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The effect of preferential diffusion on soot formation in a laminar ethylene/air diffusion flame

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The influence of preferential diffusion on soot formation in a laminar ethylene/air diffusion flame was investigated by numerical simulation using three different transport property calculation methods. One simulation included preferential diffusion and the other two neglected preferential diffusion. The results show that the neglect of preferential diffusion or the use of unity Lewis number for all species results in a significant underprediction of soot volume fraction. The peak soot volume fraction is reduced from 8.0 to 2.0 ppm for the studied flame when preferential diffusion is neglected in the simulation. Detailed examination of numerical results reveals that the underprediction of soot volume fraction in the simulation neglecting preferential diffusion is due to the slower diffusion of some species from main reaction zone to PAH and soot formation layer. The slower diffusion of these species causes lower PAH formation rate and thus results in lower soot inception rate and smaller particle surface area. The smaller surface area further leads to smaller surface growth rate. In addition, the neglect of preferential diffusion also leads to higher OH concentration in the flame, which causes the higher specific soot oxidation rate. The lower inception rate, smaller surface growth rate and higher specific oxidation rate results in the lower soot volume fraction when preferential diffusion is neglected. The finding of the paper implies the importance of preferential diffusion for the modeling of not only laminar but maybe also some turbulent flames.

Keywords: Soot; Preferential diffusion; Laminar diffusion flame; Lewis number; PAH

1. Introduction

Emission of soot not only has a detrimental effect on human health, but also contributes significantly to global warming [1]. Various restrictions have been placed on soot emission recently. The accurate modeling of soot formation in flames is critical to the understanding of the fundamental mechanism and the development of strategies to control soot emission.

Modeling soot formation in combustion systems is an extremely challenging task. Many factors affect the final results of numerical models for soot formation. One of these factors is the transport property calculation method. The impact of transport property calculation method on laminar flame speed and some other properties has been well studied. For example, Dixon-Lewis [2], Greenberg [3] and Warnatz [4] have studied the influence of transport property model on the prediction of various laminar flame properties and indicated the importance of thermal diffusion in predicting flame speeds and other properties of hydrogen/air flames. The study of Hancock *et al.* [5] concluded that thermal diffusion

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effect could not be neglected in the numerical simulation of vortex-flame interactions in hydrogen jet diffusion flames. Ern and Giovangigli [6] showed that thermal diffusion was important not only for the prediction of hydrogen/air flame structures, but also for the prediction of NO in a counterflow methane/air flame. Later, Ern and Giovangigli [7] further indicated that the prediction of extinction limits of counterflow premixed hydrogen and methane flames was sensitive to multicomponent transport model. The investigation of Williams [8] revealed that the prediction of extinction strain rate of nonpremixed flames was also sensitive to transport property model. The effect of transport property calculation method on soot formation in laminar flames has also been investigated previously. The studies of [9, 10] suggested that thermal diffusion of gas species had negligible effect on soot formation in an ethylene/air diffusion flame, but it became significant when some light components, such as hydrogen or helium, were introduced to fuel or air stream. Dworkin *et al.* [11] showed that compared to the approximate mixture-average method, the inclusion of the complex multicomponent transport property calculation method had only a minor effect on soot formation in laminar ethylene/air diffusion flames. It has been clear from the previous studies that as long as non-uniform Lewis number, which is defined as the ratio of the mixture heat conduction rate to species mass diffusion rate, is assumed and the difference in diffusion rates of different species is considered, the transport property calculation method based on most existing schemes in literature has only a minor impact on soot formation for most hydrocarbon flames but significantly affects results when there are some light components (such as hydrogen or helium) in fuel or oxidant stream [9–11]. However, few studies have investigated the influence of complete neglect of the difference in species diffusion coefficient, i.e. assumption of uniform Lewis number for all species, on soot formation in hydrocarbon flames.

