Title
Characteristics of counterflow premixed flames with low frequency composition fluctuations

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Author names
Takuya Tomidokoro\textsuperscript{a,*}, Takeshi Yokomori\textsuperscript{b}, Hong G. Im\textsuperscript{c}, and Toshihisa Ueda\textsuperscript{b}

Affiliation of Authors
\textsuperscript{a}School of Science and Technology, Keio University, Japan
\textsuperscript{b}Department of Mechanical Engineering, Keio University, Japan
\textsuperscript{c}Clean Combustion Research Center, King Abdullah University of Science and Technology, Saudi Arabia

* Corresponding author
E-mail address: takuyatomidokoro@keio.jp (T. Tomidokoro)
1. Introduction

To address increasingly strict emission requirements and the urgent need for higher efficiencies, stratified combustion is now used widely in modern internal combustion engines [1–4] and gas turbines [5,6]. Stratification plays a crucial role in controlling combustion processes in not only conventional direct-injection spark-ignition engines [3] but also advanced compression ignition engines [1,4], a promising strategy for increasing efficiency and suppressing emissions [2]. For aircraft and power-generation gas turbines, lean premixed or lean premixed prevaporized combustors have been developed to reduce NO$_x$ emissions [5–7]. However, such combustors are known to be prone to thermo-acoustic instabilities, whereby transient variations in the mixture composition play a major role in driving the feedback loops [8]. Furthermore, to stabilize the flame, such combustors are often operated in stratified configurations [5,7]. Therefore, it is of both fundamental interest and practical importance to elucidate the mechanism for flame propagation in an inhomogeneous mixture.

One of the well-known basic local effects under mixture stratification is the “back-support” effect, where the flame speed is modified when the flame propagates into a mixture with an equivalence ratio gradient in the direction of its movement [9]. Extensive research has been done to reveal the mechanism behind it. Kang and Kyritsis [10–12] conducted a series of experiments involving the one-dimensional propagation of a methane/air flame in a counterflow tubular burner and found that the flame propagated faster in mixtures that varied from being approximately stoichiometric to being gradually leaner or richer. From theoretical analysis and model development, they concluded that in both cases, the flame was thermally supported by higher temperature on the stoichiometric side. Meanwhile, recent numerical analysis of one-dimensional stratified flame propagation in a rich-to-lean mixture by Shi et al. [13–15] concluded that preferential diffusion of H for hydrogen fuel and H$_2$ for hydrocarbon fuels is the key factor in modifying the flame speed. They further developed a stratification model based on H$_2$ accumulation and successfully predicted the flame speed for monotonic and non-monotonic stratifications. Several studies have identified different mechanisms for stratification in lean and rich mixtures. Da Cruz et al. [16] conducted a numerical analysis of one-dimensional flame propagation and concluded that H$_2$ and H diffusion into the reaction zone is the key factor in modifying the flame speed, although in lean cases it is caused by larger thermal diffusion into the unburned mixture. A numerical study of steady counterflow methane/air flames by Zhou and Hochgreb [17] also concluded that H$_2$ and thermal diffusion for rich and lean cases, respectively, are dominant causes for the back-support effects. In contrast, with a similar numerical setup, Richardson et al. [18] concluded that in lean conditions, the diffusion of radicals such as OH is responsible for modifying the heat release, which is caused by increased H$_2$ and O$_2$ decomposition reactions under higher burned gas temperature.

As mentioned above, conclusions regarding the cause of the back support effect are somewhat controversial, especially in the lean region where it is unclear whether the dominant
factor in modification of the reaction rate is variation in thermal or species diffusion from the
burned gas. Therefore, it is of interest to see if the back-supporting phenomena could be
observed with a global reaction mechanism wherein detailed species diffusion effects are
excluded.

Also, several studies have reported the presence of a memory effect whereby the flame
response to an imposed equivalence ratio gradient is delayed in non-monotonic stratification
[10,14,19], but that mechanism is yet to be examined in detail. In particular, the effects of non-
on-monotonic stratification with an oscillating equivalence ratio—a situation that is important
for turbulent stratified flame dynamics in the laminar flamelet regime—are yet to be revealed.
Contrary to previous studies that have focused on an unsteady flame response under high-
frequency oscillation [20–23], a quasi-steady flame response under low-frequency oscillation is
of particular interest because the flame would have enough time to experience the back-
support effect. The low-frequency oscillation is also expected to be more practically relevant in
stratified combustion: the length scales of the mixture inhomogeneities are unlikely to be
smaller than the flame thickness.

