameter of primary particles forming the aggregate, and the total surface area of the aggregates in that section are also known. Similarly, such information is known for all the other sections. It is noted that all soot particles in the first section are spherical nascent soot particles or monomers.

Thus, aggregates and primary particles in the first section are essentially the same both meaning spherical nascent monomers.

In addition to the gas-phase governing equations, the following sectional transport equations for the number densities of soot aggregates and primary particles are formulated and solved. The formulation of the sectional transport equations follows the same methodology of Kennedy et al. [28], i.e. the transport equations have components attributed to convection, thermophoresis, nucleation (nu), coagulation (co), surface growth (sg) and oxidation (ox). Many researchers [28– 31] have found that soot transport is usually dominated by flow convection and thermophoresis while the Brownian diffusion of soot particles is small and negligible. As in reference [28], we neglect the small influence of Brownian diffusion of soot aggregates in the current study. Therefore, the velocity of a soot aggregate is assumed to be the sum of the gas velocity and the thermophoretic velocity. It should be noted that implicit in this assumption is the assumption that all soot aggregates can follow the accelerating hot gas flow in the current laminar diffusion flame [5]. This is justified as follows. Generally, suspended particles may not be able to follow the motion of an accelerating gas owing to their inertia. However, this inertia effect is not important for submicron particles [32]. Soot particles are typically submicron particles. In fact, the volume equivalent diameter of the largest soot aggregates from the modelling of the current laminar methane/air flame [5] is about 0.5 μ m (see Figures 16(a) and 16(b) later). Moreover, these largest aggregates contribute negligibly to the total number or total mass of soot aggregates (see Figures 16(a) and 16(b)). In other words, most of the aggregates found in the current laminar methane/air flame are much smaller than 0.5 μ m. Thus, the inertia effect of the soot aggregates is negligible and it is reasonable to assume that all the soot aggregates can follow the accelerating gas flow.

Mathematically, the sectional soot transport equations are

$$\rho v \frac{\partial N_j^A}{\partial r} + \rho u \frac{\partial N_j^A}{\partial z} = -\frac{1}{r} \frac{\partial}{\partial r} (r \rho N_j^A V_{Tr,s}) - \frac{\partial}{\partial z} (\rho N_j^A V_{Tz,s}) + \rho \left(\frac{\partial N_j^A}{\partial t} \Big|_{nu} + \frac{\partial N_j^A}{\partial t} \Big|_{co} + \frac{\partial N_j^A}{\partial t} \Big|_{sg} + \frac{\partial N_j^A}{\partial t} \Big|_{ox} \right) (j = 1, 2, ..., SN)$$
(5)

$$\rho v \frac{\partial N_j^P}{\partial r} + \rho u \frac{\partial N_j^P}{\partial z} = -\frac{1}{r} \frac{\partial}{\partial r} (r \rho N_j^P V_{Tr,s}) - \frac{\partial}{\partial z} (\rho N_j^P V_{Tz,s}) + \rho \left(\frac{\partial N_j^P}{\partial t} \Big|_{nu} + \frac{\partial N_j^P}{\partial t} \Big|_{co} + \frac{\partial N_j^P}{\partial t} \Big|_{sg} + \frac{\partial N_j^P}{\partial t} \Big|_{ox} \right) (j = 1, 2, ..., SN)$$
(6)

In equations (5) and (6), *r*, *z*, *v*, *u*, ρ are radial and axial coordinates, radial and axial velocities and mixture density, respectively. N_j^A and N_j^P are *jth* sectional soot aggregate and primary particle number densities in (particles/g-gas); $V_{Tr,s}$ and $V_{Tz,s}$ are the thermophoretic velocities of soot aggregates in the radial and axial directions respectively; *SN* is the total soot section number. The thermophoretic velocity of a soot aggregate is calculated by the following expression [33]

$$V_{Tx_i,s} = -0.55 \frac{\mu}{\rho T} \frac{\partial T}{\partial x_i} \qquad x_i = r, z \tag{7}$$

where μ is the dynamic viscosity of the gaseous mixture.

The terms owing to nucleation, coagulation, surface growth and oxidation in the transport equations are obtained by the fixed sectional aerosol dynamics model [11, 12] in conjunction with the nucleation, growth and oxidation sub-models [20, 23].

