Critical Peclet Numbers for the Onset of Darrieus-Landau Instability in Atmospheric-Pressure Methane-Air Flames

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1 Introduction

The spontaneous formation of cellular structures, generated by the Darrieus-Landau instability [1, 2], has been extensively observed throughout studies of spherical-flame propagation [3, 4, 5]. In the absence of initial turbulence, spherical flames initially propagate as smooth, unwrinkled, laminar flames at a speed governed by the laminar burning velocity and expansion ratio of the flame along with its sensitivity to curvature. At some critical radius, however, the spontaneous formation of cellular structures causes the flame to suddenly accelerate due to an increase in flame surface area. For a given mixture, the radius at which these cells form is a material property, which is commonly normalized by flame thickness and expressed as a critical Peclet number [6, 7].

The increase in flame surface area, and the overall magnitude of acceleration, depends on the amplitude and wavelength of the structures that form. Previous studies have found that these flames accelerate indefinitely with velocities that increase as a power law with radius [8]. In atmospheric-pressure propane-air flames, it has been found that the flame velocity increases by a factor of 1.5 when the flame radius is between three to four times the critical radius [9]. This effect is of particular importance within the context of industrial safety, where accidental explosions can occur in large clouds of flammable mixtures and the peak flame velocity is the primary factor that determines the pressures that develops.

In the work of Gostintsev et al. [8, 10], it was suggested that the formation of fractal patterns on the flame surface are responsible for the sustained acceleration. In a previous study examining propane-air mixtures [9], oscillatory flame acceleration was observed, with frequency intervals consistent with cell splitting and the formation of fractal structures on the flame surface. Based on these results, the original power law relation of Gostintsev et al. [8] was reformulated and the following relationship between propagation velocity and flame radius was obtained [9]:

$$u / u_0 = \left(R / R_0\right)^{\beta} .$$

where u_0 is the laminar flame speed of the mixture, R_0 is the critical radius of onset of instability, and β is a fractal dimension that can be obtained experimentally. This result suggests that the critical radius is an integral parameter in determining the flame speed of a spherical flame at a given radius.

The objective of this study is to examine methane-air mixtures over a range of equivalence ratios, at large scale, to experimentally determine the critical Peclet number for the onset of Darrieus-Landau instability at atmospheric pressure. This is a continuation of a previous study examining propane-air flames [9]. The results of the methane-air mixtures will also be compared with propane-air flames which have an opposite relationship between stretch/curvature and equivalence ratio. Due to the higher Markstein numbers of methane-air mixtures, the flame radius is tracked to larger scales and higher resolution imaging is used to capture the development of the cellular structures on the flame surface. This study will also examine whether oscillatory flame acceleration is also exhibited by methane-air mixtures.

2 Experimental Setup

The experiments were performed in a 64 m^3 vented enclosure with dimensions of $4.6 \times 4.6 \times 3.0$ m^3, as shown in Fig. 1, with a single 5.4 m^2 vent to maintain a constant internal pressure during flame propagation. Ignition was supplied by an automotive ignition system using two 50 μm tungsten wires as electrodes. The overall ignition energy was estimated to be less than 100 mJ and was the minimum ignition energy required to ignite the mixtures at the limits of concentration range studied. A thin polypropylene sheet was used to contain the initial mixtures during filling and mixing. The sheet was cut before each test and held in place using pneumatic clamps that were released 1 s prior to ignition.

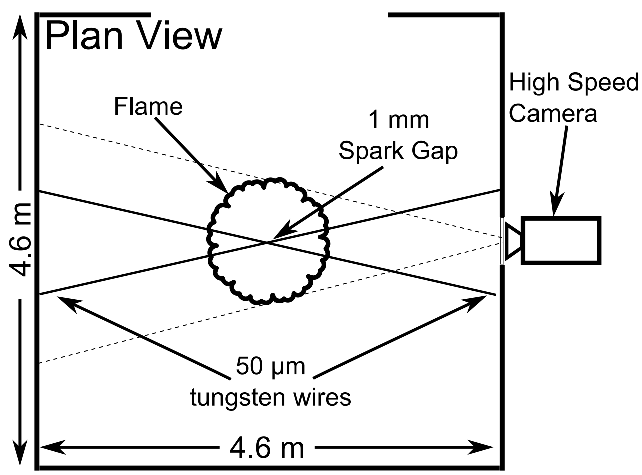

Figure 1. Schematic of enclosure and ignition configuration

A range of methane-air concentrations were examined from equivalence ratios of $\varphi = 0.81 - 1.22$. In addition, experiments were also performed for propane-air mixtures with an equivalence ratio ranging from $\varphi = 0.89 - 1.41$ to replicate the previous study and ensure that any changes to the test configuration did not influence the results. This also allowed for higher resolution images of cell formation and splitting to be obtained. Concentration was monitored and controlled using a 3.39 μm laser extinction system, calibrated prior to each experiment using a sample mixture prepared via partial pressures and ambient air, and verified using an Illinois Systems P710 paramagnetic oxygen analyzer. Overall experimental uncertainty in mixture equivalence ratio was estimated to be within $\varphi \pm 0.03$.