On the other side, in many turbulent combustion models, it was argued that species diffusion was controlled by turbulent mixing and therefore a single uniform Lewis number (usually unity) was assumed for all gas species. For example, the studies of both Pitsch *et al.* [12] and Mauss *et al.* [13] assumed unity Lewis number for all gas species, although the former considered the difference in the diffusion of soot particles. The assumption of unity Lewis number for all gas species may not significantly affect the main heat release prediction. However, in the case of soot formation, since the main reaction zone (the inner reaction layer) is different from the soot formation layer, some species that are related to soot formation may have to diffuse from one zone to another. Consequently, the diffusion rates of various species and radicals from the main reaction zone to the soot formation layer may significantly affect soot formation rate. Therefore, it is possible that the assumption of a single unity Lewis number could cause a noticeable error in modeling results of soot formation. A recent study by Lovas *et al.* [14] pointed out that the assumption of a single unity Lewis number for all gas species could result in the underprediction of soot by one order of magnitude. The difference in the diffusion rates of various species is usually referred to as preferential diffusion.

In this paper, a numerical investigation on the effect of preferential diffusion on soot formation in a laminar ethylene/air coflow diffusion flame is conducted. This flame is selected due to the existing experimental data and well validated numerical model. Although the investigation of this paper is for a laminar flame, the results and the detailed analysis are of significant implications for turbulent flames, especially for those in flamelet regime. We shall first introduce the flame configuration and numerical model. Then the overall effect of neglect of the preferential diffusion on predicted soot volume fraction will be demonstrated by comparing the results from simulations with and without preferential diffusion, followed by detailed discussion on how preferential diffusion affects soot formation. Finally conclusion remarks are drawn.

Figure 1. Schematic flame configuration.

2. Flame configuration, numerical scheme and soot model

2.1. Flame configuration

The flame used to investigate the effect of preferential diffusion on soot formation in this paper is a two dimensional laminar ethylene/air coflow diffusion flame that has been investigated experimentally and numerically previously [15]. The schematic flame configuration is shown in Figure 1. The fuel stream issued from a 10.9-mm inner diameter vertical tube, and the air from the annular region between the fuel tube and an 88-mm inner diameter concentric tube. The wall thickness of the fuel tube is 0.95 mm. The volume flow rates of air and ethylene were 284 l/min and 194 ml/min, respectively, at atmosphere pressure and room temperature condition (298 K). The spatially-resolved soot volume fraction in the flame has been previously measured by the diffuse-light two-dimensional line-of-sight attenuation (LOSA) optical diagnostic method [15]. Details of the optical diagnostic method can be found from [16]. The temperature of the flame has also been previously measured with CARS nitrogen thermometry [17].

2.2. Numerical model

The flame was modeled by numerical simulation. The low Mach number assumption was adopted. The governing equations were discretized using the finite volume method in axisymmetric cylindrical coordinates. The SIMPLE numerical scheme [18] was used to handle the pressure and velocity coupling. The diffusion terms in the conservation equations were discretized by the central difference method and the convective terms were discretized by the power law method [18]. To speed up the convergence process, the discretized governing equations of gas species and soot moments were, respectively, solved in a fully coupled fashion at each control volume [19]. Those of momentum, energy and pressure correction were solved using the tri-diagonal matrix algorithm. The thermal properties were obtained by using the algorithm given in [20].

In order to investigate the effect of preferential diffusion of gas species on soot formation, three simulations were conducted by using different transport property

Table 1. Conditions of the three sets of simulations.

	Transport property calculation	Thermal diffusion of gas species	Temperature
SIM1	Calculated by mixture-average method [21]	Considered for H ₂ and H and neglected for other species	Calculated
SIM2	Calculated by eq. (1)	Neglected	Calculated
SIM3	Calculated by eq. (1)	Neglected	Same as in SIM1

calculation methods, with one including preferential diffusion and the other two neglecting preferential diffusion. It has been shown by Dworkin *et al.* [11] that compared to the mixture-average method, the inclusion of the complex multicomponent transport property calculation method for gas species has only a minor effect on soot formation. Our previous study [10] indicated that thermal diffusion of gas species has negligible effect on soot formation in an ethylene/air diffusion flame. Therefore, in order to save simulation time, preferential diffusion was taken into account by calculating transport properties of gas species using the mixture-average method given by Kee *et al.* [21] in the first simulation (SIM1), with thermal diffusion of H and H₂ being included while that of other species being neglected. In the second simulation (SIM2), preferential diffusion was neglected and unity Lewis number was assumed for all species. Therefore, the diffusion coefficient (D_i) of each gas species in SIM2 was obtained according to local mixture thermal conductivity by

$$D_i = \lambda / \rho C_p \quad (1)$$

where λ is the local mixture thermal conductivity, ρ is the local mixture density and C_p is the local mixture specific heat. Thermal diffusion of all species was neglected in SIM2.