The nucleation terms are calculated as

$$\rho \frac{\partial N_1^A}{\partial t} \Big|_{nu} = \rho \frac{\partial N_1^P}{\partial t} \Big|_{nu} = 2r_1 N_A / C_{\min}$$
(8)

$$\rho \frac{\partial N_j^A}{\partial t} \Big|_{nu} = \rho \frac{\partial N_j^P}{\partial t} \Big|_{nu} = 0, \, j = 2, 3, \dots, SN$$
(9)

where r_1 is the rate of the nucleation reaction R1; N_A is the Avogadro number; C_{\min} is the number of carbon atoms in the incipient soot particle and is set to be 90 000 in this study according to Fairweather *et al.* [20].

The other terms $\frac{\partial N_j^A}{\partial t}|_{co.,}$ $\frac{\partial N_j^A}{\partial t}|_{sg.,}$ $\frac{\partial N_j^A}{\partial t}|_{ox.}$ and $\frac{\partial N_j^P}{\partial t}|_{co.,}$ $\frac{\partial N_j^P}{\partial t}|_{sg.,}$ $\frac{\partial N_j^P}{\partial t}|_{ox.}$ are obtained using the same method as in [11, 12]. State-of-the-art fractal aggregate collision kernel [34] is employed to calculate the coagulation terms.

2.4. Radiation model

In this study, both the optically thin approximation (OTA) radiation model [35] and the discrete ordinate method (DOM) radiation model [36] were implemented into the flame code to calculate the radiative heat transfer rate of soot particles and gaseous species H_2O , CO_2 and CO. However, little difference was found in the results in modelling of the lightly sooting methane/air diffusion flame [5]. Therefore, only the results from using the OTA radiation model are presented.

3. Numerical methods and parallelisation methodology

3.1. Numerical methods

After being written in the general transport equation form, the governing equations are discretised based on the finite volume method. SIMPLE algorithm is used to handle the pressure and velocity coupling [37]. The continuity equation is converted into the pressure correction equation. Staggered mesh is used to formulate the discretised equations for velocities and scalars. The diffusive terms are discretised by the second order central difference scheme while the convective terms are discretised by the power law scheme [37]. Pseudo-time marching method is used to arrive at the converged steady state solution from the initial guess. The radial momentum, axial momentum, pressure correction and energy equations are solved in the segregated manner by the tri-diagonal matrix algorithm (TDMA). Since the gaseous species equations are normally stiff and closely coupled, to effectively deal with the stiffness of the system and speed up the convergence process, they are solved simultaneously at every control volume [38]. The key point of this method is to linearise the current step chemical reaction source terms by using their Taylor series expansions on the previous time step values and neglecting the second- and higher-order terms. The resulting Jacobian matrices are obtained by the perturbation method. A direct solver (Gauss elimination method) is used to solve the resulting linear system at each control volume. The species equations are solved control-volume-by-control-volume until the whole computational domain is covered. A coupled solver was used to solve the sectional soot equations in the same manner as the species equations. Solving them in a segregated manner by TDMA was not implemented because we found that this method had convergence difficulties unless a very small time step was used. The Jacobian matrices for soot equations were obtained by perturbing the sectional soot number densities and calling the sectional soot aerosol dynamics subroutine for the new (after perturbation) sectional soot source terms. The reason for the effectiveness of such a coupled approach for solving the soot equations is that the



Figure 2. Numerical procedure.

sectional soot equations are strongly coupled and the equation system is stiff, similar to the gaseous species equations.

The numerical procedure to solve the equation system is summarised in Figure 2. The unknown variables are in their primitive form, i.e. axial velocity u, radial velocity v, pressure correction p', gaseous species mass fractions Y_k (k = 1, 2, ..., KK; KK is the total number of gaseous species), sectional soot aggregate number densities N_j^A (j=1,2,...,SN), sectional soot primary particle number densities N_j^P (j=1,2,...,SN) and finally temperature T. The gaseous species thermal properties, transport properties and chemical reaction rates are obtained by CHEMKIN subroutines [39] and the database associated with the selected reaction mechanism.