The full flame cross-sections were imaged using a Vision Research Phantom Flex high speed camera for flame diameters up to 2 m. Flame speed was calculated from flame radii extracted from 1,600 ×

1,600 pixel images captured at a rate of 2,000 frames per second and the onset of cellular instability was easily extracted from flame speed using the procedure described in the previous study [9].

3 Results

A summary of test conditions and critical Peclet numbers are provided in Table 1 where *T* is the initial temperature, *R.H.* is the relative humidity, Ma is the Markstein number of the mixture taken from the literature [11] and Pe_c is the critical Peclet number. The Markstein and Peclet numbers were normalized using a flame thickness, estimated from kinematic viscosity divided by laminar burning velocity, for consistency with the previous studies that are examined in the following sections.

Table 1: Summary of test conditions and experimental results.

φ	T (K)	$R.H.$ (%)	R_0 (m)	Ma [11]	Pe_c
0.81	301	43%	0.204	2.8	3305
0.83	303	51%	0.209	3.1	3572
0.88	307	46%	0.214	3.8	4263
0.89	301	48%	0.213	3.7	4146
0.89	297	91%	0.216	3.5	4003
0.92	307	53%	0.220	4.2	4692
0.93	296	92%	0.215	3.9	4283
0.99	304	40%	0.220	4.9	5068
1.00	302	43%	0.225	4.9	5124
1.04	297	55%	0.233	5.3	5239
1.07	304	53%	0.238	6.0	5567
1.12	301	43%	0.267	7.0	6074
1.14	303	59%	0.294	7.5	6577
1.17	299	84%	0.334	8.2	6943
1.19	306	52%	0.315	9.3	6762
1.22	299	50%	0.324	9.7	6319

The high-resolution camera used in this study also captured details of initial flame-surface crack formation prior to the onset of cellular instability, as shown in Fig. 2 for a $\varphi = 1.17$ methane-air flame. From the temporal evolution of the cracks observed in the high speed images, the cracks appear to be generated primarily from disturbances present in the original ignition kernel and from disturbances created near the electrodes. In some mixtures, particularly lean propane and rich methane, toroidal patterns can be observed along with spiral patterns in regions of flame propagation along electrodes.

The detailed evolution of the flame surface and the results of cell splitting were directly observed, as shown in Fig 3, where the flame surface of a $\varphi = 1.2$ propane-air flame is shown following each generation of cell formation during oscillatory flame acceleration. It is important to note that the decrease in apparent size of smaller scales is a result of resizing the images taken at different flame radii to fill the vertical space. The minimum cell size remains roughly constant throughout the flame propagation.

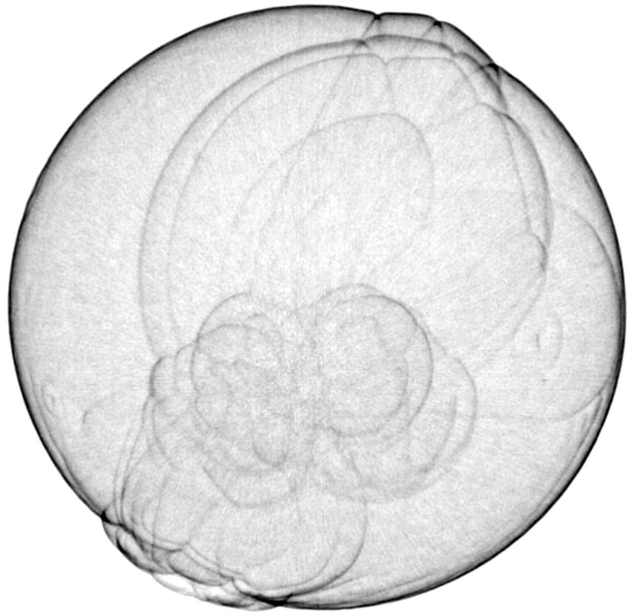

Figure 2. Disturbances on the flame surface prior to the onset of Darrieus-Landau instability for a $\varphi = 1.17$ methane-air mixture at a radius of 0.33 m.

