Morphology and Self-acceleration of Expanding Laminar Flames with Flame-front Cellular Instabilities

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Abstract

We report herein experimental observation and mechanistic interpretation of the evolution and self-acceleration of constant-pressure, spherically expanding $H_2/O_2/N_2$ flames, subjected to hydrodynamic and diffusional-thermal instabilities over a wide range of pressure, equivalence ratio and thermal expansion ratio. Results show the existence of three distinct stages of flame propagation affected by the development of the instability cells, namely smooth expansion, transition, and saturated states of cell development. The onset of the instabilities is primarily controlled by the diffusional-thermal instability, while characteristics of the subsequent transition to and maintenance of the saturated state is controlled by the hydrodynamic instability. The acceleration exponent for the fully developed saturated instability is found to be around 1.2 – 1.4, which is smaller than 1.5, the suggested value for self-turbulization.

Keywords: Expanding flame; Hydrodynamic instability; Diffusional-thermal instability; Self-similar propagation

1. Introduction

Propagating laminar flames are often subjected to various modes of flame-front instability arising from either intrinsic or external sources. Among the intrinsic instabilities, two of the most common modes are hydrodynamic and diffusional-thermal in nature, which induce cellular structure on the flame-front. The presence of these cells increases the flame surface area and as

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such could significantly augment the flame propagation speed as compared to that of the smooth flame. Furthermore, the extent of the augmentation could also continuously increase with evolution of the cells over an expanding flame, leading to the possibility of self-acceleration. Mechanistically, hydrodynamic instability sets in through sharp density change across the flame-front and characteristically occurs for either very large or very thin flames, while diffusional-thermal instability is controlled by the imbalance in the diffusivities of heat and the various species. The imbalance is usually characterized by the Lewis number, Le , defined as the ratio of the thermal diffusivity of the mixture to its controlling mass diffusivity, such that mixtures with $Le < 1$ favor the onset of the instability, while $Le > 1$ mixtures are stabilizing.

Because of their fundamental significance as well as practical importance as the primary modes of augmented flame propagation, especially within the high-pressure environments of IC engines for which the hydrodynamic instability can be substantially enhanced, there have been extensive studies on these two modes of instability. Among the experimental studies, both large-scale open flames [1–4] and confined flames in combustion vessels [5–9] have been employed. Theoretically, Sivashinsky [10–12], and Matalon and co-workers [13–16] have analyzed the critical conditions for the onset, cut-off limits, and growth rates of the instabilities using both linear and non-linear theories. Experimental investigation of the development of flame-front instability confronts considerable challenges because of the highly transient and multi-scale nature of the phenomena. For example, development of the large hydrodynamic cells in atmospheric flames requires large flame sizes, which not only requires experimentation in large open space, but the resulting large flame is also subjected to considerable buoyant distortion from spherical symmetry and as such renders it difficult to meaningfully define, say, the flame radius. Another potential difficulty, though not specifically addressed, is the need to identify the separate and coupled effects from the hydrodynamic and diffusional-thermal instabilities.

In the present study, we shall employ the well-vetted, dual-chamber, constant-pressure apparatus to conduct spark-ignited, expanding spherical flame experiments in enclosed environments of elevated pressure, which facilitate the development of hydrodynamic cells because of reduced flame thickness. In particular, we shall first suppress effects of diffusional-thermal instability by working with mixtures that are stoichiometric in composition and thereby are diffusionally neutral, and study the evolution dynamics of

flames subjected only to hydrodynamic instability, at elevated pressures. We shall then fold in the presence of diffusional-thermal instability by studying off-stoichiometric mixtures, and study its separate and coupled effects on the development of the instability cells. The specific issues of interest are the parametric boundaries for the onset of the instabilities, the possible existence, nature and extent of self-acceleration, and an assessment of the possible attainment of the state of self-turbulization.

We shall now first present the experimental and computational specifications of the present investigation, which is followed by the presentation and discussion of the various experimental components aiming to provide insight and answer to the above issues of interest.

