Transient soot dynamics in turbulent nonpremixed ethylene–air counterflow flames

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Abstract
The dynamics of soot formation in turbulent ethylene–air nonpremixed counterflow flames is studied using direct numerical simulation (DNS) with a semi-empirical soot model and the discrete ordinate method (DOM) as a radiation solver. Transient characteristics of soot behavior are studied by a model problem of flame interaction with turbulence inflow at various intensities. The interaction between soot and turbulence reveals that the soot volume fraction depends on the combined effects of the local conditions of flow, temperature, and fuel concentration, while the soot number density depends predominantly on the high temperature regions. Depending on the relative strength between mixing and reaction, the effects of turbulence on the soot formation lead to three distinct paths in deviating the data points away from the laminar flame conditions. It is found that turbulence has twofold effects of increasing the overall soot yield by generating additional flame volume and of reducing soot by dissipating soot pockets out of high-temperature regions. The relative importance between the two effects depends on the relative length scales of turbulence and flame, suggesting that a nonmonotonic response of soot yield to turbulence level may be expected in turbulent combustion.

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Keywords: Transient dynamics; Soot; Counterflow; Direct numerical simulation

1. Introduction
Soot formation in combustion devices is of serious concern for its environmental and performance impacts. Stringent regulations on pollutant emissions from combustion systems such as diesel and aircraft engines have demanded engineers to develop combustion systems to reduce NOx and soot emissions. To achieve this goal, it is important to understand the fundamental mechanisms of the physical and chemical subprocesses related to the soot formation and its interaction with turbulence and radiation.

Although there have been significant advances during the past decades in understanding the mechanisms of soot formation and destruction processes [1], detailed transient soot dynamics in the presence of turbulent momentum and scalar transport has not been fully investigated. The challenges lie not only in the uncertainties in the detailed chemical mechanisms describing the soot inception, growth, and oxidation processes, but also in the complexities in the coupling between the relevant chemical and physical processes occurring at vastly different characteristic time scales.
Effects of external flow parameters on the fundamental soot characteristics in laminar/turbulent jet flames have been extensively investigated, but mostly in the steady context [2–7]. More recently, several numerical studies adopting detailed gas-phase and soot chemistry have been conducted, but they were also limited to laminar flames [8–10]. Regarding the time-dependent characteristics of soot, a series of experimental and numerical studies on flickering flames have been undertaken, reporting that the unsteady fluctuation can yield 5–6 times more soot than under steady conditions due to the increased residence time [11–13]. In these studies, however, primary emphasis was on the overall soot generation as an integrated quantity, and few studies exist on the local interaction between soot and the neighboring flow and temperature fields that are subjected to large temporal and spatial fluctuations. Understanding of such details would allow improved predictive submodels in high-fidelity simulation.

Therefore, the objective of this study is to investigate the soot dynamics in turbulent counterflow flames using direct numerical simulations. A steady two-dimensional counterflow nonpremixed flame is subjected to interact with prescribed homogeneous turbulent flows injected at the two opposing inflow boundaries. Data from several test cases provide fine-scale details during the highly transient interactions between chemistry and turbulent transport.

2. Modeling description

A model problem of the flame-turbulence interaction is adopted in a two-dimensional counterflow configuration. The domain size is 2.48×4.96 cm with a grid resolution of 400×800, where the left and right boundaries are set to be the inflows. A steady laminar diffusion flame is established in an opposed flow using a 1-D solution profile [14]. The fuel stream is pure ethylene supplied from the right boundary and the oxidizer stream is a mixture of 22% oxygen and 78% nitrogen from the left boundary. To achieve a stable counterflow flames and to feed turbulence at the inlets, which are not a trivial task, improved nonreflecting boundary conditions are used at both inlet and outflow boundaries [15].

In this study, a compressible reacting flow code (S3D) with a 4th-order Runge-Kutta time integration and an 8th-order central differencing scheme is used [14–16]. Three different cases were simulated with different flow parameters as in Table 1. The simulations were calculated up to 2 flow times, $t_{f} / U_{0}$. To feed turbulent eddies, turbulent fluctuation velocities calculated from auxiliary isotropic turbulent spectrum field are added to the mean inlet velocities [15]. Since the characteristic eddy-turnover time ($\sim L_{1}/U'$) in the simulation is about 6 ms which is less than half of the mean flow sweep time, it is expected that the turbulent eddies reasonably well-developed by the time they reach the flame zone.