3.2. Convergence criterion

We define a mean temperature T_{mean} and mean soot volume fraction $f_{v,\text{mean}}$ as

$$T_{\text{mean}} = \frac{\sum_{i=1}^{N_z} \sum_{j=1}^{N_r} T(i, j)}{N_z \times N_r}$$
(10)

$$f_{v,\text{mean}} = \frac{\sum_{i=1}^{N_z} \sum_{j=1}^{N_r} f_v(i, j)}{N_z \times N_r}$$
(11)

where N_z and N_r are the number of control volumes in the axial and radial directions respectively. T(i, j) and $f_v(i, j)$ are the temperature and soot volume fraction at control volume (i, j), respectively. It is noted that soot volume fraction is calculated from the sectional soot aggregate number densities. Convergence is assumed to be achieved when the following inequality is satisfied and the iteration process is terminated.

$$\max\left(\left|\frac{(T_{mean}^{(n)} - T_{mean}^{(n-1)})/T_{mean}^{(n-1)}}{dt}\right|, \left|\frac{(f_{v,mean}^{(n)} - f_{v,mean}^{(n-1)})/f_{v,mean}^{(n-1)}}{dt}\right|\right) < tol$$
(12)

where the superscript (n) means the *n*th outer iteration solution, *dt* is the time step value and *tol* is the prescribed tolerance value. We choose to define the convergence criterion in terms of the rate of change rather than the magnitude of change to eliminate the dependence on the time step value. Otherwise, a small time step value might cause premature termination of the iteration process. Temperature and soot volume fraction are chosen in formulating the convergence criterion because temperature is an important indicator of the flame property and soot volume fraction is an indicator of the soot field.

3.3. Parallelisation methodology

In order to reduce the computational time, parallelisation of the two-dimensional flame code is essential. In this study, Message Passing Interface (MPI) is chosen as the parallel programming tool since it is widely used and easy to program with [40]. We found that the most elegant way to develop the parallel code is via the domain decomposition method (DDM) [41]. By DDM, the whole computational domain is divided into N_p sub-domains, with N_p being the number of processors used. Each sub-domain is assigned to one processor for calculation and the calculations in all sub-domains are carried out simultaneously. Since each processor has less equations (owing to a small number of control volumes) to solve, the run time is expected to be reduced. In MPI, transferring of data among processors is time-consuming. If some amount of data has to be transferred, it is better to transfer the whole data. Based on this consideration, we place the ghost points [40] in the boundary of each sub-domain where communications are needed, such as for the finite difference approximation of the derivatives at the boundary points. This way, the frequency of data exchange is reduced and the efficiency is improved.

The optimum method to decompose the computational domain depends on the specific problem such as the mesh type and mesh size. A parallel code with a well decomposed domain should have good flexibility (i.e. small modification to the code when a different mesh is used), low data exchange frequency among sub-domains and good load balance.

Figure 3 shows an example of the computational domain uniformly decomposed into four subdomains (for four processors) using three different schemes. In Figure 3, blocks with numbers are the sub-domains and the numbers represent individual processors responsible for the computation



Figure 3. Example of four processors used to decompose the computational domain with different schemes.

of that sub-domain. To ensure good load balance, it is desirable to decompose the domain as uniformly as possible so that each processor has a similar number of control volumes or equations to solve. In this sense, all the three schemes are equally good. Scheme 2 decomposes the domain in both the axial and radial directions. Depending on the mesh size, the total length of the overlapping boundaries of the sub-domains might be shorter than scheme 1 and Scheme 3. So, this decomposition scheme might have the advantage that less ghost points are required and thus smaller amount of data has to be transferred. However, it is not as flexible as the other two schemes. When different meshes are used, the computational domain has to be re-decomposed in both directions. In the parallel code, arrays and variables pertaining to both directions have to be re-declared or re-defined. In scheme 3, the domain is decomposed in the radial direction only. Compared to scheme 1, in which the domain is decomposed in the axial direction only, scheme 3 is not a good one. A mesh with more grid points in the axial direction than in the radial direction is usually employed for modelling laminar coflow diffusion flames. As such, the total length of the overlapping boundaries of the sub-domains is larger in scheme 3 than in scheme 1. Therefore, in this study we choose to decompose the computational domain in the axial direction only as shown in scheme 1.