Therefore, the objectives of this study are to compare the periodically-stratified flame speed
response under low-frequency equivalence ratio oscillations normal to the flame front as
obtained using a detailed reaction mechanism with that obtained using a global reaction
mechanism and to examine the roles of species diffusion under non-monotonic stratification.
The analysis is conducted separately in lean and rich regions to elucidate different
mechanisms for the back-support effects.
2. Numerical setup
2.1. Numerical model

A premixed version of the OPUS code [24] was used for the transient calculation of axisymmetric reactant-to-reactant counterflow flames. Fig. 1 shows a schematic of the counterflow geometry. Only the lower half of the geometry was calculated, considering symmetry. The governing equations were reduced to one-dimensional form under boundary layer approximation with relaxed isobaric assumption. Diffusion velocities were calculated by the mixture-averaged Fickian formula with simplified Soret effects for H and H₂, and thermal radiation was neglected. The initial condition and steady cases were calculated with the OPPST code included in the OPUS package. Both were interfaced with the CHEMKIN [25] and TRANSPORT [26] libraries for the calculation of reaction rates as well as thermodynamic and transport properties. OPUS employs adaptive time-stepping and integration order control for transient calculation. To examine the roles of certain reactive species, both detailed and global reaction mechanisms were adopted. A uniform grid, 5 μm for the global mechanism and 10 μm for the detailed mechanism, was adopted. The finer grid for the global mechanism was adopted to resolve a narrower reaction zone.

At the stagnation boundary (x = 0 mm), the axial velocity u was fixed at 0 m/s and zero-gradient conditions were applied for other variables. At the inlet (xᵢₙ = −21 mm), the axial velocity, temperature, and thermodynamic pressure were fixed at 0.5 m/s, 300 K, and 1 atm = 0.101325 MPa, respectively. The magnitude of the inlet velocity was chosen to suppress flow straining as much as possible, but be large enough to stabilize the flame under imposed equivalence ratios. In the periodically-stratified cases, the mixture composition, consisting of methane and air (mole fraction O₂:N₂ = 0.21:0.79), was varied so that the inlet equivalence ratio φᵢₙ oscillated in a sinusoidal manner as expressed by

\[ φᵢₙ = φₘ + φₐ \cos(2\pi ft), \]

where \( φₘ, φₐ, \) and \( f \) are respectively the mean equivalence ratio, amplitude, and frequency of the oscillation.

Fig. 1. Schematic representation of counterflow premixed flame and computational geometry.
2.2. Reaction mechanisms

To examine the effect of the influx of reactive species from the product side of the flame, the flame responses with detailed and global methane/air oxidation mechanisms were compared. For the detailed mechanism, GRI-Mech 3.0 [27] was used. For the global mechanism, the following four-step mechanism developed by Jones and Lindstedt [28] was selected:

\[
\begin{align*}
\text{CH}_4 + 0.5\text{O}_2 & \rightarrow \text{CO} + \text{H}_2 & \quad \text{(JL1)} \\
\text{CH}_4 + \text{H}_2\text{O} & \rightarrow \text{CO} + 3\text{H}_2 & \quad \text{(JL2)} \\
\text{H}_2 + 0.5\text{O}_2 & \leftrightarrow \text{H}_2\text{O} & \quad \text{(JL3)} \\
\text{CO} + \text{H}_2\text{O} & \leftrightarrow \text{CO}_2 + \text{H}_2 & \quad \text{(JL4)}
\end{align*}
\]

Notice that the above mechanism does not include any major radical species such as H, OH, or O, which would otherwise back-diffuse into the reaction zone. For the Arrhenius parameters and reaction orders, refer to Table 2 of Andersen et al. [29].

Adopting the detailed and global mechanisms, the consumption speed \( S_c \) was evaluated for steady flames with various \( \phi_{in} \) and was compared with the experimental results for unstretched flames [30], as shown in Fig. 2. \( S_c \) was defined by the following expression [31]:

\[
S_c = \frac{1}{\rho_u Y_{F,u}} \int_{-\infty}^{0} (-W_F \dot{Y}_F) dx,
\]

where \( W_F, \rho_u, \) and \( Y_{F,u} \) are respectively fuel molecular weight, upstream mixture density, and upstream fuel mass fraction, and \( \dot{Y}_F \) is the fuel mole production rate. In the above expression, \( Y_{F,u} - Y_{F,B} \) of the original definition [31], where \( Y_{F,B} \) is the fuel mass fraction on the burned side, was replaced by \( Y_{F,u} \) for consistency with the definition for stratified cases, which will be discussed in Section 3.4.

Although a good agreement with the experimental results was obtained with the detailed mechanism, the global mechanism substantially overestimated the consumption speed, especially in the rich region. Such deviation is due to the absence of the dissociation reactions of \( \text{H}_2 \) and \( \text{H}_2\text{O} \) [32]. However, the qualitative behavior of the consumption speed response is captured. In Fig. 3, the maximum temperature and mole fractions of the reactive species, namely, \( \text{H}_2, \text{H}, \text{OH}, \) and \( \text{O} \) for the detailed mechanism and \( \text{H}_2 \) for the global mechanism, at the flame location \( \chi_f \) are shown for various inlet equivalence ratios. Here, the flame location \( \chi_f \) is defined as the location of maximum heat release, interpolated between two grid points around the peak. Although the global mechanism overestimates the \( \text{H}_2 \) mole fraction in the rich region because of the aforementioned absence of \( \text{H}_2 \) dissociation, the \( \text{H}_2 \) mole fraction increases with \( \phi_{in} \) for both mechanisms.