2. Methodology

2.1. Experimental Details

Details of the constant-pressure, dual-chambered apparatus, in which a mixture of fuel, oxidizer and inert is filled at required concentration ratio and pressure, and then centrally ignited producing a spherically expanding flames, are described in Ref. [17, 18]. Mixtures of hydrogen, oxygen and nitrogen were used. The experimental conditions, listed in Table 1, are so chosen that they cover a wide range of the parametric space, including the equivalence ratio ϕ (various H_2/O_2 ratios), adiabatic flame temperature T_f (various N_2/O_2 ratios), and pressure P . The evolution of the flame-front morphology was recorded by a high-speed Schlieren image system, with the highest sampling rate of $15kHz$ at $600\text{pix} \times 600\text{pix}$, yielding the instantaneous flame radius $R(t)$, defined as $R = \sqrt{A/\pi}$, where A is the area of the 2D-projection of the flame, and the subsequent instantaneous flame speed, based on the burned mixture, as $S_b(t) = dR/dt$. Details of the image processing are in Ref. [8]. To avoid the effects of ignition and chamber confinement, data processing was limited to flames with radii between $5mm$ and $20mm$, the latter being 30% of the chamber size.

2.2. Computational Specifications

The fundamental parameters, namely the (downstream) laminar flame speed ($S_{b,0}$), flame thickness (δ), adiabatic flame temperature (T_f), and thermal expansion ratio (σ) for the planar laminar flame at the experimental conditions were calculated by simulating the 1D planar flame using the Chemkin-Pro Software and the H_2/O_2 kinetics model of Burke et al [19].

The Lewis number is given by $Le = Le_{H_2} < 1$, ≈ 1 , and $= Le_{O_2} > 1$ for lean, stoichiometric, and rich mixtures, respectively. Flame thickness (δ) was calculated based on temperature profile using the relationship, $\delta = (T_f - T_0)/(dT/dx)_{max}$, where T_0 is the initial temperature and $(dT/dx)_{max}$ the maximum temperature gradient. The laminar flame speed ($S_{b,0}$) and flame thickness (δ) values for the experimental conditions are listed in Appendix.

ϕ	$T_f(K)$	$P(atm)$	X_{H_2}	X_{O_2}	X_{N_2}	$\sigma(\pm 0.03)$	Le_{eff}
0.6	1600	5	0.1667	0.1389	0.6944	4.92	0.4
0.6	1700	1 to 5	0.1818	0.1515	0.6667	5.18	0.4
0.6	1800	1 to 5	0.1974	0.1645	0.6382	5.47	0.4
0.6	1900	5	0.2105	0.1754	0.6140	5.71	0.4
1.0	1600	5	0.1681	0.0840	0.7479	4.92	≈ 1
1.0	1700	1 to 10	0.1818	0.0909	0.7273	5.18	≈ 1
1.0	1800	1 to 10	0.1970	0.0985	0.7044	5.47	≈ 1
1.0	1900	5	0.2094	0.1047	0.6859	5.71	≈ 1
1.5	1600	5	0.2479	0.0826	0.6694	4.92	1.7
1.5	1700	1 to 10	0.2691	0.0897	0.6413	5.18	1.7
1.5	1800	1 to 10	0.2899	0.0966	0.6135	5.47	1.7
1.5	1900	5	0.3093	0.1031	0.5876	5.71	1.7

Table 1: List of experimental conditions. ϕ : equivalence ratio; T_f : adiabatic flame temperature; P : pressure; X_{H_2} , X_{O_2} , X_{N_2} : mole fractions of H_2 , O_2 and N_2 respectively; σ : thermal expansion ratio; Le_{eff} : effective Lewis number of the mixture. Initial temperature is $298(\pm 2)K$.

3. Flame-front Evolution and Self-acceleration

3.1. Evolution Dynamics of Hydrodynamic Instability

We first study the evolution dynamics for flames affected mainly by hydrodynamic instability, accomplished by suppressing the diffusional-thermal instability by working with stoichiometric mixtures ($T_f = 1800K$) whose Le is close to unity. The intensity of the hydrodynamic instability is varied by changing the system pressure, and thereby the flame thickness. The measured downstream flame speeds of the burned mixture, S_b , at different radii and pressures are plotted in Fig. 1(a) and clearly show substantial increase in the flame speed as the flame expands. Figure 1(I-IV) also shows that, for

the same flame radius, flames at higher pressures are highly wrinkled and hence more cellularly unstable.