One challenge of the domain decomposition method in this study is associated with the staggered grid where three sets of meshes are used for solving the u momentum equation, v momentum equation and the remaining scalar equations. All the three sets of meshes have to be decomposed. We decompose the u mesh first, and then decompose the other meshes accordingly.

4. Results and discussions

4.1. Modelling condition

The parallel code is employed to model a lightly sooting methane/air flame which has been studied numerically and experimentally by Smooke et al. [5] and numerically by Liu et al. [36]. The computational conditions in this study are the same as those assumed by Smooke et al. The burner fuel tube has an inner radius of 0.5556 cm and the wall thickness is 0.0794 cm. Both the air and fuel inlet velocities and temperatures are assumed to be uniform, i.e. $u_{\text{fuel}} = 5.52$ cm/sec, $u_{air} = 12.54$ cm/sec, $T_{fuel} = T_{air} = 420$ K. The chemical kinetic mechanism used in this calculation is GRI-Mech 3.0 [3] with the removal of all reactions and species related to NO_x formation and contains 36 species. Non-uniform mesh is used in both axial and radial directions to save the computational time while resolving the large gradients. The computational domain covers 8.655 cm (z direction) \times 4.709 cm (r direction) and is divided into 192 (z) \times 86 (r) control volumes. Fine grids are placed in the r direction between 0 and 0.75 cm with a grid resolution of 0.2 mm and in the z direction between 0 and 2.95 cm with a grid resolution of 0.2 mm. Beyond the fine grid zone, the mesh is gradually stretched in both directions. It has been checked that further refinement of the mesh has negligible impact on the results. Inlet conditions are specified for the fuel and air streams at the z = 0 boundary. Symmetry conditions are enforced at the centreline, i.e. at r = 0. Free-slip conditions are assumed at the outer radial boundary, i.e. at r = 4.709cm. Zero-gradient conditions are enforced at the exit boundary at z = 8.655 cm. The mesh and boundary conditions are shown in Figure 4. Thirty-five sections are used to represent the solid soot phase with a section spacing factor of 2 (i.e. the representative mass of one section is twice of that of the preceding section). The resulting system is a set of 110 governing equations to be solved at each control volume. The calculation is performed on the NRC/ICPET high-performance parallel computing cluster which is comprised of AMD OpteronTM64bit RedhatTMLinux machines with the Portland Group PGITM6.2 FORTRAN compiler and MPI library.



Figure 4. Non-uniform mesh and boundary conditions.

4.2. **Two-dimensional representation**

The two-dimensional contour plots of the calculated temperature and soot volume fraction fields are shown in Figure 5 and Figure 6 respectively. The temperature and soot volume fraction both peak in the flame wing (annular region) in agreement with Smooke et al. [5] and Liu et al. [36]. According to Smooke et al. [5], a good estimate of the flame height is the lowest axial location at which the temperature peaks on the centreline. By examining the data of Figure 5, the flame height is found to be 4.1 cm approximately. It is noted from Figure 6 that a sharp decay of soot volume fraction occurs at z = 4.1 cm at the centreline, indicating that the calculated luminous



Figure 5. Temperature field. The unit is in K.

2.9E-07 2.5E-07 2.1E-07 1.7E-07 1.3E-07 9.0E-08 5.0E-08 1.0E-08

Figure 6. Soot volume fraction field.



Figure 7. Temperature profile at different heights above the burner.

flame height is also about 4.1 cm. The predicted peak soot volume fraction is 0.42 ppm which is very close to the measured peak value of 0.47 ppm [5].

4.3. Temperature profiles at different heights above the burner

The radial temperature profiles at four different heights above the burner are compared with the experimental data in Figure 7. At all four heights, as r increases from zero, i.e. from the centreline to the peak radial temperature locations, the model slightly under-predicts the temperature. Beyond the peak radial temperature locations, the model somewhat over-predicts the temperature. The predicted flame front is shifted to the outer radial region, i.e. the model predicts a wider flame than the experiment. At z = 1.0 cm, the centreline temperature is under-predicted by about 100 K. As z increases, the centreline temperature is predicted better. At z = 2.5 cm, the predicted centreline temperature is about 25 K lower than the experimental one. The lower predicted centreline temperature and the trend of the radial temperature distribution at all four heights are reasonably well predicted. The overall performance of the present flame model is very similar to the previous studies [5, 36] as far as the calculated temperature field is concerned.