Since preferential diffusion also causes variation in temperature which in turn affects PAH and soot formation, the third simulation (SIM3) was conducted to exclude the effect of temperature variation due to preferential diffusion on soot formation. In SIM3, flame temperature was kept the same as in SIM1, while the diffusion coefficient of each gas species was obtained by the method that is the same as in SIM2. Table 1 gives the summary of the calculation conditions for all three simulations.

Due to radial symmetry, only half of the flame was simulated. The computational domain covers an area from 0 to 3.0 cm in the radial (r) direction and 0 to 11.0 cm in the axial (z) direction. The inflow boundary ($z = 0$ cm) corresponds to the region immediately above the fuel nozzle. Totally 160 (z) $\times$ 190 (r) non-uniform grids were used in the simulations, with finer grids placed in the primary reaction zone and near the fuel nozzle exit region. The symmetric condition was used for the centerline in the simulation. The free slip boundary condition was used for the side boundary, and zero-gradient condition was employed for the top outlet. At the bottom of the domain, uniform velocities, temperatures and compositions were specified for the center fuel tube region and outer concentric space, respectively, based on supplied fuel and air. The fuel was pure ethylene and air was assumed to consist of 79% nitrogen and 21% oxygen (by volume). Radiation heat transfer was calculated by the discrete ordinate method coupled to a statistical narrow-band correlated-K (SNBCK) based wide band model for the radiating properties of CO, CO₂, H₂O and soot

[22]. Other details of the numerical methods can be found from our previous publications [23, 24].

2.3. Soot model

The formation and evolution of soot particles were simulated by the method of moments [25]. Six concentration moments were used. The soot particle moments are defined as

$$M_r = \sum_{i=1}^{\infty} m_i^r N_i \quad (2)$$

where M_r is the r th moment of soot particle distribution, and m_i and N_i are the mass and the particle number density, respectively, of the soot particles of size class i . The soot particle mass is represented by the number of carbon atoms. Six concentration moments (i.e. $r = 0, 1, 2, 3, 4, 5$) are used.

The governing equation for each soot concentration moment is

$$\begin{aligned} \rho u \frac{\partial (M_r/\rho)}{\partial z} + \rho v \frac{\partial (M_r/\rho)}{\partial r} = \frac{\partial}{\partial z} \left(\rho D_{p,1} \frac{\partial}{\partial z} \left(\frac{M_{r-2/3}}{\rho} \right) \right) \\ + \frac{1}{r} \frac{\partial}{\partial r} \left(r \rho D_{p,1} \frac{\partial}{\partial r} \left(\frac{M_{r-2/3}}{\rho} \right) \right) - \frac{\partial}{\partial z} (V_{T,z} M_r) - \frac{1}{r} \frac{\partial}{\partial r} (r V_{T,r} M_r) + Q_r \end{aligned} \quad (3)$$

where ρ is density (g/cm^3), u and v the axial (z) and radial (r) direction velocities (cm/s), respectively, Q_r the source term, and $M_{r-2/3}$ the fractional moments obtained by interpolation between the whole moments. Quantity V_{T,x_i} is the thermal diffusion velocity of soot in z or r direction, and is calculated by

$$V_{T,x_i} = -0.55 \frac{\nu}{T} \frac{\partial T}{\partial x_i} \quad (x_i = z \text{ or } r) \quad (4)$$

where ν is the kinematic viscosity. Note that thermal diffusion of soot particles is included in all three simulations, since the focus of this paper is the effect of preferential diffusion of gas species. Quantity $D_{p,1}$ is the diffusion rate of the smallest soot particles, and is given by

