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An experimental investigation on self-acceleration of cellular spherical flames

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Abstract

The cells that continuously develop over the flame surface of an expanding spherical flame increase its area and thereby the global propagation rate, resulting in the possibility of self-acceleration. The present study examines whether this self-acceleration could be self-similar, and, if so, whether it could also be self-turbulizing. Extensive experiments at elevated pressures and thereby reduced laminar flame thicknesses and enhanced propensity to exhibit Darrieus-Landau instability were conducted for hydrogen/air mixtures over an extensive range of equivalence ratios. The results demonstrate the strong possibility of self-similar flame acceleration, weak influence of the system pressure and diffusional-thermal instability, and a corresponding moderate spread in the power-law acceleration exponent.

Keywords: Self-acceleration; Expanding spherical flame; Hydrodynamic (Darrieus-Landau) instability; Cellular flames; Self-similar propagation

1. Introduction

Propagation of laminar premixed flames is subjected to the excitation of flame front diffusional-thermal, hydrodynamic, and buoyancy induced instabilities, which are respectively caused by the non-equidiffusive nature of the diffusive scalars of species concentration and heat, the density jump across the flame, and body force in the presence of density gradient. The presence of cells over the flame surface increases its surface area and consequently also the global propagation speed of the flame. Furthermore, since new cells continuously evolve as the flame propagates, it is reasonable to expect that the flame speed will also continuously increase, leading to the possibility of self-acceleration. The phenomenon is of particular interest for the propagation of the globally spherical flame because of the well-defined, yet continuously increasing flame radius, which yields a characteristic dimension and the dynamic parameters against which excitation of the various instability modes evolve. Specifically, during the initial stage of flame propagation when the flame radius is small, the small-scale diffusional-thermal instability is expected to dominate. As the flame becomes larger and its thickness is small compared to its radius, the hydrodynamic, Darrieus-Landau (DL) instability sets in, progressively dominating

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the flame morphology and the accelerative dynamics.

This self-acceleration can be quantified, and an assessment of the propagation mode made, by determining the history of the average flame radius, R_{av}, through a power-law expression, $R_{av} \sim t^{\alpha}$, where t is the time. There has been considerable interest in the specific values that α can assume. That is, if α is a constant, then the flame acceleration is either self-similar in t, if it holds for all t, or locally similar if it holds only for a specific period of t. Furthermore, it has been suggested [1–3] that if α is not an integer, then the wrinkled flame front can be considered as a fractal surface with the total surface area of the front given by $A_f \sim R_{av}^{2+d}$, where the constant $d = 1 - \alpha^{-1}$ is the fractal excess. In the limit of an infinitesimally thin flame, diffusional-thermal effects are suppressed and the local flame speed over the wrinkled flame surface is that of the laminar flame speed, S_L, and is as such a constant. The fractal hypothesis can then be related to the self-similar propagation of the flame front through,

$$4\pi R_{av}^2 \frac{dR_{av}}{dt} \approx S_L A_f, \qquad \frac{dR_{av}}{dt} \sim R_{av}^d$$

Finally, for fully-developed Kolmogorov turbulence it has been suggested by Mandelbrot [4,5] that the iso-surfaces (e.g., iso-temperature) also have a fractal character, and the fractal dimension $2 + d$ has been theoretically estimated by Sreenivasan et al. [6] to be 7/3 based on Reynolds number similarity. Experimental values by Sreenivasan and Meneveau [7,8] for a variety of turbulent flows fall in the range of 2.35 ± 0.05, in close agreement with the theoretical prediction of 7/3. Consequently, if α assumes the value of 3/2 for the case of expanding laminar flames under hydrodynamic instability, then $2 + d = 7/3$, and the flame has possibly undergone self-turbulization and thereby assumed the character of turbulence.

Considerable experimental and computational studies on laminar flames have been conducted, aiming to demonstrate the possible attainment of $\alpha = 3/2$ and hence self-turbulization, although with inconclusive results. In fact, in the interest of demonstrating such an attainment, interpretations of the results have been conducted rather loosely, causing considerable confusion in the rigorous understanding of the fundamental phenomena.

In view of the above considerations, we shall perform a well-controlled experimental investigation, aiming to provide reliable data to answer the following three increasingly demanding claims on α, namely:

- Does the flame self-accelerate, i.e., is $\alpha > 1$?
- Is α a constant such that the propagation is self-similar?
- Is α approximately equal to 3/2 such that the wrinkled laminar flame has undergone self-turbulization?