Since the back-support effect is induced by relative variation in the flame properties under varying mixture compositions, no quantitative match of the flame properties between different mechanisms is required for comparison. In the following analysis, the transient response of flame speed to equivalence ratio variations are qualitatively contrasted between different reaction mechanisms, while a quantitative comparison of flame speed and structure is conducted only between flames with the same mechanism.
**Fig. 2.** Comparison of the consumption speed $S_c$ between experimental results [32] and steady numerical results obtained with the detailed and global mechanisms under the mixture equivalence ratio $\phi_{in}$.

**Fig. 3.** Comparison of maximum temperature and species mole fraction at the flame location $x_f$ for steady flames obtained with the detailed and global mechanisms under mixture equivalence ratio $\phi_{in}$. 
3. Evaluation of the back-support effect

3.1. Notes on the stretch effect

Unlike in previous studies of transient one-dimensional flame propagation [13–16,33,34], the flame is stretched in the present counterflow configuration. Fig. 4 shows the consumption speed $S_c$ of steady counterflow flames, normalized by one-dimensional laminar flame speed obtained with PREMIX solver [35], versus different local stretch rates, evaluated from the local radial velocity gradient at $x_f$. While the detailed mechanism resulted in qualitatively similar behavior to previous reports [36,37], the global mechanism failed to reproduce the response curve. However, this is not an issue for the current study because of the following reason. Although the inlet velocity is fixed in the transient analysis, the local stretch rate fluctuates due to variation in the heat release rate and consequently the flow expansion ratio. The shaded area in the figure corresponds to the maximum range of local stretch rate fluctuation in the transient analysis. It can be seen that the variation in $S_c$ caused by stretch is less than 1 % for all conditions, which is small enough as compared to the maximum magnitude of $S_c$ modification, as will be shown later. Consequently, the stretch effect is ignored in the following analysis. However, there have been relatively few studies of how stretch affects stratification [9,17], and it may be an interesting topic for future work.

![Normalized flame speed response](image)

**Fig. 4.** Normalized flame speed response of steady counterflow flames with detailed and global mechanisms in lean ($\phi_{in} = 0.8$) and rich ($\phi_{in} = 1.2$) mixtures. The shaded area represents the maximum range of local strain rate fluctuation in the following transient analysis.

3.2. Equivalence ratio

To evaluate the back-support effect, we compare a periodically-stratified flame (SF) under equivalence ratio oscillation with a series of corresponding steady homogeneous flames (HFs) that experience the same mixture composition as that of the SF at each instant. The conventional species-based definition of the equivalence ratio cannot represent the unburned
mixture composition at $x_f$, where fuel has been consumed. Therefore, an element-based equivalence ratio $\phi$ has been used in previous studies [13–15,17],

$$\phi = \frac{4Z_C + Z_H}{2Z_O},$$

(3)

where $Z_C$, $Z_H$, and $Z_O$ are the mole fractions of C, H, and O atoms, respectively.

Contrary to the conventional definition, the element-based definition retains its unburned value even on the product side of the flame. Following a common method applied in previous studies on laminar stratified flames [13–18,34], the value of $\phi$ at the flame location, $\phi_f$, was matched to for SF and HF at each value of $\phi_f$, so that their properties are compared under the same mixture composition.

3.3. Fuel consumption speed

To determine the flame speed in a transient counterflow field, the consumption speed as given by Eq. (2) has been reported to be a better choice than the displacement speed [31] given that the latter fails to represent the chemical processes in the reaction zone [38,39]. Consequently, previous numerical studies on a transient counterflow geometry have often applied the consumption speed as a measure of the flame speed [20,38–41]. However, some difficulties arise when the consumption speed is evaluated under transient variation in the mixture composition. First, especially in rich cases, the variation in the fuel mass fraction, $Y_{F,u} - Y_{F,b}$, cannot be evaluated unambiguously because the value of $Y_{F,b}$ is uncertain on the product side. Therefore, following the convention adopted in previous studies [13–16], $Y_{F,u} - Y_{F,b}$ is replaced by $Y_{F,u}$. Such simplification is fairly reasonable because $Y_{F,b}$ is relatively small even in the rich region around stoichiometry. Another difficulty arises when evaluating $\rho_u Y_{F,u}$ because $\rho_u Y_{F,u}$ varies spatially throughout the fuel consumption layer at every moment. In the present study, because the fuel consumption layer is sufficiently thin as compared to the variation in the mixture composition, the characteristic value of $\rho_u Y_{F,u}$ was estimated from $\phi_f$ and the temperature at $x_f$.