$$D_{p,1} = \frac{3}{2\rho} \sqrt{\frac{\bar{m} k_B T}{2\pi}} \left(1 + \frac{\pi \alpha_T}{8} \right)^{-1} \frac{1}{d_l^2} \quad (5)$$

with $\bar{m}$ being the mean mass of the gas (g), k_B the Boltzmann's constant ($\text{erg mol}^{-1} \text{K}^{-1}$), T the temperature (K), α_T the thermal accommodation coefficient (0.9), and d_l the diameter of the smallest soot particle (cm). The source term Q_r in each moment equation accounts for particle nucleation, coagulation, surface growth and oxidation of soot particles.

The nucleation of soot particles is assumed to be due to the coalescence of two large size PAH molecules, pyrene (A4), into a dimer. Then the particle size increases or decreases due to the particle coagulation, surface growth and oxidation. The gas phase chemistry and the calculation methods for the particle nucleation, coagulation, surface growth and oxidation are basically those developed by Appel *et al.* [26]. However, some modifications have been

Figure 2. Soot volume fraction from experiment and the three simulations.

made for gas phase chemistry and surface growth calculation because the original method and chemistry significantly underpredicted the soot volume fraction for the flame studied in this paper. More details of the soot model can be found elsewhere [15].

3. Results and discussion

Figure 2 displays the distribution of soot volume fraction obtained from experiment and the three simulations for the studied flame. It is observed that when preferential diffusion was taken into account (SIM1), simulation successfully captured the measured soot volume fraction by experiment. Both the peak value and the distribution of soot volume fraction are close to those from experiment. However, when the preferential diffusion was neglected and unity Lewis number was assumed for all species, the simulations (SIM2 and SIM3) significantly underpredicted soot volume fraction for the flame. This is qualitatively consistent with the observation by Lovas *et al.* [14] who used a different flame configuration. The peak soot volume fractions are 8.0 and 8.3 ppm in experiment and SIM1, respectively, while drops to 2.0 and 1.85 ppm in SIM2 and SIM3, respectively. Compared to the significant difference between SIM1 and SIM2, the difference between SIM2 and SIM3 is very small.

Figure 3. Flame temperature distribution from experiment and the first (SIM1) and second (SIM2) simulations.

This suggests that the effect of preferential diffusion caused temperature variation on soot formation is not significant.

Figure 3 illustrates the distributions of flame temperature obtained from experiment, SIM1 and SIM2. The temperature distribution of SIM3 is not shown since it is exactly same as that of SIM1. Clearly, the simulation neglecting preferential diffusion (SIM2) predicted higher temperatures over a more extended region than the simulation including preferential diffusion (SIM1). The peak temperatures from SIM1, SIM2 and experiment are 2063, 2153 and 2156 K, respectively. Although the simulation including preferential diffusion

underpredicted the peak value, it predicted a reasonable overall temperature distribution. In both SIM1 and experiment, the peak temperature occurs in the flame wing region and the higher temperature region does not converge to the upper centerline flame tip region. As has been discussed in our previous publication [22], this is because of the effect of radiation heat loss due to the presence of soot. However, the simulation neglecting preferential diffusion (SIM2) predicted an unreasonable temperature distribution, although the calculated peak value is closer to the measured than that by SIM1. Firstly, temperature in SIM2 is higher than in SIM1. Secondly, the higher temperature region in SIM2 converges to the upper centerline flame tip region where the temperature in SIM1 and experiment is significantly lower than the corresponded peak value that occurs in the flame wing region. Therefore, we can conclude that the neglect of preferential diffusion results in a significant underprediction of soot formation rate and an overprediction of flame temperature. It should be pointed out that although preferential diffusion also affects flame temperature due to reaction zone shift, the higher temperature in SIM2 is more closely related to the lower soot volume fraction owing to radiation effect. Although not shown, additional simulations show that the peak temperature difference between the cases with and without preferential diffusion reduces to 35 K if soot is neglected, which is much smaller than the 90-K peak temperature difference between SIM1 and SIM2. The focus of this paper is the influence of preferential diffusion on soot formation. Therefore we will examine the detailed effect of preferential diffusion on soot formation below.