Figure 1(a) further shows that at the lower pressure of $1atm$, S_b barely exhibits any acceleration, while at $2.5atm$, S_b starts to accelerate significantly after reaching a certain radius. At the higher pressure of $5atm$, the flame exhibits three stages: starting with a relative constant flame speed, followed by large acceleration, and then mild acceleration. Furthermore, at the highest pressure of $10atm$, only the latter two stages are observed as upon ignition the flame immediately starts propagation with large acceleration and then settles to mild acceleration.

To conduct a rational interpretation of the data shown in Fig. 1(a), we first recognize that there are three factors affecting the burning rate of the flame with increasing pressure, namely the increase in the density and thereby the amount of mass needed to be processed by the propagating flame, the increase in the chemical reactivity with increasing concentration and hence pressure, and the increase in the surface area due to the flame-front wrinkling. To systematically investigate the influence due to these three factors, and eventually isolate the influence due to the instability effect, we first note that since the fundamental parameter indicating the burning rate of a flame is the density-weighted flame speed instead of the flame speed itself, Fig. 1(b) plots this quantity, the mass burning rate ($\rho_b S_b$), which then clearly shows that the flame indeed has larger mass burning rate at higher pressures, as should be. This density-weighting effect has been amply demonstrated for laminar flames, with the understanding that the laminar burning rate, namely the density-weighted flame speed, is the proper rate parameter indicating the efficiency with which a flame consumes the reactive mixture.

To (largely) eliminate the influence of increased chemical reactivity, we plot in Fig. 1(c) the normalized flame speed $\bar{S}_b = S_b/S_{b,0}$, where $S_{b,0}$ is the unstretched, laminar flame speed, so that effects of chemical reactivity can be considered to be mostly canceled out as the same chemical reactivity affects both the stretched and unstretched flames; note that the density weighting effect is also cancelled out for both flame speeds. Consequently $\bar{S}_b$ should primarily indicate the increase of the flame burning rate due to the development of the hydrodynamic cells and hence the increased flame surface area. Furthermore, in Fig. 1(c), $\bar{S}_b$ is plotted versus the scaled flame radius $Pe = R/\delta$, which measures the extent of the hydrodynamic destabilizing force that increases with decreasing flame thickness, δ . Figure 1(c) then clearly collates the three-stage evolution behavior for the different pressures, indicated by

(a)

(b)

(c)

Figure 1: Schlieren images and flame speeds (dR/dt) for $H_2/O_2/N_2$ flames at stoichiometry ($\phi = 1$), $1800K$, and different pressures. Top row are images with same radius ($1.5cm$) at different pressures: (I) $1atm$, (II) $2.5atm$, (III) $5atm$ and (IV) $10atm$, respectively. 2nd and 3rd rows are (a) dimensional flame speed vs flame radius, (b) dimensional mass-burning rate vs flame radius, and (c) nondimensional flame speed vs normalized flame radius, $Pe = (R/\delta)$.

the solid line. Specifically, when the flame is small ($Pe < 100$ in Fig. 1(c)), it is devoid of any instabilities as depicted by the smooth flame-front shown in Fig. 1(I), and the propagation is affected by flame stretch alone, instead of cellular instabilities. This stage is hereafter referred to as the *smooth expansion stage*. As the flame grows large enough such that Pe exceeds a critical value Pe_c ($Pe_c \approx 100$ in Fig. 1(c)), cellular instability starts to appear over the flame-front [7, 13, 15], causing the flame to accelerate rapidly due to the excess flame area generated by the cells. Figure 1(II and III) shows the flame development during this stage, hereafter referred to as the *transition stage*. During this stage, the first large-scale cells that appear on the flame-front break down to smaller cells leading to a cascading process to produce small-scale cells. Consequently, as the flame grows, the largest cell size increases while the continuous cascading process extends the limit of the smallest cell sizes as well. However, after a certain limit, the smallest cell size reaches the hydrodynamic cut-off limit, and leads to saturation of the cascading process. As the flame is still radially expanding, the largest characteristics length (flame radius) grows continuously, hence increases the number of cells and the flame surface area. Saturation of the smallest cells, thus, reduces the acceleration rate after the flame grows beyond a critical Peclet Number, Pe_s ($Pe_s \approx 150$ in Fig. 1(c)), hereafter referred to as the *saturated stage*. The typical flame-front is shown in Fig. 1(IV).