3.4. Equivalence ratio gradient

Because the back-support effect is fundamentally a diffusive process, it seems feasible that its magnitude can be described based on the spatial gradient of $\phi$ [13,17]. However, the local gradient at $x_f$, namely the reaction zone, cannot appropriately represent the global gradient imposed on the flame because preferential diffusion of intermediate species cause the element-based $\phi$ to fluctuate around the reaction zone [18]. Instead, we define the equivalence ratio gradient at the flame location $(\partial \phi / \partial x)_f$ as the time derivative of $\phi_f$ divided by the distance that the flame propagates relative to the flow, namely

$$
(\frac{\partial \phi}{\partial x})_f = \frac{-(\phi_f/\partial t)}{u_f - dx_f/dt},
$$

(4)

where $u_f$ is the axial flow velocity at $x_f$. 
4. Results and discussion

The magnitude of the back-support effect is evaluated by comparing the consumption speed $S_c$ of an SF under equivalence ratio oscillation with the $S_c$ of a steady HF at each $\phi_f$ values. The oscillation parameters considered in the following analysis are listed in Table 1. Two types of analyses were conducted for both lean and rich mixtures. In Section 4.1, using the detailed reaction mechanism, flame response to periodical stratification under oscillation frequencies of 1.25, 10, and 80 Hz were compared. Here, a larger oscillation amplitude $\phi_a$ was imposed for the 80 Hz case as compared to $\phi_a$ of lower frequencies, so that the effective amplitude of $\phi_f$ felt at the flame location $x_f$, which would be smaller than $\phi_a$ at the inlet due to attenuation, is approximately the same as the low-frequency cases. Then in Section 4.2 and 4.3, flame responses with the detailed and the global reaction mechanisms were compared, and the roles of species diffusion in back-supported flames were examined.

All of the oscillation timescales are sufficiently longer than the flame timescales, which are on the order of 1 ms, thus the flame responds quasi-steadily for all frequencies under consideration. As mentioned above, any higher frequency would result in a significant attenuation of the amplitude before it reaches the flame. Therefore, 80 Hz was chosen as the maximum frequency in the current analysis.

The results are presented for one cycle of the SF response after it reached a limit cycle. In the current study, the maximum equivalence ratio gradient of the unburned mixture was around $\pm 3.5$ cm$^{-1}$, which is on the order of the magnitude in actual conditions, for example around 1 cm$^{-1}$ in homogeneous charge compression ignition engines [33].

<table>
<thead>
<tr>
<th>Reaction mechanism</th>
<th>$\phi_m$</th>
<th>$\phi_a$</th>
<th>$f$ [Hz]</th>
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<td>4.1. Frequency response</td>
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<td>Detailed</td>
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<td>4.2. Roles of species diffusion – lean mixture</td>
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<tr>
<td>Detailed</td>
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<tr>
<td>Global</td>
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<td>4.3. Roles of species diffusion – rich mixture</td>
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<tr>
<td>Detailed</td>
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<td>Global</td>
<td>1.35</td>
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4.1. Frequency response

Fig. 5 shows the responses of the consumption speed $S_c$ and the equivalence ratio gradient $(\partial \phi / \partial x)_f$ of the SF with the detailed mechanism to different oscillation frequencies, along with the corresponding HF properties.

For both the lean and rich mixtures, while low-frequency cases result in the same value of $S_c$ as the corresponding HFs, the variation in $(\partial \phi / \partial x)_f$ is amplified with increasing frequency, and $S_c$ deviates from the steady case. As expected, it is the amplitude of $(\partial \phi / \partial x)_f$ variation that directly affects the magnitude of the back-support, rather than the amplitude of $\phi_f$ variation. In both cases, decreasing (increasing) $\phi_f$, or positive (negative) $(\partial \phi / \partial x)_f$, leads to an increase (decrease) in $S_c$ as compared to that of the corresponding HFs. The above behaviors agree with previous reports [14,33,34] and, especially in the present study, results in an increased phase delay of $S_c$ relative to the oscillating $\phi_f$ as the frequency is increased. The phase delay has also been reported in previous studies [20,22], but with oscillation timescales as short as the order of the flame timescale. It is confirmed here that even quasi-steady flame response to imposed equivalence ratio oscillation can be delayed due to the back-support effect. Furthermore, rich cases show a larger deviation than do lean results for the same $(\partial \phi / \partial x)_f$, as also reported in previous studies [14,33,42].
Fig. 5. Responses of consumption speed $S_c$ and equivalence ratio gradient $(\partial \phi/\partial x)_f$ of SF, compared with those of the HFs, under different oscillation frequencies in (a) lean and (b) rich mixtures respectively.
4.2. Roles of species diffusion – lean mixture

In Section 4.1, it was shown that when the flame propagates away from the stoichiometric condition in the lean region, the consumption speed $S_c$ is increased from that of the corresponding HF s. As shown in Fig. 3, the flame temperature is higher, and the amount of reactive species is larger around the stoichiometric condition. Therefore, possible causes of the increase in $S_c$ are an increase in the diffusion of heat and/or reactive species from the burned side of the flame. Conversely, when the flame propagates toward stoichiometric condition, the $S_c$ is decreased from that of HFs due to a lack of back-diffusion of heat and/or reactive species.

In the present section, such roles of the reactive species are examined through different reaction mechanisms and subsequent analysis with modified diffusion coefficients.