4.4. Methane and acetylene profiles at different heights above the burner

The radial profiles of the calculated and measured methane mole fraction at four axial locations are compared in Figure 8. It is clear that the radial distributions of methane concentration at all four heights are reasonably predicted. The model over-predicts the methane concentrations in the centreline region, i.e. the model predicts a slower consumption rate of methane than the experiment. This is related to the under-prediction of flame temperature along the centreline. The methane levels decay faster in the outer radial regions, which is consistent with the fact that the flame temperature is over-predicted in these regions.

Acetylene is a crucial species in soot nucleation and growth steps in this study. Thus, a good predictability of the acetylene concentration is essential for the model to well predict



Figure 8. Methane profile at different heights above the burner.

the soot field. The calculated radial profiles of acetylene mole fraction at four flame heights are compared to those from experiment in Figure 9. It is evident that the centreline acetylene level is under-predicted, especially low in the flame at $z \le 1.5$ cm. It is believed that the under-prediction of acetylene concentration in this region is also tied to the under-prediction of centreline flame temperature shown in Figure 7. The flame model predicts a sharper decay of acetylene concentration in this is connected to the over-prediction of the temperature



Figure 9. Acetylene profile at different heights above the burner.



Figure 10. Soot volume fraction profile at different heights above the burner.

in these regions. Nevertheless, the present flame model predicts well the general trend of the acetylene distribution and the peak value of the acetylene level at all four heights examined.

4.5. Soot volume fraction profiles at different heights above the burner

The radial distributions of the calculated soot volume fraction at four axial locations are compared with the experimental data in Figure 10. The soot volume fraction distribution is well predicted at z = 2.0 cm. As z increases, the agreement between the model prediction and the measurement gradually deteriorates. The predicted radial location of the peak soot volume fraction does not move towards the flame centreline as fast as the experiment. This might be related to the temperature and acetylene concentration predictions. Nevertheless, the soot model used in this study generally well predicts the trend and absolute level of the radial soot volume fraction distribution.

4.6. Soot aggregate formation and size distribution

The major advantage of the fixed sectional aerosol dynamics model is its capability of modelling soot aggregate formation and size distribution. Knowing the structure of soot aggregates and the size distribution is very important to understand the roles of various physical and chemical processes involved in soot formation.

Figure 11 shows the total number density of soot primary particles summed over all sections along the flame centreline. As can be seen, the primary particle number density first increases then decreases with the height above the burner. The increase of primary particle number density is owing to the nucleation of nascent soot particles while the decrease of primary particle number density is owing to the oxidation of soot particles. Such can be confirmed from Figure 12 where the centreline nucleation rate and surface rate (growth rate + oxidation rate) are shown. From Figure 12, nucleation persists until z reaches 3.9 cm and surface growth persists until z reaches



Figure 11. Total number density of primary particles along the centreline.

3.6 cm. As *z* further increases from 3.6 cm, surface rate becomes negative meaning that oxidation becomes dominant and soot particles starts to be oxidised. As such, aggregates start to move from high sections to low sections and the first section aggregates (or equivalently primary particles; note that first section aggregates and first section primary particles are the same, both being monomers, as assumed in the fixed sectional aerosol dynamics model) become gaseous products. This leads to the disappearance of primary particles and accordingly the decrease of primary particle number density.

Figure 13 shows the centreline distribution of the number-averaged primary particle diameter $d_{av,num}^P$, defined as

$$d_{av,\text{num}}^{P} = \sum_{j=1}^{SN} d_{j}^{P} N_{j}^{P} / \sum_{j=1}^{SN} N_{j}^{P}$$
(13)

with d_j^P being the primary particle diameter in *j*th section. From Figure 13, $d_{av,num}^P$ first increases and then decreases with *z*, similar to the primary particle number density distribution in Figure 11. The causes are, however, slightly different. Here, the first increase of $d_{av,num}^P$ is owing to surface growth and the following decrease is, same as in Figure 11, owing to oxidation.