3.2. Coupled Hydrodynamic and Diffusional-Thermal Instabilities

For $Le < 1$ mixtures, not only the diffusional-thermal instability is favored, it also extends the spectrum of the cell sizes as its characteristic length is of the order of the flame thickness, which is smaller than that of the hydrodynamic cells. On the other hand, for $Le > 1$ case, the flame-front is diffusional-thermally stable, and as such could actually damp the development of the hydrodynamic cells.

To investigate the influence of the additional presence of the diffusional-thermal cells, we compare the results of $H_2/O_2/N_2$ flames for lean ($\phi = 0.6$; $Le < 1$), stoichiometric ($\phi = 1.0$; $Le \approx 1$) and rich ($\phi = 1.5$; $Le > 1$) mixtures, again with $T_f = 1800K$ (Fig. 2a). It is seen that, as expected from the above discussion, the normalized flame speed indeed increases for the lean mixtures and decreases for the rich mixtures, implying the possible increase in the extent of wrinkling by the diffusional-thermal cells for the former, and the corresponding decrease for the latter. It is also of interest

to note the continuous nature of the correlation for the rich flames, indicating the preservation of the hydrodynamic cells. This is to be contrasted to the disjointed nature of the lean (and stoichiometric) flames, indicating the participation of the diffusional-thermal cells together with the hydrodynamic cells in the cellular spectrum, and as such the inadequacy of the $\overline{S_b} - vs - Pe$ plot based on consideration of the hydrodynamic instability alone.

Figure 2: Effects of equivalence ratio. Nondimensional flame speeds $\overline{S_b}$ for different equivalence ratios and pressures at $T_f = 1800K$. Grayscale represents various equivalence ratios. Shapes indicate different pressures: 1atm - circle; 2.5atm - square; 5atm - triangle; 10atm - diamond. Legends are in the form of $(P \text{ in atm}, \phi)$. Only 1 in every 5 data points is shown here.

3.3. Effect of Thermal Expansion (σ)

In addition to being affected by the flame thickness through pressure, hydrodynamic instability is also strongly influenced by the thermal expansion ratio, σ , across the flame, which can be represented by the adiabatic flame temperature, T_f . Figures 3(a), (b) and (c) show the results of $\overline{S_b}$ of $H_2/O_2/N_2$ flames with different T_f for lean, stoichiometric and rich mixtures respectively. Figure 3(a) then shows significant influence of thermal expansion for lean mixtures. At the smooth *expansion stage*, for $Pe < Pe_c$, there is relatively small difference between various T_f . This is because in the absence of instability, for mixtures with the same Le , σ (or T_f) primarily affects $S_{b,0}$, which is accounted for in the flame speed scaling. We next note that the two

critical Peclet numbers (Pe_c and Pe_s) decrease with increasing T_f (or σ). Mechanistically, while the propensity of hydrodynamics instability is weaker for flames with lower T_f (or σ), the propensities of the diffusional-thermal instability for various T_f (or σ) are similar because they have the same Le . Therefore, relatively speaking, diffusional-thermal instability dominates hydrodynamic instability for flames with lower T_f (or σ), and consequently extends the spectrum of the cell size towards the smaller values.

For stoichiometric ($\phi = 1.0$) mixtures the role of the thermal expansion is weaker, mostly because preferential diffusion plays a neutral role for $Le \approx 1$ mixtures. Therefore, compared to the lean case (Fig. 3(a)), the difference in $\overline{S_b}$ between various T_f (or σ) for stoichiometric mixtures (Fig. 3(b)) is smaller. However, since the actual Le is slightly smaller than 1, indicated by the negative slope at the *smooth expansion stage*, the flames speeds (in the *transition* and *saturated* stages) are higher for the lower T_f (or σ), bearing effects of sub-unity Le .