4.2.1. Consumption speed

Fig. 6 shows the responses of $S_c$ and $(\partial \phi / \partial x)_f$ of SFs with the detailed and global mechanisms at a fixed frequency of 80 Hz, along with $S_c$ of the steady HF at each $\phi_f$. With the detailed mechanism, the variation in $(\partial \phi / \partial x)_f$ leads to a deviation from the steady $S_c$. Meanwhile, with the global mechanism, the SF continues to exhibit the same value of $S_c$ as the HFs despite the similar amplitude of $(\partial \phi / \partial x)_f$ variation.

For quantitative discussion, Fig. 7 shows the responses of the relative deviation of the fuel consumption speed of SFs, $S_{c, SF}$, from that of the HF s, $S_{c, HF}$, at each value of $\phi_f$. The results with the detailed mechanism show a positive correlation between $(\partial \phi / \partial x)_f$ and the deviation. Also, the phase delay between the two has been reported as the “memory effect” of non-monotonic stratification [10,14,19], which is a hysteresis of the back-support effect under transient $(\partial \phi / \partial x)_f$ variation. Meanwhile, the corresponding results with the global mechanism show a smaller, if not negligible, variation in $S_c$, which does not correlate with the variation in $(\partial \phi / \partial x)_f$. 
Fig. 6. Responses of consumption speed $S_c$ and equivalence ratio gradient $(\partial \phi / \partial x)_f$ of SF, compared with those of the HFs, with (a) detailed and (b) global mechanism in a lean mixture.
Fig. 7. Responses of relative deviation in consumption speed $S_c$ in a lean mixture with detailed and global mechanisms.

4.2.2. Transient flame structures

In Fig. 8, the transient flame structures of the SF are visualized at the moments DL1–4 and GL1–4 indicated in Figs. 6 and 7, in comparison with those of the corresponding HFs. The odd indices represent the moments of maximum or minimum $(\partial \phi / \partial x)_f$, and the even indices represent the moments when $(\partial \phi / \partial x)_f = 0$. Consequently, DL1/GL1 (DL3/GL3) represents the moments with the largest decreasing (increasing) equivalence ratio gradients toward the burned gas. Meanwhile, DL2/GL2 and DL4/GL4 represent the moments when the flame is at the peak and trough locations of the equivalence ratio oscillation, but may still be under the influence of modified temperature or species profiles on the burned side.

For both mechanisms, the heat release rate response corresponds to the $S_c$ response in Fig. 6: compared to the corresponding HF profiles, the heat release rate is decreased (increased) at DL1 (DL3) with the detailed mechanism, while no relative variation is observed with the global mechanism for all moments. Similar responses are observed for the species profiles. On the other hand, with both mechanisms, the burned gas temperature profiles are lower (higher) at DL1/GL1 (DL3/GL3).

The above behavior implies that both species and thermal back-diffusion are present for the SF with the detailed mechanism, while the thermal effect plays the primary role with the global mechanism. Consequently, contrary to the findings in previous studies [10,16,17,43], the variation in the burned gas temperature does not seem to help modify the reaction rate. As such, we will make a hypothesis that the back-diffusion of light reactive species such as $H_2$ and $H$, $OH$, and $O$ radicals play dominant roles in modifying the reaction rate under stratification. In the next part, the roles of these species are verified through an analysis of
flame response with modified diffusion coefficients.

Furthermore, profiles at DL2 and DL4 show that even when the instant equivalence-ratio gradient is zero, the flame is still under the influence of modified species profiles as compared to the HFs, leading to a phase delay of the back-support effect to the imposed \((\partial \phi / \partial x)_{f}\) variation, or the “memory effect”, as seen in Fig. 7. This aspect is revisited in Section 4.3.

### 4.2.3. Modification of diffusion coefficients

Following the methodology by Shi and Chen [14], in order to verify the influence of species back-diffusion into the reaction zone, diffusion coefficients were modified for SFs and HFs with the detailed mechanism, and the resulting magnitude of the back-support effect for each case was qualitatively compared. Fig. 9 shows the \(S_c\) response of SFs under the equivalence ratio oscillation of 80 Hz along with the \(S_c\) of HFs at each \(\phi_f\). Case (a) is the reference case with standard diffusion coefficients, while cases (b-f) are the modified-coefficient cases. Here, the inlet oscillation amplitudes \(\phi_a\) of the cases (b-f) were adjusted so that the average amplitudes of \((\partial \phi / \partial x)_{f}\) variation for each case were within \(\pm 0.005\) 1/mm of that of the reference case.

Comparing the reference case (a) with case (b), in which Lewis numbers were modified so that \(Le_{H,OH,O} = 1\), case (b) shows smaller deviations from the HF curve, implying that the preferential diffusion of H, OH, and O radicals amplify the magnitude of the back-support. Similarly, in case (c), in which \(Le_{H_2} = 1\), it is suggested the preferential diffusion of \(H_2\) also amplifies the back-support. Furthermore, examining case (d), in which \(Le\) for both \(H_2\) and the radicals are modified to unity, the back-support is not observed: a similar behavior was obtained for case (e), in which \(Le\) for all species were set to unity. From the above results, it can be concluded that the preferential diffusion of both \(H_2\) and the radicals is the primary cause of the back-support in a lean mixture.