As an effort to model a representative sooting nonpremixed flame at amenable computational cost, a single-step ethylene–air chemistry proposed by Westbrook and Dryer [17] was used. For the soot calculation, we adopted a semi-empirical model proposed by Moss and coworkers [18, 19]. The transport equations for the soot number density, $N_{r}$, and the soot mass fraction, $Y_{s}$, are solved, which are given in normalized form by

$$\begin{align*}
\frac{\partial \rho Y_{s}}{\partial t} + \nabla \cdot (\rho \mathbf{u} Y_{s}) &= \frac{1}{\rho_{0}} \left( \frac{\partial}{\partial \rho_{0}} \frac{\partial}{\partial \rho_{0}} \left( \frac{\partial}{\partial \rho_{0}} \left( \frac{\partial}{\partial \rho_{0}} \rho_{0} Y_{s} \right) \right) \right) \\
\frac{\partial \rho Y_{s}}{\partial t} + \nabla \cdot (\rho \mathbf{u} Y_{s}) &= \frac{1}{\rho_{0}} \left( \frac{\partial}{\partial \rho_{0}} \frac{\partial}{\partial \rho_{0}} \left( \frac{\partial}{\partial \rho_{0}} \rho_{0} Y_{s} \right) \right)
\end{align*}$$

where $N_{0}$ is the Avogadro number, $Sc$ is the Schmidt number of soot which is specified to be 1000, and $V_{t, j}$ is the thermophoresis velocity of a soot particle in direction $j$ [20].

The reaction source terms in Eq. (1) are given by

$$\begin{align*}
\frac{\partial \rho_{0} Y_{s}}{\partial t} &= \rho f_{\text{f}} T^{1/2} X_{F} \exp(-T_{s, f}/T) \\
\frac{\partial \rho_{0} Y_{s}}{\partial t} &= \rho f_{\text{f}} T^{1/2} X_{F} \exp(-T_{s, f}/T) N_{s} \\
\frac{\partial \rho_{0} Y_{s}}{\partial t} &= \rho f_{\text{f}} T^{1/2} X_{F} \exp(-T_{s, f}/T) N_{s}
\end{align*}$$

such that the rate for $N_{r}$ is represented by the nucleation and coagulation rates, and $Y_{s}$ results from surface growth, nucleation, and oxidation, respectively. $X_{F}$ is the mole fraction of fuel (C$_{2}$H$_{4}$), $X_{O}$ is the mole fraction of oxidizer (O$_{2}$), and $S$ is the soot surface area per unit volume. $f_{\text{f}}$ is the soot volume fraction defined by $f_{\text{f}} = \rho Y_{s} / \rho_{s}$, where $\rho_{s}$ is the soot density specified as 1800 kg/m$^{3}$. The soot oxidation due to oxygen is given by $\omega_{\text{ox}} = 1.085 \times 10^{5} X_{O} T^{1/2} \exp(-179778/T)$ [21].

We considered radiative heat transfer associated with soot, H$_{2}$O, and CO$_{2}$. The absorption coefficients of H$_{2}$O and CO$_{2}$ are calculated from the Planck mean absorption coefficients [22] and the absorption coefficient for soot is given by $\kappa_{\text{ax}} = 1.307 \times 10^{5} f_{\text{f}} T^{1/2} 1/[m]$ [23].

The net radiative heat loss from a control volume is determined by the difference between emitted energy and absorbed irradiation [24, 25]:

$$-\nabla \cdot \mathbf{q}_{\text{rad}} = \kappa_{\text{a}} \int_{4\pi} I(r, s) d\Omega - 4\pi I_{b}(r),$$

where $\mathbf{q}_{\text{rad}}$ is the radiative heat flux vector, $\kappa_{\text{a}}(r)$ is the gray absorption coefficient of the medium, $I_{b}(r)$ is the intensity of blackbody radiation at the
position, \( r \). \( I(\mathbf{r}, \hat{s}) \) is the radiative intensity in the direction, \( \hat{s} \), at the position, \( \mathbf{r} \) and \( \Omega \) is the solid angle. Since the intensity of blackbody radiation is only a function of temperature, the radiation loss can be calculated if the intensity of the medium is already given.