To understand how preferential diffusion affects the prediction of soot formation, the radial profiles of soot inception and surface growth rates at main soot formation region (from $z = 1.0$ to 4.0 cm) are displayed in Figure 4. Please note that the inception and surface growth rates from SIM2 and SIM3 at some axial positions are so small that some curves cannot be clearly observed in the graph. Surface growth rate in Figure 4 is the total growth rate, i.e. soot formation rate per unit volume due to surface growth. It is noted that both inception and surface growth rates from SIM2 or SIM3 are significantly smaller than those from SIM1 at all axial heights, suggesting that the neglect of preferential diffusion results in the underprediction of both soot inception and surface growth rates. Clearly, the smaller inception and surface growth rates cause the lower soot volume fractions in SIM2 and SIM3 than in SIM1, as shown in Figure 2. Relatively the difference between SIM2 and SIM3 is smaller, implying that the effect of temperature variation due to preferential diffusion on soot formation rate is small.

We first examine how preferential diffusion affects inception rate. In the soot model employed, particle inception is assumed to be due to the coalescence of two large size PAH molecules, pyrene (A4). Therefore, inception rate depends on temperature and concentration of pyrene. Since Figure 3 has shown that the temperature in SIM2 is higher than that in SIM1 and the temperature in SIM3 is the same as that in SIM1, the lower inception rates in SIM2 and SIM3 should be the sole result of lower concentration of pyrene. This is confirmed by the radial profiles of pyrene at different axial heights in Figure 5(a), which reveals that the concentrations of pyrene in SIM2 and SIM3 are much lower than that in SIM1. The lower concentrations of pyrene directly results in the lower inception rates in SIM2 and SIM3 than in SIM1. The significantly lower concentrations of pyrene in SIM2 and SIM3 than in SIM1 can be further explained.

The formation of pyrene is closely related to smaller size PAHs and precursors of PAH. Figure 5(b) shows the radial profiles of propargyl (C_3H_3) whose recombination is the primary reaction for the formation of benzene, the first aromatic ring. It is found that although the concentrations of propargyl in SIM2 and SIM3 are also lower than in SIM1, the difference between SIM1 and SIM2 or between SIM1 and SIM3 is smaller than in

Figure 4. Radial profiles of inception and surface growth rates at different axial heights. (a) Inception rate; (b) surface growth rate.

the concentration of pyrene at each corresponding axial height, which implies that the difference between SIM1 and SIM2 or between SIM1 and SIM3 in the concentrations of PAHs increases with the increase in the size of PAH. Although the temperature variation due to preferential diffusion may affect the formation and growth of PAH due to the shift of the direction of reversible reactions, the smaller difference in the concentration of pyrene between SIM2 and SIM3 (both of which used unity Lewis number assumption but have different and same temperature as SIM1, respectively) than that between SIM1 and SIM2 or between SIM1 and SIM3 suggests that this shift is not the primary reason for the significant difference in the concentration of pyrene between SIM1 and SIM2. The increase in the difference between SIM1 and SIM2 or between SIM1 and SIM3 in the concentrations of larger size PAHs might be due to the fact that the PAH growth and soot formation layer is different from the main reaction zone. The diffusion rates of some species from main reaction zone to the layer of PAH growth and soot formation affect the growth of PAH. The

Figure 5. Radial profiles of pyrene (A4) and propargyl (C_3H_3) at different axial heights. (a) Pyrene; (b) Propargyl.

neglect of preferential diffusion in SIM2 and SIM3 might slow the diffusion of the species that play important role in PAH growth and reduce the PAH growth rate. Because of the accumulation of this effect during the PAH growth, the difference between the simulations with and without preferential diffusion in the concentration of pyrene, a larger size PAH, is significant.