Additionally, the result of case (f), in which diffusion coefficient of \(H_2\) was set to zero, also shows that even when there is no \(H_2\) diffusion into the reaction zone, the back-support is still induced solely through variation in the downstream radical profiles. As discussed later in Part 4.3.3, this turns out to be an essential difference in the back-supporting mechanism in lean and rich mixtures.
Fig. 8. Temperature $T$, heat release rate $Q$, and mole fractions of $H_2$, $H$, $OH$, and $O$ for the SF and corresponding HFs with the detailed (upper row) and global (lower row) mechanisms at the moments indicated in Figs. 6 and 7. The coordinates are shifted to match the flame location at $x = 0$. 
Fig. 9. Responses of consumption speed $S_c$ of SF along with that of the HFs in a lean mixture with (a) normal diffusion coefficients and (b–f) modified diffusion coefficients: (b) $L_e_{\text{H, OH, O}} = 1$, (c) $L_e_{\text{H}_2} = 1$, (d) $L_e_{\text{H}_2, \text{H, OH, O}} = 1$, (e) $L_e = 1$ for all species, and (f) $D_{\text{H}_2} = 1$. 

\[ S_c [\text{cm/s}] \]

\[ \phi_i \]
4.3. Roles of species diffusion – rich mixture

In Section 4.1, when the flame propagates toward the stoichiometric condition in a rich mixture, the consumption speed $S_c$ was increased from that of the corresponding HFs. As seen in Fig. 3, the flame temperature and the amount of radical species $H$, $OH$, and $O$ peak around the stoichiometric condition and decreases with increasing equivalence ratio $\phi_{in}$, whereas the amount of $H_2$ monotonically increases with $\phi_{in}$. Consequently, an increase in the back-diffusion of $H_2$ is assumed to be the primary cause of the increase in $S_c$. Several previous studies have also ascribed the back-support effect in a rich mixture to the $H_2$ back-diffusion [14–17]. In this section, the mechanism of the stratification in a rich mixture is discussed from the perspective of the species diffusion.

4.3.1. Consumption speed

Fig. 10 shows the response of $S_c$ and $(\partial \phi / \partial x)_f$ of SFs with the detailed and global mechanisms at a fixed frequency of 80 Hz, along with $S_c$ of the steady HF at each $\phi_f$. As shown in Table 1, the mean equivalence ratio $\phi_m$ of the SF with the global mechanism was shifted from 1.20 to 1.35 so that the range of $S_c$ variation is approximately the same as that with the detailed mechanism, considering that the global mechanism overestimates $S_c$ in a rich mixture. Similar to the lean-mixture results, the SF with the detailed mechanism results in deviation of $S_c$ from that of HFs at each $\phi_f$, whereas with the global mechanism SF shows little deviation despite the same extent of variation in $(\partial \phi / \partial x)_f$.

With the detailed mechanism, the correlation between $(\partial \phi / \partial x)_f$ and the relative deviation of the fuel consumption speed, $(S_{c, SF} - S_{c, HF})/S_{c, HF}$, in Fig. 11 is considerably stretched in the direction of negative deviation, which indicates that the memory effect of back-support is much more substantial around the richer turning point (DR2) than around the stoichiometric turning point (DR4). This behavior is peculiar as compared to the relatively symmetric correlation in the lean region seen in Fig. 7. The cause of this memory effect is discussed in Part 4.3.2. Meanwhile, with the global mechanism, the relative deviation of $S_c$ is less significant and does not correlate well with the $(\partial \phi / \partial x)_f$ variation.
Fig. 10. Responses of consumption speed $S_c$ and displacement equivalence ratio gradient $(\partial \phi/\partial x)_f$ of the SF, compared with those of the HFs, with the (a) detailed and (b) global mechanisms in a rich mixture.
**Fig. 11.** Responses of relative deviation in consumption speed $S_c$ in a rich mixture with detailed and global mechanisms.

### 4.3.2. Transient flame structures

In Fig. 12, the transient flame structures are compared with those of the corresponding HF$s$ at the moments DR1–4 and GR1–4 in Figs. 10 and 11. A noticeable difference from the results in a lean mixture is observed in the SF structures with the global mechanism: as shown in GR1–4, the heat release rate and the H$_2$ profiles are clearly modified from those of corresponding HF profiles, despite the insignificant modification of $S_c$ in Fig. 10. In fact, as shown in Fig. 13, the response of the maximum heat release rate $Q_{\text{max}}$ of SF deviates substantially from that of the HF$s$ regardless of the reaction mechanisms.

Meanwhile, with both mechanisms, no relative variation is observed in the burned gas temperature profiles, which can be explained as follows: As the SF propagates toward stoichiometry (richer mixture), the burned gas temperature becomes lower (higher) than that of the corresponding HF. However, the lower (higher) temperature is compensated by the increase (decrease) in the heat release rate mentioned above. Accordingly, thermal diffusion is not responsible for modifying the SF structure.