Before doing so, it is necessary to first present a brief assessment of the state of the determination of α in order to demonstrate the various subtleties involved in conducting an unambiguous determination. A more detailed discussion on this issue is given in [9].

2. Review of previous works

2.1. Experimental studies

Lind and Whitson [10] conducted large-scale explosion experiments in quiescent condition at atmospheric pressure with various mixtures including H_2/air, H_2/O_2, C_3H_8/air, CH_4/air, C_4H_6/air and C_2H_2/air. These experimental data were later analyzed by Gostintsev et al. [11] using $\alpha = 3/2$ for all mixtures studied and concluded that this value is consistent with the data. However, the data analysis was not clearly described, and more significantly, the original reported value of α was lowered to $1.25 \sim 1.5$ in two later publications [12,13], with the revised assessment that the value of 1.5 is only attained in limiting cases. Most of the subsequent studies by others have either justified or focused their investigations on the presumed validity of $\alpha = 1.5$.

Bradley et al. [14,15] experimented with large-scale flames at atmospheric pressure, and allowed the outwardly propagating flames to grow to radii as large as 3 m so that hydrodynamic cells could be developed. A constant α of 1.5 was again used in their data analysis. Furthermore, due to strong buoyancy with typical Froude numbers, V^2/gR, being around 2, the experimental flame was largely hemispherical, as shown in [14]. This severe lack of spherical symmetry fundamentally complicates the data interpretation and analysis.

Haq [16] conducted experiments for atmospheric CH_4/air flames in a combustion vessel. It was shown that the data obtained at small radii could be blended with those of Lind and Whitson [10] at large radii, with good match. Furthermore, the results showed that α is 1.24 for the horizontal radii and 1.32 for the vertical radii; the difference could be due to buoyancy effects. The primary concern with this series of experiments is whether mature hydrodynamic cells were developed for flames of such small dimension at atmospheric pressure, at which the flame thickness would require cells of large dimension, recognizing that the flame of Bradley et al. [14] required 3 m to develop.

Kwon et al. [17] experimented with $H_2/O_2/N_2$ flames, using a constant and high-pressure dual-chamber vessel at a pressure of 15 atm. It is noted that the elevated pressure reduces the flame thickness and thereby the hydrodynamic cell size, hence allowing the development of hydrodynamic cells for the small, laboratory-scale flames. The reported values of α are 1.26, 1.36 and 1.23 for $\phi = 0.50$, 1.00 and 1.50, respectively.

2.2. Numerical studies

A number of numerical investigations [2,18–28] have been conducted with focus on the issue of self-acceleration of expanding wrinkled flames. Due to the high computational cost, almost all studies were performed for 2-D cylindrical flames. Because the power-law exponent α for the 2D and 3D flames are necessarily different, as noted by Blinnikov and Sasorov [2], caution is needed in drawing direct comparison for values of α obtained from 2D and 3D flames. Furthermore, most simulations were also conducted using simplified models, such as the Sivashinsky equation or Frankel equation. Finally, there were studies that not only claimed $\alpha_{2D} = 1.5$ from calculations obtained by using 2D cylindrical flames, but the relevant slopes were also determined from results exhibiting various degrees of nonlinearity [18,20,21]. Since the goal of the present investigation is to experimentally assess the possible values of α, and in view of space limitation, the reader is referred to [9] for a more detailed review of these computational studies, which show not only considerable differences in the reported value of α but also the lack of any unambiguous evidence of the attainment of $\alpha = 1.5$.

2.3. Fitting formulas

In addition to disagreement in the values of α, there is also inconsistency among different studies in the extraction of α. The early studies of Gostintsev et al. [11] and Bradley et al. [14] simply assumed $\alpha = 1.5$ and applied the following self-similar law:

$$R = R_0 + At^{3/2}$$

where R is the flame radius, t the time after ignition, and R_0 and A are the parameters for best linear fitting. Obviously, such a fitting automatically assigns $\alpha = 1.5$, although an examination of the reported data shows that a plot of R–$t^{3/2}$ is not exactly linear.

To evaluate α directly from data, other experimental and numerical studies [17–19] treated all R, A and α as fitting parameters:

$$R = R_0 + At^{\alpha}$$

where R is the flame radius and t is either the time after ignition in experiments or the time after calculation starts in the simulation. This formula allows determination of the acceleration exponent, α, directly from the data. However, because of the definition of t, the fitting result will automatically depend on the instant of ignition, or the simulation starting point. For example, as shown in Fig. 1, when a set of experimental data is artificially shifted in time (which in real experiments can be caused by the different ignition conditions), different values of α are obtained.