Figure 12. Nucleation and surface rates along the centreline. Surface rate is the sum of surface growth rate and oxidation rate.



Figure 13. Number-averaged primary particle diameter along the centreline.

Figure 14 shows the number of primary particles per aggregate as a function of aggregate volume equivalent diameter at four heights along the flame centreline. At each height considered, the curve shows a monotonically increasing trend indicating that larger aggregate always contains more primary particles. This is attributed to coagulation by which two small aggregates collide with each other to form a large aggregate with the primary particles in the two small colliding aggregates all being transferred to the resultant large aggregate. Thus, the resultant large aggregate contains more primary particles than each of the small aggregates. From Figure 14, it can also be found that the curve ends at a higher aggregate volume equivalent diameter indicating that larger and larger aggregates are formed as z increases from 2.0 cm to 3.5 cm. Those large aggregates are formed by coagulation and surface growth happening between these two heights. As z further increases from 3.5 cm to 4.0 cm, the curve does not extend further to higher aggregate volume equivalent diameter, indicating that no larger aggregates are formed in between these two heights. This is because oxidation becomes dominant and overrides the coagulation and surface growth. Also from Figure 14, for aggregates with the same volume equivalent diameter (or mass), the ones higher in the flame contain more primary particles. Because the aggregate is made up of spherical primary particles, the aggregates higher in the flame have smaller primary particle diameter. This observation is illustrated more intuitively in Figure 15. It should be noted that aggregate 2 is



Figure 14. Number of primary particles per aggregate as a function of aggregate volume equivalent diameter at four heights along the centreline.



Figure 15. Number of primary particles per aggregate of the same volume equivalent diameter at 3.0 cm and 3.5 cm above the burner along the centreline. Aggregate 1 and 2 have the same volume equivalent diameter or mass and belong to the same section. Aggregate 2 contains more primary particles than aggregate 1 while aggregate 1 contains larger primary particles than aggregate 2. Note that aggregate 1 does not lead to the

not evolved from aggregate 1 because aggregate 1 will become larger as it travels from z = 3.0 cm to z = 3.5 cm owing to surface growth and coagulation. The different structures of the same section aggregates 1 and 2 suggest different coagulation rates at these two locations. At z = 3.5 cm, coagulation is stronger than at z = 3.0 cm thus small aggregates collide faster to form new large aggregates. For a new aggregate with some mass to be formed, aggregate 2 will contain a larger number of primary particles owing to the faster collision at this location.

The soot aggregate size distribution function (ASDF) at each control volume is available from the fixed sectional aerosol dynamics model. Studying ASDF can help clarify the individual roles of different sooting processes. Here, two ASDFs are shown: the number-based ASDF n(D)defined as n(D) = dN/dLog D and the mass-based ASDF m(D) defined as m(D) = dM/dLog D, where D is the soot aggregate volume equivalent diameter in nm, N is the soot aggregate number density in (particles/cm³-gas) and M is the soot aggregate mass density in (g/cm³-gas).



Figure 16. (a) Number-based aggregate size distribution function n(D); (b) Mass-based aggregate size distribution function m(D).

formation of aggregate 2.

We examine the ASDFs at five selected locations, four at the centreline (z = 2.0, 3.0, 3.5, 4.0 cm) and one at the peak soot volume fraction (z = 2.77 cm, r = 0.31 cm). Figure 16(a) and Figure 16(b) show n(D) and m(D) as a function of D on the log-log plot, respectively. It is noted that D starts from 12.4 nm which is the diameter of the incipient soot particle containing 90 000 carbon atoms as assumed by Fairweather *et al.* [20]. Unfortunately, there is no experimental data on soot ASDF in this flame to validate the model prediction.

It can be found from Figure 16(a) that n(D) monotonically decreases as D increases at all the five locations considered. This means small aggregates dominate the aggregate number density. No bimodal n(D) is observed, suggesting that nucleation is stronger than coagulation [42]. As z increases, n(D) widens until z reaches 3.5 cm, indicating that large aggregates are gradually formed. At even larger z, n(D) starts to narrow indicating that large aggregates start to be oxidised and disappear. The area under the n(D) curve is an indication of the soot aggregate number density. One can find that the area first increases then decreases as z increases from 2.0 cm to 4.0 cm. This trend suggests that there are soot aggregates formed and then oxidised in between these two heights along the centreline, which can be confirmed by the soot volume fraction contour plot and also Figure 12. At the peak soot volume fraction location, i.e. z = 2.77 cm, r = 0.31 cm, n(D) is much wider, indicating there are more large aggregates at this location.