For rich mixtures ($\phi = 1.5$, $Le > 1$), Fig. 3(c) shows that $\overline{S_b}$ collapse nicely for the four values of σ studied; confirming again that in the absence of diffusional-thermal instabilities, the present $\overline{S_b} - vs - Pe$ scaling properly describes the dynamics of hydrodynamic instability, at least within our range of data.

4. Critical Peclet numbers (Pe_c and Pe_s)

As discussed earlier, the change in the morphology and propagation mode of the expanding flame is defined by two critical Peclet numbers, namely the *onset Peclet number* (Pe_c) designates the minimum flame radius for the cellular structure to appear, below which the flame-front is smooth [7, 13, 15] and the *saturation Peclet number* (Pe_s) which designates the flame radius when the cellular structure saturates the flame-front, leading to a distinct slowdown in acceleration from that in the *transition stage*.

Figures 4(a) and (b) show the variation in Pe_c and Pe_s at various σ (or T_f), ϕ (or Le) and pressures. It is seen that Le is the most influential parameter that affects both Pe_c and Pe_s . Furthermore, the Pe_c for the lean cases ($Le \approx 0.4$) is about 1/3 of the stoichiometric cases ($Le \approx 1$), which in turn is almost 1/3 of the rich cases ($Le \approx 1.7$) for a given σ . This is because with increasing Le , the flame-front tends to stabilize from the diffusional-thermal instability, and therefore requires larger flame sizes for inception and for saturation of the cell growth, resulting in larger Pe_c

Figure 3: Effects of adiabatic flame temperature. Nondimensional flame speeds $\overline{S_b}$ for $H_2/O_2/N_2$ flames of: (a) lean ($\phi = 0.6$), (b) stoichiometric ($\phi = 1.0$), (c) rich ($\phi = 1.5$) mixtures at various pressures and different adiabatic flame temperatures. For clarity, 5atm data at four T_f for stoichiometric mixture are shown in the inset of (b). Grayscales represent various temperatures. Shapes indicate different pressures: 1atm - circle; 2.5atm - square; 5atm - triangle; 10atm - diamond. Legends are in the form of (P in atm, T_f in K). Only 1 in every 5 data points is shown

and Pe_s . Figure 3 also shows that the effect of hydrodynamic instability is stronger with increasing σ , indicating that the cascading process of the flame cells continues for longer period, hence delaying the transition to the *saturated stage*.

Figure 4: Critical Peclet numbers for (a) onset (Pe_c) and (b) saturation (Pe_s) for $H_2/O_2/N_2$ flames of various equivalence ratios (ϕ) at different thermal expansion ratios (σ) and pressures. Grayscales indicate different ϕ : lean ($\phi = 0.6$): white; stoichiometric ($\phi = 1.0$): gray; rich ($\phi = 1.5$): black. Shapes indicate different pressures: 2.5 atm - square; 5 atm - triangle; 10 atm - diamond. Legends are in the form of (P in atm, ϕ). Uncertainty is less than 15%.

5. Acceleration Exponents

An intriguing aspect of the flame acceleration phenomena is to determine the possible values of the acceleration exponent, α , defined through $R \sim t^\alpha$ and the fractal excess, d , where $d = 1 - \alpha^{-1}$, so as to assess if these flames can reach a “self-turbulizing” condition [20, 21]. Specifically, Sreenivasan and co-workers [22–24] theoretically and experimentally showed that for fully-developed Kolmogorov turbulence the fractal dimension, defined as by $2 + d$, takes a value of $7/3$, which corresponds to $\alpha = 1.5$. Consequently much experimental and computational effort has been expended towards assessing if the flame acceleration exponent can indeed reach the critical value of 1.5, which would suggest a fractal dimension similar to that of Kolmogorov turbulence. A comprehensive assessment [8] has however shown that there has been considerable inappropriateness in the comparison in the literature, and there has not been any conclusive evidence that the acceleration exponent can indeed reach this value.

