Brief Communications

Diffusive-thermal effect on local chemical structures in premixed hydrogen–air flames

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ABSTRACT

The diffusive-thermal effect plays an important role in the intrinsic instability of premixed flames. A two-dimensional direct numerical simulation of the propagation of premixed hydrogen–air flames was performed using a detailed chemical kinetics model. The cellular behavior of a lean hydrogen–air flame was analyzed on the basis of its chemical structure. The primary consequence of the diffusive-thermal effect was found to be a change in the buildup process of reactive intermediates by the chain reaction mechanism at the preheat zone. The resultant chemical structure at the main reaction zone can be explained by the local composition change of the gas flowing into the reaction zone from the preheat zone.

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1. Introduction

The intrinsic instability of premixed flames [1–4] affects its shape and structure, resulting in the formation of a cellular flame front and the acceleration of flame speed. Hydrodynamic instability caused by thermal expansion is a ubiquitous and essential factor in the flame instability. In addition, the diffusive-thermal effect due to nonequidiffusion of mass and heat also plays a major role depending on the Lewis number of reactants. Detailed mechanistic understanding of these effects is of fundamental interest in combustion studies.

Dynamics of unstable flames have been the subject of many numerical investigations, from which considerable phenomenological insight has been obtained [5–12]. Nevertheless, only a few studies have reported on the role of chemical reactions in flame instability (e.g., [6,10,12]). In principle, reaction kinetics plays a minimal role in hydrodynamic instability and therefore the increment of flame velocity can be interpreted as a consequence of increased flame surface area. The diffusive-thermal effect influences the gas composition supplied to the flames and induces spatial variations of the reaction rate [7], which can further destabilizes the flame when the Lewis number is sufficiently low. Although this variation

results in a further change in the flame velocity, the role of chemical processes on the acceleration mechanisms remains uncertain.

The present study aims to provide further mechanistic insight into the chemistry involved in the diffusive-thermal effect. Numerical simulations were performed for two-dimensional hydrogen–air flames using a detailed kinetic model. The results were analyzed from the viewpoint of chemical structure to uncover underlying chemical mechanism responsible for the instability.

2. Method

Free propagation of the two-dimensional premix hydrogen–air flame was simulated with the Navier–Stokes equations on a rectangular domain with a fixed length of $-L/2 \leq x \leq L/2$ and a fixed height of $0 \leq y \leq h$, where $L = 10.0$ cm and $h = 0.4$ cm. Detailed descriptions on the computational method with relevant Refs. [13–20] are given in the supplemental material. The premixed hydrogen–air mixtures at equivalence ratios of $\phi = 0.5$ and 1.0 entered the domain from the left ($x < 0$) boundary at a uniform unburned gas temperature ($T_u = 300$ K), pressure ($p = 1$ atm), and velocity (laminar flame velocity, S_L). The boundary for the outflow is the non-reflecting condition. Periodic boundary conditions were imposed in the y-direction. As an initial condition, a steady planar flame structure was placed on the domain and perturbed by a sinusoidal disturbance with amplitude of 0.004 cm and wavelength equal to the domain height. As a reference, one-dimensional (or planar) laminar flame speed, structure, and flame thickness ($\delta_T \equiv (T_b - T_u)/(dT/dx)_{max}$ where T_b is the adiabatic flame

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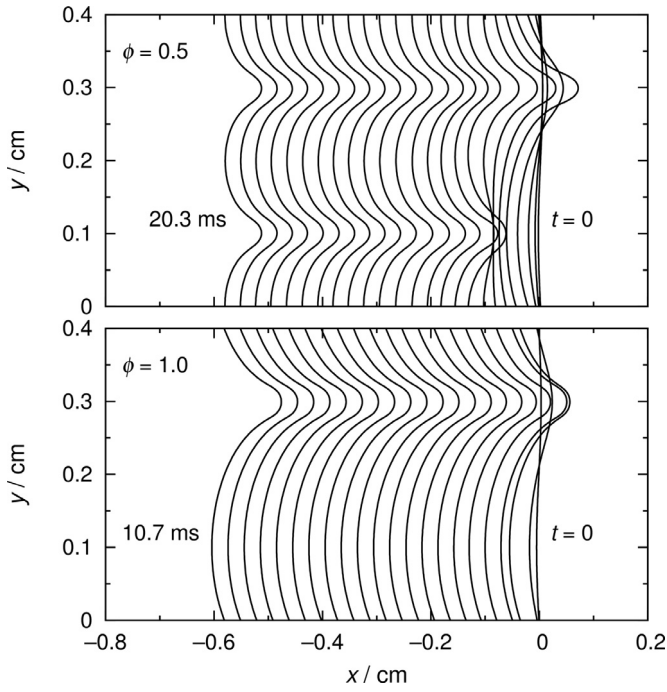*E-mail addresses:* a.matsugi@aist.go.jp (A. Matsugi), htera@eng.hokudai.ac.jp (H. Terashima).