The mass-based ASDF m(D) shown in Figure 16(b) is quite different from the number-based ASDF n(D). At all locations except z = 2.0 cm r = 0 cm, m(D) first increases then decreases. This means aggregates of intermediate size dominate the aggregates mass density at these locations. The area under the m(D) curve is an indication of the aggregates mass density. From Figure 16(b), the area is larger at the peak soot volume fraction location, i.e. z = 2.77 cm, r = 0.31 cm, indicating there is more soot mass at this location, consistent with the fact that the soot volume fraction peaks at this location. It is also an indication that there is significant surface growth at this location since the increase of aggregates mass is mainly by surface growth.

5. Parallel code efficiency analysis

Using 12 processors, each outer iteration takes about 7 s with soot equations turned off, i.e. without calculating soot. Typically, about 3000 outer iterations are required to get the converged flame solution. After that, soot equations are turned on. The run time for each outer iteration increases to 21 s. Another 3000 outer iterations are required to obtain the final converged solution. The total run time is about 24 h.

To gain a quantitative understanding of the CPU time spending in each outer iteration, the CPU time breakdown is monitored and is shown in Figure 17. It can be seen that most of the CPU time



Figure 17. Percentage of CPU time for solving the governing equations in each outer iteration.



Figure 18. Speed-up obtained using different number of air processors.

is spent on solving the soot equations (64%) and the species equations (28%). The momentum equations, pressure correction equation, and energy equation only take a small percentage of CPU time to solve (8%). This CPU time breakdown analysis confirms that the fixed sectional aerosol dynamics model is computationally intensive.

Speed-up, a key indicator of the performance of a parallel code, is defined by the ratio of sequential code run time to the parallel code run time. Figure 18 presents the speed-up curve for 1, 4, 6 and 12 processors. The speed-up curve can be linearly fitted as speed-up = $0.8 \times N_p + 0.2$ approximately. The slope is less than unity owing to the overhead associated with the transfer of data among different processors in the calculation process. Nevertheless, the speed-up scales quite well with the number of processors and a parallel efficiency of 0.8 is considered good. Using 12 processors, a speed-up of almost 10 can significantly reduce the total run time.

Another key indicator of the performance of a parallel code is the load balance. A good parallel code should have a good load balance, i.e. each processor should perform as equal amount of work as possible. For a homogeneous parallel computing cluster, this translates to the requirement that each processor should have similar CPU time. It has been checked that all processors had a CPU time within $\pm 1\%$ of master processor's time and thus a good load balance is achieved. This results from the uniform domain decomposition.

6. Conclusions

An advanced fixed sectional aerosol dynamics model has been successfully implemented to model soot formation in a laminar axisymmetric coflow methane/air diffusion flame. This sectional model solves for two variables per section to model the formation of soot aggregates. The main concern of this article is the implementation strategy. The transport equations of the sectional soot particles were formulated. These equations were found to be strongly coupled and have to be solved simultaneously to ensure convergence. Parallel computation is essential for the implementation of this aerosol dynamics model. The best way to develop the parallel code is by the domain decomposition method and it is generally better to decompose the computational domain only in the axial direction compared with other domain decomposition schemes.

Compared to available experimental data from the literature, the flame temperature, species concentrations and soot volume fraction are generally well predicted, although the present flame code tends to predict a wider flame. The structures of soot aggregates are determined by the nucleation, coagulation, surface growth and oxidation processes. The aggregate number density

is found to be dominated by small aggregates while the aggregate mass density is in general dominated by aggregates of intermediate size.

Most of the CPU time is spent on solving the soot and species equations showing that the fixed sectional aerosol dynamics model is computationally intensive. The speed-up and load balance of the parallel code are shown to be good. Using 12 processors, a speed-up of almost 10 is achieved which makes it possible to model soot formation in laminar coflow diffusion flames with detailed chemistry and detailed aerosol dynamics within a reasonable amount of time.

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