Other studies [20,21] set R_0 to be the visual onset of the transition from a smooth to a wrinkled flame, and consider the following formula:

$$R = R_0 + A(t - t_0)^{\alpha}$$

where R_0 and t_0 are the radius and time of the onset of wrinkling. This method avoids the influence of the ignition condition; however, it introduces two other concerns. First, the determination of the onset of cellularity from experimental or numerical flame images can be quite subjective. The fitting result are very sensitive with respect to the values of R_0 and t_0, especially for the range of data points that immediately follows the onset, because both R–R_0 and t–t_0 are close to zero. Second, since there is possibly a transitional period after the onset of cellularity but before the acceleration reaches a saturated self-similar regime [19,22,23], using this formula still depends on the transitional period, as shown herein.

Some numerical studies [22–24] fitted the calculated propagation velocity, $U = \mathrm{d}R/\mathrm{d}t$, by a power-law:

$$U = A\alpha^{-1}t^{\alpha-1}$$

while others [16] ignored the constant *R0* or *t0* and directly considered the power-law:

$$R = At^{\alpha}$$

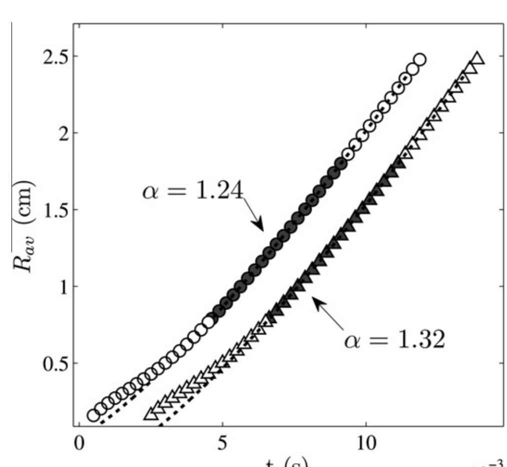

Fig. 1. A set of experimental data from the present study; Circles: original data, Triangles: same set of data after an artificial time shift $\Delta t = 2$ ms. Filled markers indicate data used for fitting. The dot lines are fitting curves.

However, the influence of the initial condition still exists as t is still defined as the time after ignition in experiments or the time after the calculation starts in simulations.

3. Present experiment and data analysis

3.1. Experimental considerations and specifications

The key consideration for the present experiment is to reduce the flame thickness so that hydrodynamic cells can be readily excited and rapidly grow to maturity in accordance with the Landau limit of flame-sheet propagation [29]. This goal was achieved by employing two unique features. First, experiments were conducted using a dual-chamber design of a closed combustion vessel [30] that allows observation of constant-pressure flame propagation with initial pressures as high as 60 atm. The high-pressure flame propagation therefore substantially reduces the flame thickness. The reader is referred to [30] for details of the apparatus design and the experimental procedure.

Second, the flame thickness was further reduced by using hydrogen as the test fuel because of its high flame speed. In particular, lean hydrogen flames were observed to almost immediately exhibit cellular instabilities after initiation of propagation because of the simultaneous excitation of the diffusional-thermal cellular instability [31,32]. Consequently, they are particularly suitable for the present investigation, with their cellular propagation mode taking place over 10^2–10^3 flame thicknesses in our investigation. This provides ample time and space for the development of the cell and exploration of the possible existence of the self-similar propagation mode. The flame propagation was imaged with schlieren photography and recorded using a high-speed digital camera at 2–25 k frames per second, depending on the propagation speed of the flame.

3.2. Data processing

Figure 2 shows three typical sequences of the images of propagating hydrogen/air flames. Specifically, it is seen that, at a pressure of 5 atm, the flame surface is wrinkled for lean but not rich mixtures, hence demonstrating the stabilizing effect of the diffusional-thermal instability on rich hydrogen/air mixtures whose Le is greater than unity. Flames of these rich mixtures, however, also become wrinkled at the higher pressure of 25 atm, demonstrating the manifestation of hydrodynamic instability as the flame becomes progressively thinner with increasing pressure.