Using the present data we have evaluated the acceleration exponents for the *transition stage* (α_t) and the *saturated stage* (α_s), shown respectively in Figs. 5(a) and (b). It is seen that both α_t and α_s are insensitive to Le ,

indicating that rapid and significant flame *acceleration* is dominated by the growth of the hydrodynamic cells, even though the actual values of the flame speed can be slightly elevated by the diffusional-thermal cells during the initial period of transition. This also implies that the fractal excess (d) and fractal dimension ($2 + d$) for the saturation stage is independent of pressure, Le and the thermal expansion ratio, at least within the present experimental conditions. We finally note that the values of α_s lie between $1.2 - 1.4$, which is smaller than 1.5 , the value suggested as the acceleration exponent for self-turbulization.

Figure 5: Acceleration exponents for (a) *transition stage* (α_t) and (b) *saturated stage* (α_s) for $H_2/O_2/N_2$ flames of various equivalence ratios (ϕ) at different thermal expansion ratios (σ) and pressures. Colors indicate different ϕ : lean ($\phi = 0.6$): white; stoichiometric ($\phi = 1.0$): gray; rich ($\phi = 1.5$): black. Shapes indicate different pressures: 2.5 atm - square; 5 atm - triangle; 10 atm - diamond. Legends are in form of (P in atm, ϕ).

6. Summary

In summary, we report herein a comprehensive study on the effects of flame-front instabilities on flame propagation, covering a wide range of pressure, Lewis number and thermal expansion ratio, using $H_2/O_2/N_2$ mixtures. It is first demonstrated that the use of normalized flame speed and normalized flame radius (*i.e.*, Peclet number) clearly identifies three stages of flame propagation, namely *smooth expansion*, *transition* and *saturated states* of cell development. The smooth propagation of the flame continues until the *onset Peclet number*, Pe_c , at which the instability is triggered as cells starts to

grow. At this stage the flame accelerates rapidly until the smallest cell size reaches the cut-off limit inhibiting further reduction in the cell size. Beyond this *saturation Peclet number*, Pe_s , the flame still accelerates but with reduced rate. The activation of the diffusional-thermal cells for the $Le < 1$, lean mixtures expands the spectrum of the hydrodynamic cells at the small wavelength, hence facilitating the two transitions (Pe_c and Pe_s), while the opposite holds for $Le > 1$, rich mixtures for which differential diffusion has a stabilizing effect on the hydrodynamic cells, hence inhibiting the transition. At the *saturated stage*, the acceleration exponent is insensitive to Le , pressure or thermal expansion ratio and its value is around 1.2 to 1.4, which is smaller than the suggested exponent of 1.5 for self-turbulization.

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Appendix A. Flame Speed and Flame Thickness

Table A.1: The (downstream) laminar flame speed ($S_{b,0}$ in cm/s) and flame thickness (δ in cm) for the experimental conditions

		$P = 1atm$		$P = 2.5atm$		$P = 5atm$		$P = 10atm$	
$T_f(K)$	ϕ	$S_{b,0}$	δ	$S_{b,0}$	δ	$S_{b,0}$	δ	$S_{b,0}$	δ
1600	0.6	-	-	-	-	88.14	0.0145	-	-
1700	0.6	291.98	0.0428	218.49	0.0176	155.47	0.0103		
1800	0.6	423.86	0.0379	332.47	0.0146	249.28	0.0080		
1900	0.6	-	-	-	-	350.34	0.0068	-	-
1600	1.0	-	-	-	-	86.44	0.0166	-	-
1700	1.0	278.83	0.0518	207.00	0.0210	142.01	0.0124	82.60	0.0089
1800	1.0	389.87	0.0467	307.52	0.0178	224.98	0.0098	143.41	0.0063
1900	1.0	-	-	-	-	308.95	0.0084	-	-
1600	1.5	-	-	-	-	306.92	0.0073	-	-
1700	1.5	645.75	0.0366	565.77	0.0125	457.38	0.0062	323.28	0.0035
1800	1.5	821.09	0.0353	749.50	0.0117	634.50	0.0055	477.97	0.0029
1900	1.5	-	-	-	-	825.14	0.00513	-	-

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