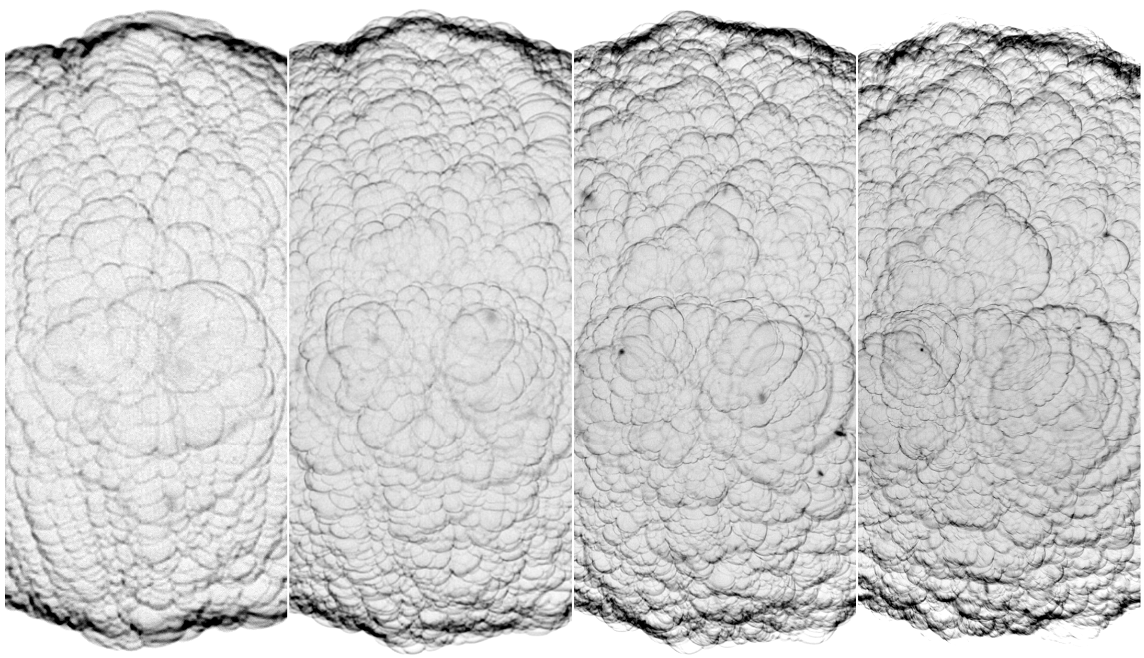

Figure 3. Slices of the flame surface from a single $\varphi = 1.2$ propane-air experiment at different times where $R = 0.20$, 0.35, 0.55, 0.75 m.

4 Discussion

The previous propane-air test series found that the critical Peclet numbers decreased monotonically with increasing propane concentration while the present study show the critical Peclet numbers increase with methane concentration. This is to be expected as the flame response to local curvature is the primary mechanism through which the flame is stabilized prior to the onset of instability. In fact, when plotted in terms of the Markstein number, a linear relationship between Markstein length and critical Peclet number is observed, as shown in Fig. 4. Compared to other studies performed at smaller scale over a range of initial pressures, a stronger variation in critical Peclet number with Markstein number was observed along with reduced scatter. This reduced scatter may be a result of the experiments being performed at a single initial pressure and at a larger scale, which reduces the influence of ignition.

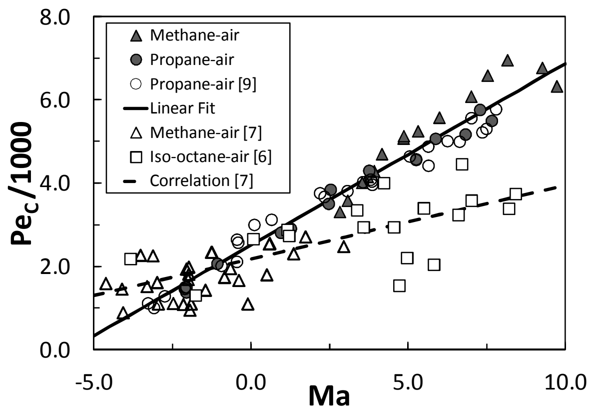

Figure 4. Comparison of critical Peclet numbers with the empirical correlation of Gu et al. [7] and existing experimental data [6, 7, 9].