Accurate tracking of the flame front from schlieren cine images is crucial, especially for a subject like this current study with high sensitivity. Due to the fact that wrinkled flame fronts contain many troughs and cusps, only an averaged flame radius of the entire flame front has statistical meaning. In this study, the Canny Edge Detection method was applied on the experimental flame images and the flame radius, $R(\theta, t)$, was determined as a function of time, *t, and angle,* θ. The tracked lines are plotted in the third row of each column in Fig. 2. From the tracking results, the average flame radius of a wrinkled flame front is defined as:

$$R_{av}(t) = \frac{1}{\Theta}\int_0^{\Theta} R(\theta, t)d\theta$$

where Θ is the angle that the selected portion of flamefront encompasses. Θ is slightly less than 2π for cases where the disturbances by the ignition wires become visible, for example, the flame shown in the third column of Fig. 2.

Realizing the inconsistency in the fitting methods in previous studies; we note that the fundamental hypothesis on the self-similar propagation is applied to the velocity instead of the radius [1,2]:

$$U_{av} = \frac{dR_{av}}{dt} = BR_{av}^{d}$$

where $B = \alpha A^{1/\alpha}$ is a constant that only depends on the properties of the mixture. This relation directly comes from the fractal description of the flame propagation. Therefore, this formula is employed in the present study, treating B and d as parameters for optimal fitting. As a result, the current fitting method removes the influence of initial conditions through the removal of t in the fitting equation; and the issues in the determination of R_0 or t_0.

The only concern remaining is then the accuracy in the numerical differentiation of R_{av} to obtain U_{av} For any discrete data, a straightforward numerical differentiation, such as the central differencing, will introduce additional random errors. For example, in a number of previous studies [16,20,22,28], although the R–t plot appears to be smooth, the U–t plot is often quite scattered after the differentiation. This is either because the time step in the differentiation is too small to acquire a statistically meaningful quantity, or the poor performance of the flame front tracking technique. To suppress the random error introduced by numerical differentiation, smoothing techniques are frequently employed. However, these techniques, such as the spline-fit or moving average, use a much longer range of the R–t data than central differencing and can thus potentially produce artificial effects, especially considering that the influences of ignition and wall confinement are to be eliminated [33,34]. With these considerations in mind, we did not use any curve