The discrete ordinate method (DOM) is used to solve the radiative transfer equation with a discrete set of equations for a finite number of ordinates and corresponding weighting factors. The integrals over the entire solid angle for scattering are replaced by numerical quadrature summed over each discrete ordinate. The discrete equations can be written for each ordinate,

\[
\hat{s}_i = \sum_{i=1}^{n} \frac{n_i}{c_i} + \sum_{j=1}^{n} g_{ij} + \sum_{k=1}^{n} l_{ik},
\]

where \( S_i \) represents the radiative source function, \( k_e \) is the extinction coefficient, and \( n \) is the number of discrete ordinates.

A finite difference form of Eq. (4) is solved by iterative method with appropriate boundary conditions. The radiative heat loss, Eq. (3), is updated using the converged radiative intensity at the end of every time step.

### Table 1

Parameters for three different cases for flame–turbulence interaction

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Case 1</th>
<th>Case 2</th>
<th>Case 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean inlet velocity, ( u_0 ) (m/s)</td>
<td>1.56</td>
<td>1.56</td>
<td>3.12</td>
</tr>
<tr>
<td>Turbulence intensity, ( u'/u_0 )</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>Integral length scale, ( L_{11}/l_x )</td>
<td>0.1</td>
<td>0.05</td>
<td>0.1</td>
</tr>
<tr>
<td>Flame thickness, ( l_f ) [cm]</td>
<td>0.215</td>
<td>0.215</td>
<td>0.158</td>
</tr>
<tr>
<td>Length scale ratio, ( L_{11}/l_f )</td>
<td>1.16</td>
<td>0.58</td>
<td>1.58</td>
</tr>
</tbody>
</table>

To examine the chemical aspects of soot formation, various terms contributing to the net reaction rate for \( Y_s \) in Eq. (2) are investigated along Line A in Fig. 1. Figure 3 shows the profiles, indicating that the overall soot formation mainly comes from the surface growth reaction except near the oxidizer-rich zone where the oxidation rate becomes significant. Although not shown, consistent behavior was found in other regions of the flame, such as along Lines B and C.
C in Fig. 1. Therefore, for the semi-empirical soot model considered in this study, it is concluded that the surface growth reaction is the most dominant chemical path, and will be considered a representative rate for soot formation for subsequent discussion.

Considering Figs. 1 and 2, it is evident that the significant enhancement in \( f_v \) in turbulent flames is not entirely caused by the increased chemical production; additional effects such as mixing and transport must be also taken into account. Figure 4 shows the budget analysis of various terms contributing to \( f_v \) appearing in Eq. (1). Results along Lines A–C indicated in Fig. 1 represent three typical situations encountered in wrinkled nonpremixed flames. Along Line A, the local maximum \( f_v \) is 2.3 ppm, which is almost twice as large as that in the steady flame (~1.26 ppm). The soot enhancement in this region is caused by the local convective terms as well as by the soot formation (~0.8 kg/m³s) in the flame. It was found that the maximum \( f_v \) in this local soot pocket subsequently reaches even up to 3.0 ppm, which is approximately 2.4 times larger than the corresponding steady value. In contrast, along Line B, the local maximum \( f_v \) is reduced to 0.37 ppm. Since this region is subjected to high tangential strain rate at approximately 600/s compared to the mean steady value (approximately 280/s), the soot formation is counter-balanced by the convective loss, resulting in a smaller amount of \( f_v \). Along Line C, local maximum \( f_v \) is increased up to 1.8 ppm but the soot formation rate is almost negligible in the region of high \( f_v \). This implies that a large amount of soot convected toward fuel rich zones maintains its level without further soot generation or oxidation reactions. These results suggest that the instantaneous soot dynamics in turbulent flames can be affected by the intricate coupling between the soot chemistry and transport.