Contrary to the expectation based on the equivalence ratio dependence in Fig. 3 that the H, OH, and O radical profiles of the SF should be increased (decreased) from those of HF at DR1 (DR3), they are actually decreased (increased) especially around the reaction zone for OH, and O radicals. As reported by previous studies [14,17], the distributions of those radical species were modified by the variation in the H$_2$ decomposition reactions, namely

- $\text{H} + \text{O}_2 \rightarrow \text{O} + \text{OH}$ \hspace{1cm} (D1)
- $\text{O} + \text{H}_2 \rightarrow \text{H} + \text{OH}$ \hspace{1cm} (D2)
- $\text{OH} + \text{H}_2 \rightarrow \text{H} + \text{H}_2\text{O}$ \hspace{1cm} (D3)
The same studies [14,17] also reported that the modification of the radical species profiles around the reaction zone increases or decreases the fuel consumption rate and the heat release rate. With the global mechanism, however, the variation in H₂ back-diffusion only modifies the heat release rate through the overall oxidation reaction (JL3) and ends up having little effect on the consumption speed $S_c$.

Lastly, with the detailed mechanism, the greater memory effect around the richer turning point DR2 than around the stoichiometric turning point DR4 is apparently due to the larger pool of H₂ and lower reaction rate, meaning that it takes longer to compensate for the previous back-support effect. In contrast, in an overall lean mixture, the larger H₂ pool corresponds to a higher reaction rate near stoichiometry, and thus the memory effect becomes less pronounced, which resulted in the similar magnitude of phase delay at DL2 and DL4 as seen in Fig. 7.
Fig. 12. Temperature $T$, heat release rate $Q$, and mole fractions of $H_2$, $H$, $OH$, and $O$ for the SF and corresponding HFs with the detailed (upper row) and global (lower row) mechanisms at the moments indicated in Figs. 10 and 11. The coordinates are shifted to match the flame location at $x = 0$. 
Fig. 13. Responses of maximum heat release rate $Q_{\text{max}}$ of the SF with the (a) detailed and (b) global mechanisms in a rich mixture, compared with the that of HFs.

4.3.3. Modification of diffusion coefficients

The roles of the radical species under periodical stratification is further examined through modification of the diffusion coefficients. Fig. 14 shows the $S_c$ response of SFs under 80 Hz equivalence ratio oscillation along with the $S_c$ of HFs at each $\phi_f$, similar to Fig. 9. Again, the inlet oscillation amplitudes $\phi_a$ of cases (b-f) were adjusted to match the $(\partial \phi/\partial x)_f$ variation amplitude with the reference case (a). It is seen in the figure that both in case (b), in which the Lewis numbers were modified so that $Le_{\text{H,OH,0}} = 1$, and case (c), in which $Le_{\text{H}_2} = 1$, the magnitude of $S_c$ deviation from the HF values are smaller than that in the reference case (a). On the other hand, the back-support effect is hardly observed for both cases (d) and (e), in which preferential diffusion of both $\text{H}_2$ and the radical species was omitted. Consequently, similar to the back-support in a lean mixture, the preferential diffusion of $\text{H}_2$ and radical species has also been shown to be a primary factor for the back-support in a rich mixture. This conclusion conforms to the report by Shi and Chen [14], who had conducted a similar study on a one-dimensional flame propagating in a rich-to-lean stratified mixture.

Furthermore, a larger magnitude of back-support is observed in case (b) than in case (c), which implies that the preferential diffusion of $\text{H}_2$ is more influential than that of radical species. In fact, when $\text{H}_2$ diffusion is entirely removed in case (f), the $S_c$ response of the SF is reversed from the standard back-supported SF behavior: When $\phi_f \gtrsim 1.2$, $S_c$ of the SF is decreased (increased) from HF values at each $\phi_f$ under decreasing (increasing) $\phi_f$. This is a consequence of richer (more stoichiometric) burned gas composition with a decreased (increased) amount of radical species. The above behavior confirms the previous conclusion that the radical species distribution is modified through variation in their production rate, rather than the burned gas composition.

The roles of $\text{H}_2$ and major radical species in a flame propagating under decreasing
equivalence ratio in lean and rich mixtures, which results in an increased $S_c$, are contrasted in Fig. 15. In a lean mixture, stratification induces both $H_2$ and the radical distributions to increase as compared to those of HFs, which in turn increases the back-diffusion of radicals into the reaction zone. On the other hand, in a rich mixture, while $H_2$ distribution is increased with the stratification, the distributions of radicals are decreased in the burned gas. However, due to the presence of a large $H_2$ pool in a rich flame, the increase in the radical production through the chain reactions (D1·3) overrides the decrease in the radical distribution around the reaction zone. Consequently, the net back-diffusion of radicals is increased as compared to that of HFs.