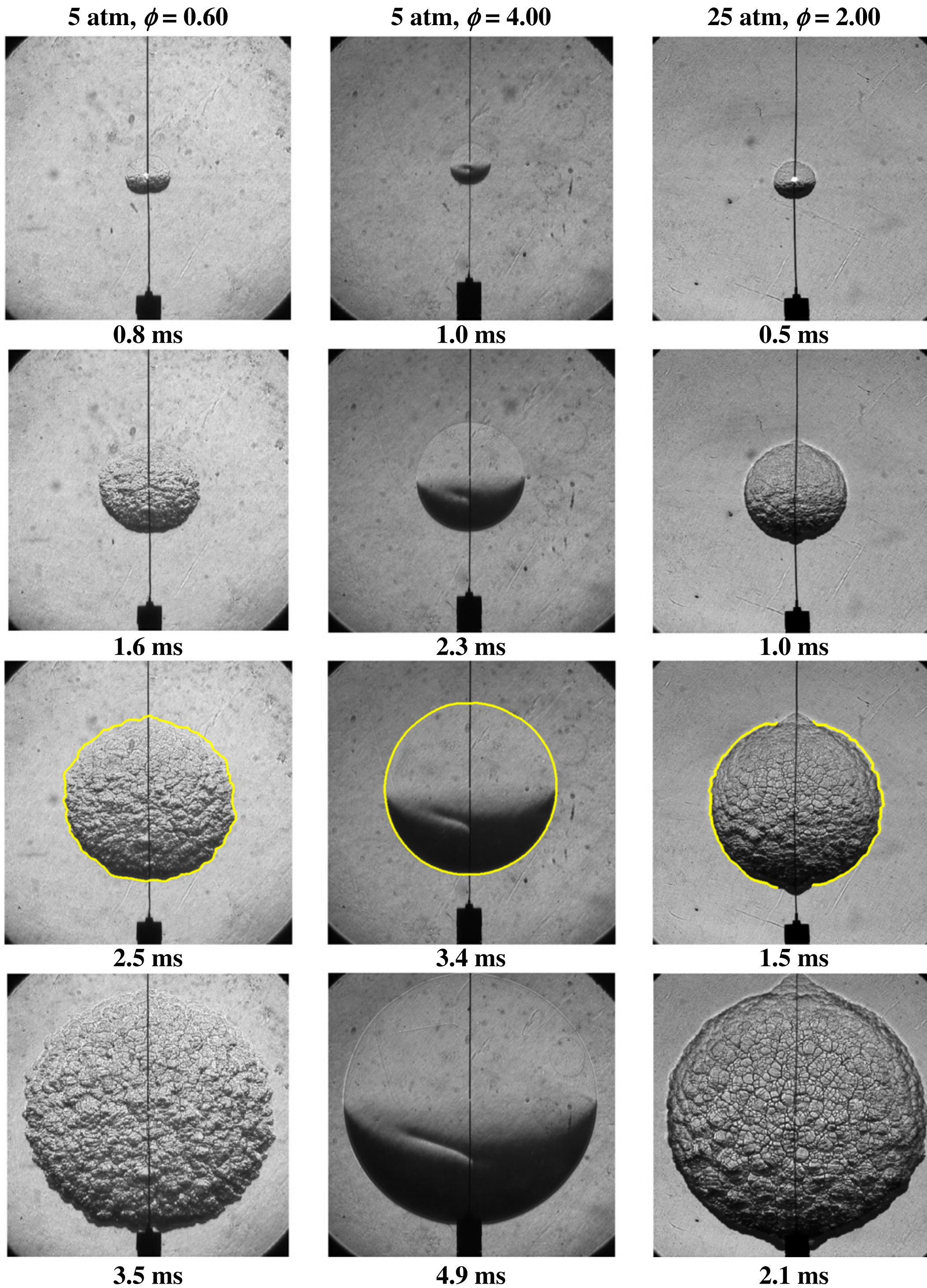

Fig. 2. Sequences of flame propagation in hydrogen/air mixtures, showing that the development of cellular flamefront instability is promoted for $Le < 1$ mixtures and in high pressure environments.

smoothing technique; rather, U_{av} was calculated by taking central difference on R_{av}–t. Owing to the quality of flamefront tracking and the averaging technique used when calculating R_{av}; the local spatial effects caused by each cusp or trough approximately cancel out. For the time step between each camera exposure in the experiment (typically 0.06–0.13 ms), a good statistically meaningful propagation velocity is automatically captured. Therefore, the resulting velocity curve is quite smooth.

4. Results

We are primarily interested in assessing if the propagation of wrinkled flames indeed self-accelerates, and if the acceleration follows a self-similar law. In other words, if we fit the velocity data with a power-law formula, not only should the exponent α be greater than unity but it should also be a constant, at least within a certain range of the propagation. This question is important in its own right, and needs to be addressed before the specific values of α are considered.

4.1. Self-similar propagation regime

Figure 3a and b are logarithmic plots of the propagation velocity versus the flame radius for two experimental runs with the same mixture compositions ($\phi = 0.60$ and 1.00, respectively) but with different ignition energies. Experiments with different ignition energies were conducted to eliminate effects of ignition and cell development on flame acceleration. It is seen that the two curves overlap after certain transition periods. In the overlapping region, the slope of the $\log(U_{av})$ vs. $\log(R_{av})$ remains a constant until chamber confinement starts to have an influence, at about $R_{av} = 2.0$ cm. This result suggests that a self-similar regime does exist for wrinkled laminar flame propagation, after an initial transition period. Note that in the transition period the slope is higher than that in the self-similar regime, which agrees with numerical results [22,23]. The same phenomenon was also observed for other conditions.

4.2. Acceleration exponents and fractal dimensions

In Fig. 4 we plot the experimentally measured acceleration exponent α for hydrogen/air mixtures as a function of the fuel equivalence ratio, ϕ, for various pressures. Three trends are observed. First, the values of α for the lean mixtures as a group are higher than those for the rich mixtures. It is reasonable to suggest that this is due to the effect of diffusional-thermal instability, which is cellularly de-stabilizing for lean mixtures and stabilizing for rich mixtures, and as such can lead to stronger and weaker wrinkling for these mixtures respectively. The second trend is that, for the strongly burning mixtures, with ϕ ranging from 0.60 to 1.20, increasing the pressure results in larger values of α, which implies more wrinkling. This is in agreement with our anticipation in that, for a given ϕ, increasing pressure reduces the flame thickness and thereby increases the propensity of the flame to become hydrodynamically unstable, leading to earlier onset, and possibly more wrinkling. We note, however, that for safety reasons the highest pressure studied for these flames was 10 atm. Therefore, additional experiments at higher pressures and thus thinner flames in this range of ϕ are needed. Third, in the range of $\phi = 1.40-2.00$, increasing the pressure does not change the value of α, within the experimental uncertainty. This can be interpreted as a sign that the maximum value of the acceleration exponent has been reached, as using flames that become unstable at an earlier point does not change the value of the acceleration exponent. It is noted that while diffusional-thermal cellular instability is suppressed for the rich cases ($Le > 1$), pulsating instability [35,36] is activated and its coupled effects with the hydrodynamic cells have yet to be understood. As such; inclusion of detailed