PAH growth and soot formation usually happen in the fuel rich region (on the near centerline side of the stoichiometric zone), while the main chemical reactions of the flame occur in the near stoichiometric zone. One of the species that play significant role in PAH growth and has to diffuse from the main reaction zone to soot formation layer is atomic hydrogen (H). It is a key radical controlling PAH growth and soot formation. Figure 6 shows the radial profiles of H at different axial heights. It demonstrates that at each axial height, compared to the radial positions of peak pyrene concentration and peak soot formation (inception and surface growth) rate (as shown in Figures 4 and 5), the position of the peak

Figure 6. Radial profiles of H.

Figure 7. Radial profiles of surface area and specific acetylene (C_2H_2) addition rate. (a) Surface area; (b) specific acetylene addition rate.

H concentration is further away from centerline. This is due to that the primary H formation reactions occur in the main reaction zone that is further away from centerline compared to PAH growth and soot formation layer. Although the peak concentrations of H in SIM2 and SIM3 are higher than that in SIM1 at $z = 1.0$ and 2.0 cm, the concentrations of H on the near centerline side (left) of the peak value position are lower in SIM2 and SIM3 than in SIM1. This is because of the neglect of preferential diffusion and unity Lewis number assumption for all species in SIM2 and SIM3. Radical H is a light species that has much smaller Lewis number (higher mass diffusion coefficient) than most other species. Therefore, the neglect of preferential diffusion in SIM2 and SIM3 results in the significant underprediction of mass diffusion rate of H from the main reaction zone to the near centerline region at $z = 1.0$ and 2.0 cm where significant PAH growth and soot formation happen. As a result, the PAH formation/growth and soot inception rates in SIM2 and SIM3 were significantly underpredicted compared to SIM1 in lower flame region. In higher flame region ($z = 3.0$ and 4.0 cm), the concentration of H on the near centerline side of the peak value position gradually becomes higher in SIM2 and SIM3 than in SIM1 due to the gradual diffusion of H formed in lower flame region. However, the lower PAH formation and growth rates in lower flame region result in that pyrene concentration in higher flame region is still lower in SIM2 and SIM3 than in SIM1. Therefore, the lower diffusion rate of radical H from the main reaction zone to PAH growth and soot formation layer in the lower flame region is the primary reason for the significantly lower concentration of pyrene and low soot inception rate in SIM2 and SIM3 than in SIM1.

Now we examine how preferential diffusion affects surface growth. Although Figure 4 shows that the surface growth rates in SIM2 and SIM3 are lower than that in SIM1, it does not mean that the neglect of preferential diffusion results in the lower surface growth reaction rates, since surface growth rate is the product of the specific surface area (area per unit volume, cm^{-1}) and specific surface growth reaction rate (the rate per unit surface area, $\text{g}/\text{cm}^2\text{s}$). Specific surface area heavily depends on particle number density and inception rate. Figure 7(a) shows the radial profiles of specific surface area, indicating that the surface areas in SIM2 and SIM3 are significantly lower than that in SIM1. This is mainly due to the lower inception rate which results in the lower particle number density in SIM2 and SIM3. The lower specific surface area is one factor that causes the lower surface growth rates in SIM2 and SIM3 than in SIM1.

Specific surface growth consists of PAH condensation and acetylene (C_2H_2) addition. Because of the lower concentration of PAH in SIM2 and SIM3 than in SIM1, it is easy to imagine that the specific PAH condensation rates in SIM2 and SIM3 are lower than in SIM1. Besides, simulation shows that acetylene addition dominates specific surface growth in the studied flame. Therefore, we only examine the specific acetylene addition rate. Figure 7(b) shows the radial profiles of the specific acetylene addition rate at different axial heights. It is found that although the radial positions of the peak rates are different, overall there is no significant difference in specific acetylene addition rate between SIM1 and SIM2 or between SIM1 and SIM3, suggesting that preferential diffusion does not causes significant prediction error in specific acetylene addition rate. This might be due to that specific acetylene addition rate depends on the combined effects of temperature and concentrations of acetylene and atomic hydrogen. In the soot model employed, the acetylene addition is calculated by the H-abstraction and carbon-addition (HACA) mechanism developed by Appel *et al.* [26]. Based on the mechanism, the specific acetylene addition rate primarily depends on temperature and concentrations of acetylene and atomic hydrogen. The temperature and concentration of H have been shown in Figures 3 and 6, respectively. Figure 8 displays the radial profiles of acetylene. We observe that although the concentration of H in SIM2 and SIM3 is lower

Figure 8. Radial profiles of acetylene (C_2H_2).