To further investigate this issue, the behavior of \( f_v \) is mapped as a function of the mixture fraction, \( \xi \), following the definition by Bilger [26]. Figure 5 shows a scatter plot of \( f_v \) in terms of \( \xi \) for the entire cumulative data up to 32 ms, with an increment of 0.25 ms. The white line denotes the initial steady state profile. As the flame and turbulent eddies interact, large scatters are observed in \( f_v \), deviating from the steady behavior. These data points can be categorized into three distinct groups, referred to as Paths A–C. As will be discussed later, these three paths are closely related to Lines A–C shown in Fig. 1.
To characterize the distinct paths of soot formation, the data points shown in Fig. 5 were conditionally sampled based on two additional parameters: the scalar dissipation rate, $\chi = 2\phi |\nabla \phi|^2$, where $\phi = \lambda/\rho c_p$ is the local thermal diffusivity [14]; and the soot surface growth rate, $\dot{\omega}_{\text{surf}}$. The two parameters, respectively, represent the characteristic mixing and reaction rates determining the soot formation process.

Figure 6 shows that the cumulative data points shown in Fig. 5 can be clearly distinguished into the three paths: (a) Path A for low mixing and high reaction ($\chi < 30 \text{ s}^{-1}$ and $\dot{\omega}_{\text{surf}} > 0.1 \text{ kg/m}^3\text{s}$), (b) Path B for high mixing and ($\chi > 100 \text{ s}^{-1}$) and (c) Path C for low mixing and low reaction ($\chi < 30 \text{ s}^{-1}$ and $\dot{\omega}_{\text{surf}} < 10^{-4} \text{ kg/m}^3\text{s}$). The data points not included in any of these paths fall close to the steady behavior. Therefore, the three paths shown here represent the data points that deviate significantly from the steady laminar values. Note that the maximum values of $\chi$ and $\dot{\omega}_{\text{surf}}$ of the steady flame are about $93 \text{ s}^{-1}$ and $1.2 \text{ kg/m}^3\text{s}$, respectively.

Along the Path A (Fig. 6a), the maximum $f_v$ grows up to 3.0 ppm due to the additive effects of both the enhanced surface growth rate and increased convective transport into the region (negative stretch). This behavior is represented by Line A in Fig. 1, where the compressive flow strain (as high as 2000/s) broadens the flame, rendering it ripe for enhanced soot generation by creating a larger high-temperature region. Along Path B (Fig. 6b), which typically corresponds to situations represented by Line B in Fig. 1, soot reaction rate is significantly reduced from the steady value due to the large tangential strain rate. This represents a typical example of reaction suppression by positive strain rate. Finally, Path C (Fig. 6c) corresponds to the regions at low mixing and reaction rates. The many data points showing enhanced soot volume fraction in this path are represented by Line C in Fig. 1, where a soot pocket originating from the flame zone is convected into the cold and fuel-rich region. As indicated in Fig. 4c, the positive convective transport appears to be primarily responsible for the enhanced soot volume fraction in this region.

Recalling Figs. 4c, however, it is found that a large part of the $f_v$ enhancement in Path C is simply due to the temperature drop in the gas phase fluid surrounding the soot particles. Consequently, the local $f_v$ increases relative to the surrounding gases even if the soot mass remains the same. Therefore, the large scatter in Path C shown in Fig. 6c may not necessarily result in the corresponding increase in the total soot mass.
From Fig. 6, it is also noted that Paths A and C are statistically the dominant mechanisms for the $f_v$ enhancement in turbulent flames. Path A is more prominent in the relatively fuel-lean region (near $\xi = 0.20$), while Path C is more important in the highly fuel-rich region (near $\xi = 0.75$). In general, path A tends to yield larger local peaks in $f_v$.

Other simulation cases show consistent behavior. Figure 7 shows the similar scatter plots along with typical instantaneous snapshots of $f_v$ for Cases 2 and 3. For Case 2 which has smaller $L_{11}$ than Case 1, the soot distribution spreads out more widely with a larger level of scatters compared to Case 1. This is attributed to the fact that smaller turbulent eddies are more effective in the mixing process that is responsible for the local increase in $f_v$. In contrast, for Case 3 in which inlet velocity and mean strain rate are twice as large as those in the other cases, the data scatter is significantly reduced. In particular, the soot enhancement along Path C is hardly visible. This is because the flame thickness is reduced due to the larger strain rate, such that the ratio of the turbulence to flame length scales is decreased. Consequently, the scatter in the data is significantly mitigated compared to the other cases and the overall behavior follows more closely to the laminar flame behavior.