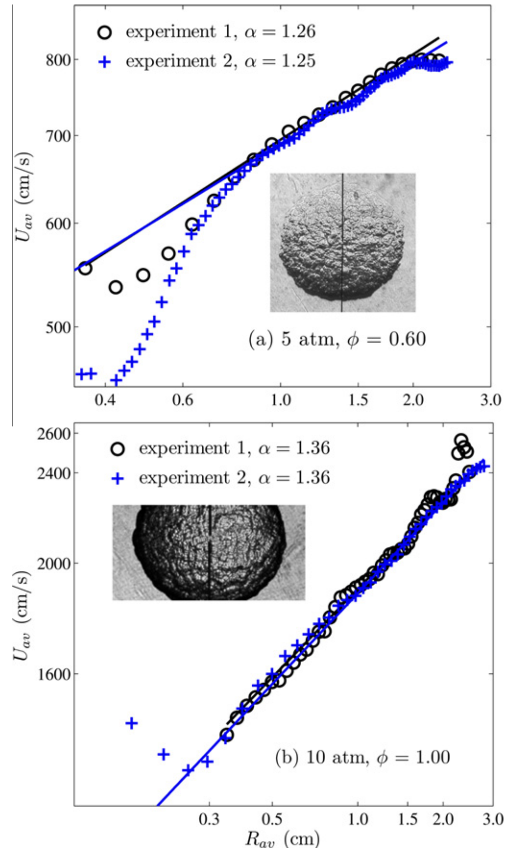

Fig. 3. Propagation velocity for two experiments for H_2/air, at (a) 5 atm, $\phi = 0.60$, and (b) 10 atm, $\phi = 1.00$. Experiments 1 and 2 are runs with the same mixture but different ignition energies.

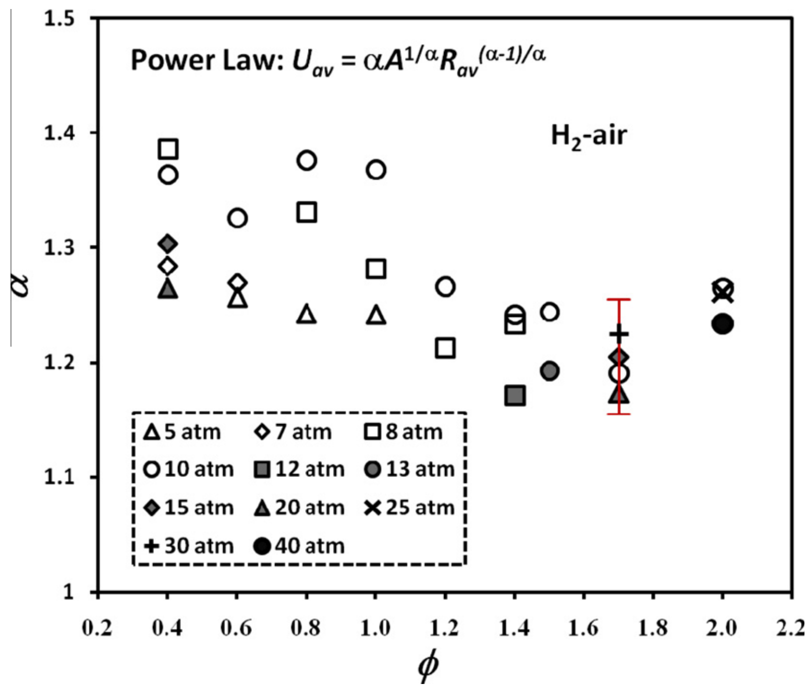

Fig. 4. Experimental acceleration exponents for H_2/air at various pressures and equivalence ratios (all data points are estimated to have uncertainty ± 0.05; however, error bar is indicated only at one point for display clarity).