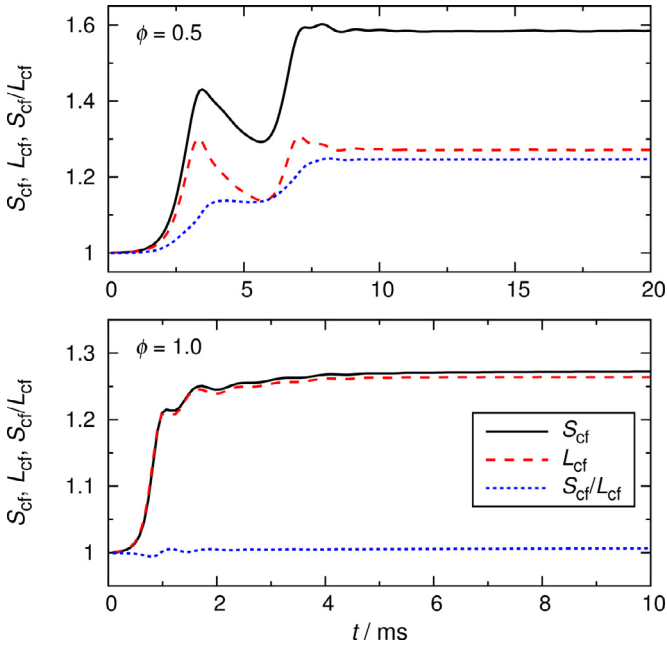**Fig. 1.** Time evolution of the flame front of the hydrogen–air flames at $\phi = 0.5$ and 1.0.

Fig. 2. Time history of the normalized burning velocity (S_{cf}), normalized flame front length (L_{cf}), and their ratio (S_{cf}/L_{cf}) of the hydrogen–air flames at $\phi = 0.5$ and 1.0.

temperature and $(dT/dx)_{max}$ is the maximum temperature gradient) were also calculated using the Cantera program [19].

3. Results and discussion

Temporal evolution of the flame fronts is shown in Fig. 1. Here, the flame fronts are defined as isolines of temperature as $T = (T_u + T_b)/2$. The $\phi = 1.0$ flame quickly evolved to a single-cell structure that stably propagated upstream, whereas formation of the secondary cell was observed for the $\phi = 0.5$ flame.

Figure 2 shows the time history of the normalized flame velocity (S_{cf}; calculated as the burning rate integrated over the whole computational domain and normalized by the planar flame value),

the normalized length of the flame front (L_{cf}; normalized by the planar flame value), and their ratio. As expected, the normalized velocity is approximately equal to the normalized flame front length for the $\phi = 1.0$ flame because the hydrodynamic effect dominates the cellular structure of this flame. On the other hand, the cell formation in the $\phi = 0.5$ flame also increases the velocity but the increment cannot solely be accounted for by elongation of the flame front, suggesting some changes of chemical reaction structure along the flame. Local spatial profiles of the temperature and major species fractions at the top of the convex (toward the unburned gas) front and at the bottom of the concave front in the $\phi = 0.5$ flame are shown in Figs. 3 and 4, respectively. The profiles are taken to be normal to the flame front and plotted along the relative distance x'. The profiles differ from those of the 1-D flame shown as solid lines. The diffusive-thermal effect preferably supplies H_2 to the convex side of the flame, which increases/decreases the reactivity of mixtures (and hence the temperature) at the crest/trough part of the flame.

The concentration of radicals build up in the preheat zone of the flame due to the well-known chain reaction mechanism:

$$H + O_2 = HO_2 \tag{R1}$$

$$H + O_2 = OH + O \tag{R2}$$

$$O + H_2 = OH + H \tag{R3}$$

$$OH + H_2 = H_2O + H \tag{R4}$$

In this region, the rate of each reaction is pseudo-first-order since there exist large excess amounts of H_2 and O_2 relative to the radicals. Thereby, the preferable diffusion of H_2 toward the convex part affects the buildup rate of the chain carriers. The chain reactions continue until the consumption of H_2 reaches a certain level. Then, the main heat release occurs by consumption of the chain carrier radicals, HO_2, and H_2O_2 mainly through the reactions:

$$H + O_2 = HO_2 \tag{R1}$$

$$O + H_2O = OH + OH \tag{R5}$$

$$OH + HO_2 = H_2O + O_2 \tag{R6}$$

$$OH + H_2O_2 = H_2O + HO_2 \tag{R7}$$

This region can be considered as the main reaction zone since most of the heat is released at this stage. Importantly, the heat release rates of these reactions are not affected by the concentration of H_2; the kinetics here is insensitive to the amount of H_2 supplied to the main reaction zone. Therefore, the primary consequence of the preferable diffusion of H_2 is the change in the buildup process of the reactive intermediates at the preheat zone.

The above consideration suggests that the reaction and heat release rates at the main reaction zone can be determined from the gas composition of the mixture provided from the preheat zone. As previously noted, the chain reactions proceed with pseudo-first-order at the preheat zone; hence, the composition should reflect the amount of H_2 (and O_2) consumed there. This indicates that the chemistry at the main reaction zone is similar to that of the 1-D premixed flame with different equivalence ratio. To validate this hypothesis, the corresponding *effective* equivalence ratio, ϕ_{eff}, was calculated on the basis of the peak concentration of H atoms ($[H]_{max}$); namely, the relationship between $[H]_{max}$ and the equivalence ratio was determined from the 1-D premixed flame calculations, which was used as a calibration curve to estimate ϕ_{eff} from