3.2. Overall soot characteristics

It is of practical interest to investigate the integrated effect of the detailed transient characteristics on the overall soot yield. Figure 8 shows the integrated $N_s$ versus the flame volume defined as the region at temperature above 2000 K. The choice of different definition of the flame volume was from the fact that the soot particle generation process is dominated by the temperature-sensitive nucleation reaction. All quantities are normalized by the corresponding steady values. Each data point represents the quantity integrated over the entire computational domain for an instantaneous solution field. It is clearly seen that a strong correlation exists between the integrated $N_s$ and the flame volume. Since most soot particles exist in high temperature region and quickly coagulate in low temperature region, the overall $N_s$ is hardly affected by turbulent mixing and flow conditions but depends predominantly on the high temperature flame volume. Note that the strong correlation between the soot number density and high temperature zones may be primarily due to the formation of small nascent soot particles in the adopted simple chemistry model. In an actual flame, the temperatures in the rapid soot formation zones may be somewhat lower than implied here, since soot formation occurs on the rich side of the flame somewhat removed from the highest temperatures. Nevertheless, the results suggest that the number of soot particles is relatively small in the low temperature regions as in Path C in Fig. 5.

Figure 9 shows the integrated soot mass production rate as a function of the total flame volume defined as the region at temperature above 1300 K. As before, all quantities are normalized by the corresponding steady values. As a general observation, the soot production rate is larger than the steady value for all turbulent cases, implying that more soot yield is expected from turbulent flames. Furthermore, the good correlation against
Fig. 9. The integrated soot production rate as a function of the flame volume ($T > 1300$ K) for Cases 1–3.

The flame volume suggests that the increased soot production rate in turbulent flames is primarily due to the increased flame volume by turbulence. Incidentally, some data points that fall below the correlation lines (the data points appearing as a loop) are caused by the fact that some high temperature regions move out of the computational domain, such that the integrated soot production rate drops suddenly.

As a more detailed analysis, we now consider the effect of the relative ratio of the turbulence to flame scales. Figure 9 shows that the integrated soot production rate increases as the relative turbulence length scale is increased (Case 2 → 1 → 3). By inspecting the instantaneous solution fields, it is apparent that larger eddies effectively generate a larger flame volume at high temperature, while smaller eddies tend to break into the laminar flame structure to induce more rapid dissipation of the soot pockets. Note that, although the integrated soot production rate is higher than the steady value, it is less than the soot yield rate of the corresponding laminar flame with the same flame volume, which implies that turbulence mixing not only enhances soot yield by increasing flame volume but also reduces the soot chemistry by rapid transport of soot pockets out of the high temperature regions. For case 2, the data spreads out more.

From the above results, it is thus concluded that the effects of turbulence on soot formation are twofold: (a) soot enhancement by increased flame volume and (b) reduction of soot chemistry by rapid transport of soot pockets out of the high temperature regions. Based on this observation, it is conjectured that a nonmonotonic soot response may occur as the turbulence intensity is increased. Since the increased turbulence intensity results in reduced Kolmogorov eddy scale, the soot yield increases first due to the flame volume generation and then drops down as the effect of dissipation starts to override the flame volume enhancement. These two effects of turbulence must be properly accounted for in the soot formation model in large scale simulations.

4. Conclusions

The effects of turbulence-chemistry interaction on the transient soot dynamics was investigated via direct numerical simulations of turbulent ethylene–air nonpremixed counterflow flames using a semi-empirical soot model and the discrete ordinate method for gray gas radiation. Detailed analysis of data revealed that the effects of turbulence on the soot formation can be categorized into three distinct paths depending on the relative strength between mixing and reaction. The effects of turbulence on the overall soot yield characteristics were also investigated. It was found that the soot number density behavior is predominantly controlled by the high temperature flame region. In general, turbulence has twofold effects of increasing the overall soot yield by generating additional flame volume and of reducing soot by dissipating soot pockets out of high-temperature regions. It was found that the relative importance between the two effects depends on the relative length scales of turbulence and flame. These results demonstrate that the detailed information of the local and transient flow-chemistry interaction is important in the accurate prediction of the soot formation characteristics.

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