![Graph showing responses of consumption speed $S_c$ of SF along with that of the HFs in a rich mixture with modified diffusion coefficients.](image)

**Fig. 14.** Responses of consumption speed $S_c$ of SF along with that of the HFs in a rich mixture with (a) normal diffusion coefficients and (b–f) modified diffusion coefficients: (b) $Le_{H, OH, O} = 1$, (c) $Le_{H_2} = 1$, (d) $Le_{H_2, OH, O} = 1$, (e) $Le = 1$ for all species, and (f) $D_{H_2} = 1$. 
**Fig. 15.** Mechanism of the back-support effect on a flame propagating under decreasing equivalence ratio in (a) lean and (b) rich mixtures. A solid/striped arrow represents an increasing/decreasing effect in the direction of the arrow, respectively, and their thickness the magnitude of the effect.

4.4. Implications for turbulent flames under mixture inhomogeneity

Contrary to the previous findings [20–22] that focused on unsteady flame responses to high-frequency compositional oscillations, whose characteristic time scales are shorter than that of the flame, the present study targets low-frequency, quasi-steady flame responses to the oscillation. However, as shown above, such low-frequency compositional fluctuation can still lead to nonnegligible modification of the flame speed because of the back-support effect. The implication here is that the quasi-steady assumption of the flame response to compositional fluctuation does not necessitate the flame speed to be the value that corresponds to the local mixture composition.

Furthermore, the back-support effect can influence variation in the flame surface area when the flame propagates in inhomogeneous mixtures. The right-hand side of each case in Fig. 16 shows a flame surfaces perturbed by equivalence ratio variation, and hence different flame speeds, along the flame surface in overall lean and rich mixtures. When the flame propagates into the mixture on the left-hand side, where the equivalence ratio gradient along the flame surface is switched, the perturbation of the flame surface is attenuated. However, the rate of attenuation differs depending on the overall stoichiometry of the mixture. It has been shown in the previous sections that, when the flame propagates into a mixture with a lower/higher equivalence ratio, the flame speed is increased/decreased as compared to the flame speed without the back-support effect. In an overall lean mixture (Fig. 16(a)), decrease in the higher flame speed (upper half of the figure) and increase in the lower flame speed (lower half of the figure) are both delayed as compared to the flame with no back-support effect (broken line). This behavior results in a slower attenuation of the flame surface perturbation.

Contrarily, in an overall rich mixture (Fig. 16(b)), as the flame propagates into the left-hand
side, the decrease/increase in higher/lower flame speed is accelerated, leading to faster attenuation of the perturbation. However, as noted by Lipatnikov [9], the perturbation of the flame surface area due to mixture inhomogeneity may only be pronounced under weakly turbulent flames.

\[ S_c(\phi_1) > S_c(\phi_2) \]

**Fig. 16.** Attenuation of flame surface perturbation in a flame propagating under overall (a) lean and (b) rich mixture inhomogeneity. On the right-hand side of each case is the initial flame surface perturbed by equivalence ratio variation along the flame surface. On the left-hand side are the attenuated flame surface with (solid lines) and without (broken lines) the back-support effect. Note that \( S_c(\phi_1) > S_c(\phi_2) \).
5. Conclusions

The “back-support” effect on methane/air reactant-to-reactant counterflow flames under low-frequency equivalence ratio oscillation was studied numerically. The analysis was conducted separately in lean and rich mixtures. By using a fully detailed reaction mechanism, the consumption speed $S_c$ of periodically-stratified flames (SF) showed increasing deviation from $S_c$ of steady homogeneous flames (HFs) with larger variation in the equivalence ratio gradient $(\partial \phi / \partial x)_f$. That is, $S_c$ of the SF relatively increased (decreased) with decreasing (increasing) $\phi_f$, which caused a phase delay of $S_c$ response to the $\phi_f$ oscillation. It was shown that even when the imposed oscillation timescale is much longer than the flame timescale, the flame response can still be delayed under sufficiently large $(\partial \phi / \partial x)_f$ variation.

The roles of H$_2$ and radical species in the above phenomena were elucidated. The SF responses were compared with those with a four-step global mechanism that does not include H, OH, and O radicals. In a lean mixture, despite similar variations in the burned gas temperature for both mechanisms, the response of $S_c$ with the global mechanism showed negligible back-supporting phenomena. By analyzing the SF response with modified mass diffusion coefficients, it was found that preferential diffusion of H$_2$ and radical species played a dominant role in modifying the reaction rate. Contrarily to the previous findings [12,16,17], variation in burned gas temperature was found to play a minimal role in modifying $S_c$.

In a rich mixture, although the global mechanism resulted in much smaller modification of $S_c$ from the HF properties, the heat release rate and H$_2$ profiles were similarly modified from those of HFs regardless of the reaction mechanisms. Further analysis of diffusion coefficient modification has shown that modification of radical distributions around the reaction zone, which was induced by variation in H$_2$ decomposition rate, played an essential role in modifying the fuel consumption rate, confirming the previous findings. Additionally, the “memory effect,” or the hysteresis of non-monotonic back-support effect was revealed to be more prominent on the richer side in an overall rich mixture because of the larger pool of H$_2$ and the lower reaction rate.

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References


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