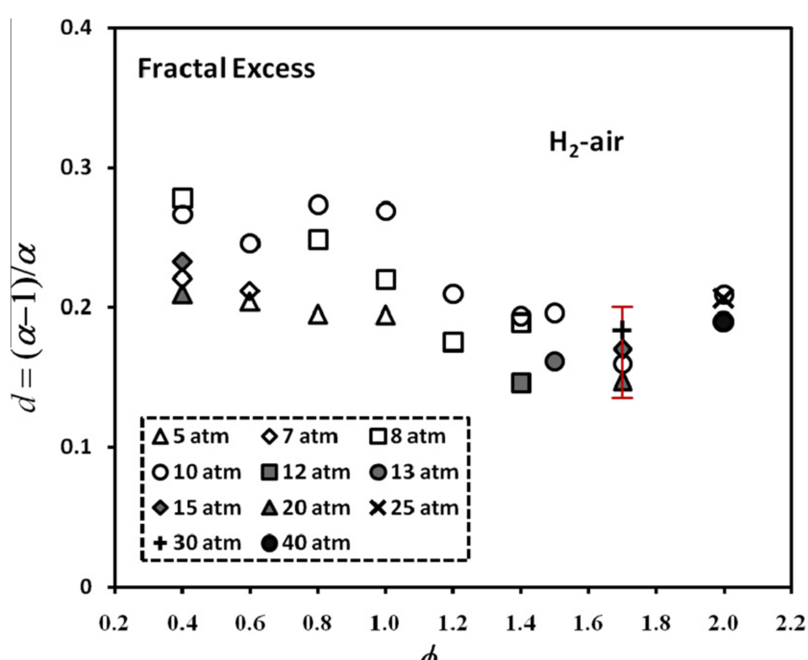

Fig. 5. Experimental fractal excess for H_2/air at various pressures and equivalence ratios (estimated uncertainty is ± 0.03; however, error bar is indicated only at one point for display clarity).

chemistry in the modeling would be beneficial, in order to capture potential subtle effects associated with high-pressure chemistry.

The promoting effect of pressure is also less conclusive for the ultra-lean case of $\phi = 0.40$, for which a consistent trend is not observed. Recognizing that the lean flammability limit for hydrogen/air mixtures at atmospheric pressure is about $\phi = 0.17$, and that increasing pressure could elevate this limit due to enhanced three-body termination reactions, it is reasonable to anticipate the possible occurrence of local extinction and re-ignition, which would introduce additional factors in the flame propagation that are not amenable to direct interpretation.

Recognizing the potential complication due to diffusional-thermal effects in the interpretation of hydrodynamic instability on the self-similar propagation, it is prudent to consider equidiffusive mixtures for which these effects are suppressed. For hydrogen/air mixtures this is close to the stoichiometric value of $\phi = 1.00$. Figure 4 then shows that the acceleration exponent for such mixtures assumes a maximum value of about $\alpha = 1.37$, occurring at 10 atm pressure. This value, determined in a manner that can be considered to be rigorous based on the current understanding and state-of-the-art investigation, is close to; but still sufficiently smaller than $\alpha = 3/2$, the value purportedly related to the concept of self-turbulization.

Having determined the acceleration exponents of the laminar flames; we plot the fractal excess, $d = 1-\alpha^{-1}$, in Fig. 5, showing the approach to $d = 1/3$ for the high-pressure, stoichiometric mixture.

5. Concluding remarks

In the present study we have first briefly, albeit critically, discussed the extent of rigor in the various claims of self-similarity in previous studies of the self-acceleration of expanding spherical flames, particularly on the attainment of 3/2 for the power-law acceleration exponent. Our conclusion is that the evidences presented in some of these prior studies were not rigorously interpreted, and as such the claim of the 3/2 exponent, motivated by the initial suggestion of Gostintsev et al. [11] based on results of uncertain accuracy, and by the attractiveness of its potential implication on self-turbulization, is not justified.

Being mindful of the above concern, we have designed, conducted and analyzed our experiments with considerable caution and thoroughness. Our results suggest that self-acceleration definitely exists and self-similarity, subsequent to the initial transient, most likely exists. Our power-law exponent, however, still seems to weakly depend on the system pressure and the intensity of the diffusional-thermal instability, and as such has not reached the hydrodynamic, flame-sheet limit. Further experiments with even thinner flames and longer observation times are needed to resolve this scientifically and practically important issue on flame propagation.

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