Figure 9. Radial profiles of specific oxidation rate and OH concentration. (a) Specific oxidation rate; (b) OH concentration.

than in SIM1 in the surface growth region (near centerline side in Figure 6) at $z = 1.0$ and 2.0 cm, the concentration of acetylene is higher in SIM2 and SIM3 than in SIM1 in the surface growth region (note that surface growth happens on the right side of the peak value position in Figure 8). Therefore, the combined effects of temperature and concentrations of atomic hydrogen and acetylene results in the overall similar specific acetylene addition rates in SIM1, SIM2 and SIM3 at $z = 1.0$ and 2.0 cm, although the peak positions in radial direction slightly differ in the three simulations. Accordingly, the neglect of preferential diffusion does not significantly affect specific acetylene addition rate in lower flame region. In higher flame region ($z = 3.0$ and 4.0 cm), there is slight difference in the specific acetylene addition rate among three simulations, but the significantly low specific area in SIM2 and SIM3 than in SIM1 causes the much lower total surface growth rates in SIM2 and SIM3. Therefore, the lower surface growth rates in SIM2 and SIM3 than in SIM1 are primarily due to the underprediction of surface area and PAH condensation rate.

Finally we examine if preferential diffusion affects soot oxidation. Figure 9(a) shows the radial profiles of specific soot oxidation rate (oxidation rate per unit surface area). It is illustrated that specific oxidation rates in SIM2 and SIM3 are higher than in SIM1 at most axial heights. This might be due to the higher OH concentrations in SIM2 and SIM3, as shown in Figure 9(b), since OH is the primary agent responsible for soot particle oxidation. Therefore, the neglect of preferential diffusion may also cause the overprediction of specific soot oxidation rate.

Overall, above results show that the neglect of preferential diffusion results in significant underprediction in soot formation rate in a diffusion flame. The primary reason for this is that the neglect of preferential diffusion reduces the diffusion of some key species/radicals, such as atomic hydrogen, from the primary reaction zone to the PAH growth and soot formation layer. This leads to the underprediction of PAHs growth rate and the overprediction of soot oxidation, which causes the lower net soot formation rate. Although these results are obtained from a laminar diffusion flame, they may be also of importance for some turbulent flame modeling, since soot formation layer differs from the inner reaction zone for local laminar flamelets. Therefore, it would be of interest to further investigate the effect of preferential diffusion on turbulent flames in the future.

4. Conclusion remarks

The effect of preferential diffusion on soot formation in a laminar ethylene/air diffusion flame has been investigated. Three simulations were conducted by using different transport property calculation methods with one including preferential diffusion and the other two neglecting preferential diffusion. The results show that the simulation including preferential diffusion predicted the peak value and soot volume fraction distribution that are very close to the experimentally measured, while the simulations neglecting preferential diffusion significantly underpredicted soot volume fraction in the flame. A further analysis of the numerical details suggests that the underprediction of soot formation rate by the simulations neglecting preferential diffusion is due to the lower diffusion rate of some species, such as H, from the main reaction zone to PAH growth and soot formation layer. This results in the lower soot formation rate due to the smaller inception and surface growth rates in the simulations without preferential diffusion. The lower inception rate is a direct result of the lower PAH concentration and the smaller surface growth rate is due to the smaller surface area caused by lower inception rate. Besides, the neglect of preferential diffusion causes higher OH concentration in the flame and therefore higher specific soot oxidation rate. This may also contribute to the underprediction of soot volume fraction in

the simulations neglecting preferential diffusion. The results of the paper may also imply that preferential diffusion should be taken into account in the modeling of some turbulent flames, since soot formation layer differs from the inner reaction zone for local laminar flamelets.

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