Figure 5 shows the measured flame speed as a function of flame radius normalized by laminar flame speed and critical radius, respectively. Oscillations are clearly observed for all of the mixture compositions studied. At the end of several curves, particularly for rich mixtures, the flame velocity drops compared to the regular oscillatory trend. This drop is likely due to wall effects which reduce the upstream flow velocity as the flame diameters are approaching half the chamber width. It is important to note, however, that shifts in the frequency of oscillations, which were observed in rich propane-air mixtures, were not observed in the methane-air mixtures due to significantly higher Markstein numbers.

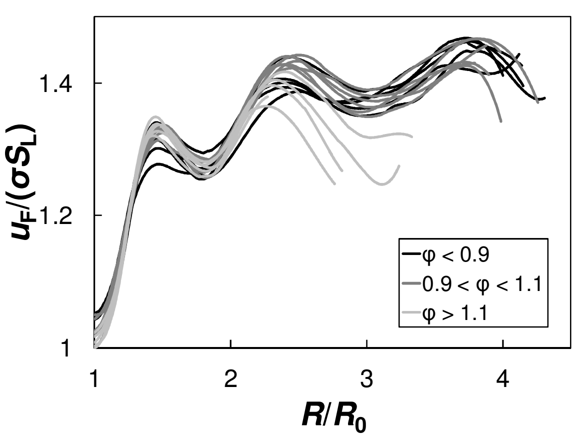

Figure 5. Normalized flame speed vs. normalized flame radius showing self-similar flame propagation for methane-air mixtures.

5 Summary and Conclusions

Experiments were performed examining the critical Peclet number for the onset of the Darrieus-Landau instability in spherical methane-air flames with flame diameters up to 2 m. For these atmospheric pressure methane-air flames, critical radii on the order of 20 – 30 cm were observed. Throughout the range of methane concentrations studied, critical radii increased with methane concentration, consistent with changes in mixture Markstein number. A linear relationship between critical Peclet number and Markstein length was also found across both methane-air and propane-air results. In addition, oscillatory flame propagation was observed in the cellular methane-air flames, consistent with results of previous large scale experimental observations of propane-air mixtures.

References

[1] G. Darrieus, unpublished work presented at La Technique Moderne, and at Le Congrès de Mécanique Appliquée (1945) and (1938)

[2] Landau, L. D. (1944). On the theory of slow combustion. Acta Physicochim. URSS, 19(1), 77-85.

[3] Istratov, A. G., & Librovich, V. B. (1966). On the stability of propagation of spherical flames. Journal of Applied Mechanics and Technical Physics, 7(1), 43-50.

[4] Bradley, D., Cresswell, T. M., & Puttock, J. S. (2001). Flame acceleration due to flame-induced instabilities in large-scale explosions. Combustion and Flame, 124(4), 551-559.

[5] Law, C. K., Jomaas, G., & Bechtold, J. K. (2005). Cellular instabilities of expanding hydrogen/propane spherical flames at elevated pressures: theory and experiment. Proceedings of the Combustion Institute, 30(1), 159-167.

[6] Bradley, D., Hicks, R. A., Lawes, M., Sheppard, C. G. W., & Woolley, R. (1998). The measurement of laminar burning velocities and Markstein numbers for iso-octane–air and iso-octane–n-heptane–air mixtures at elevated temperatures and pressures in an explosion bomb. Combustion and Flame, 115(1), 126-144.

[7] Gu, X. J., Haq, M. Z., Lawes, M., & Woolley, R. (2000). Laminar burning velocity and Markstein lengths of methane–air mixtures. Combustion and flame, 121(1), 41-58.

[8] Gostintsev, Y. A., Istratov, A. G., & Shulenin, Y. V. (1988). Self-similar propagation of a free turbulent flame in mixed gas mixtures. Combustion, Explosion, and Shock Waves, 24(5), 563-569.

[9] Bauwens, C. R., Bergthorson, J. M., & Dorofeev, S. B. (2014). Experimental study of spherical-flame acceleration mechanisms in large-scale propane–air flames. Proceedings of the Combustion Institute.

[10] Gostintsev, Y. A., Fortov, V. E., & Shatskikh, Y. V. (2004, July). Self-similar propagation law and fractal structure of the surface of a free expanding turbulent spherical flame. In Doklady Physical Chemistry (Vol. 397, No. 1, pp. 141-144). MAIK Nauka/Interperiodica.

[11] Taylor, S. C. (1991). Burning velocity and the influence of flame stretch (Doctoral dissertation, University of Leeds).