An Experimental Determination of the Laminar Burning Velocities and Extinction Stretch Rates of Benzene/Air Flames

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By using the counterflow technique, the burning velocities and lean extinction stretch rates of premixed, prevaporized and preheated benzene/air mixtures were determined for the adiabatic limit, as well as for the sub- and superadiabatic situations characterized by downstream heat loss/gain. Values of laminar burning velocities are obtained at atmospheric pressure and elevated temperatures. Flame stretch, in the presence of substantial diffusional imbalance due to the relatively small mass diffusivity of benzene, was found to have a strong effect on the burning velocity. Downstream heat loss was also shown to have some measurable influence, and the effect in terms of the fuel concentration for the lean extinction states can be substantial. The results will be of use in the simulation of engine combustion utilizing fuel blends with substantial aromatic contents, and the modeling of the flame kinetics of aromatic fuels.

INTRODUCTION

In the past two decades extensive progress on understanding the oxidation kinetics of paraffinic fuels has been made. Such progress has been facilitated by two major advances which allow the close coupling of experimental and computational investigations. The first is the computational simulation and comparison of combustion phenomena and parameters with detailed chemistry [1, 2]. Through such a comparison the adequacy and weaknesses of a given reaction scheme can be identified by using, for example, sensitivity and reaction path analyses. The experimental information which has been used for such a comparison includes ignition delays, laminar burning velocities, extinction stretch rates, and the scalar structure of flames. The second advance is the recognition [3, 4] that previous experimentally determined values of laminar burning velocities were frequently, and extensively, complicated by stretch effects such that significant uncertainties existed when they were used to "validate" or "partially validate" a kinetic mechanism. Law [3, 4] subsequently proposed the counterflow twin flame technique to eliminate these stretch effects by first determining appropriate reference burning velocities of flames with various stretch rates, and then linearly extrapolating these reference burning velocities to zero stretch rate, to yield the stretchless laminar burning velocity. This technique has since been refined to allow for nonlinear extrapolation [5, 6] when the nozzle separation distance is not sufficiently large and/or the Karlovitz number not sufficiently small [7]. The general concept of including stretch effects in processing experimental laminar burning velocity data has also been extended to spherical flames [8–10].

Although advances in flame kinetics have been made for paraffinic fuels, the progress for aromatic fuels has been relatively less [11]. Thus the primary objective of the present investigation is to provide accurate experimental data on the laminar burning velocities and extinction stretch rates of benzene/air flames, using the counterflow technique, with the anticipation that these data will be useful for further studies on benzene oxidation. We have also investigated the effects of downstream heat loss/gain on the laminar burning velocity and flame extinction, and provided quantitative information on the magnitudes of such influences. The experimental laminar burning velocities and extinction stretch rates are also expected to be of value in the study of engine combustion, because aromatics constitute a portion of the automotive fuel blend.

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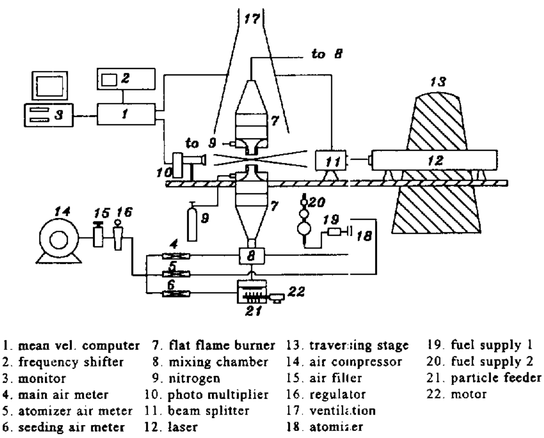

Fig. 1. Apparatus.

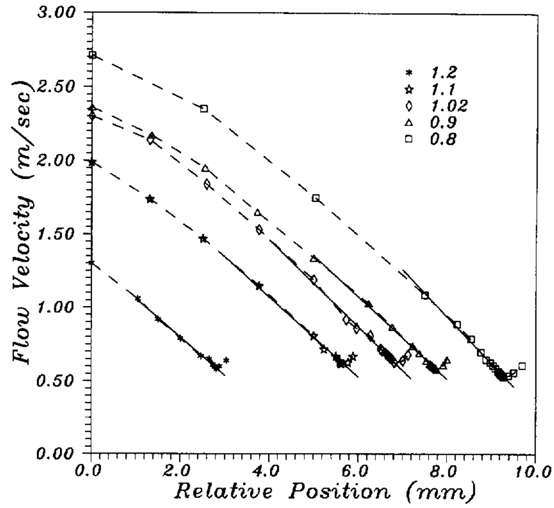

Fig. 2. Representative velocity profiles along the central axis.

EXPERIMENTAL METHODOLOGY

The apparatus (Fig. 1) consists of a one-dimensional LDA system, a 3-axis traversing stage, an axisymmetric counterflow burner, fuel and air supplies, particle seeding units, and heating elements with voltage regulators. The LDA system consists of a 25-mw He–Ne laser with modular beam splitter and Dantec Type 55N11 Frequency Shifter. The forward scattered light signal from the $0.1 \sim 1$ μm MgO seeding particles is collected at a photomultiplier and the average scattered light frequency is computed using a Dantec 55L94 velocity computer to yield the average particle velocity. The axisymmetric counterflow burner consists of two identical opposed nozzles, with an area contraction ratio of 9:1, an exit diameter of 18 mm, a separation distance of 18 mm, and nitrogen co-flow.

In order to ensure accurate velocity measurement, the LDA system was first calibrated by a motor-driven rotational disc in the velocity range from 0.4 to 2 m/s; the accuracy was found to be $\pm 0.3\%$. The cold air axial velocity profiles at the nozzle exit plane were also tested before taking the flame data. The distribution at different air flow rates was found to be uniform at most central regions and changed rapidly only close to the free boundary, thus demonstrating that the design of the nozzle was satisfactory.

Technical-grade liquid benzene with a purity exceeding 99.5% was steadily supplied by two independent and identical DC micro-gear pumps. It was subsequently vaporized by flowing part of the air through a venturi to generate a high-speed, low-pressure air stream to draw it through a small hole inside the venturi. The fuel formed a liquid spray which vaporized either within the air stream or on the hot surface of copper tubing. The temperature of the hot surface was carefully controlled so as not to exceed 473 K, to prevent thermal pyrolysis of the benzene. All the mixture supply lines and the burner were wrapped with electric heating coils and thermal insulators, such that the temperature was much higher than the dew point of the mixture, in order to prevent the benzene vapor from condensing in the flow circuit. The mixture temperature was measured at the exit plane of the nozzle by a J-type thermocouple, and was controlled to within 1 K by the supply voltage to the heating coils.

By carefully adjusting the air, fuel, and nitrogen flow rates as well as the heating power, stable stationary flat flames were obtained. The mixture equivalence ratio was calculated from the reading of the flow meters, accurate to 2%. The flow velocities of the lower and upper flows were measured along the central axis from $3 \sim 4$ mm upstream of the flame to a location slightly downstream of the velocity minimum, V_{min}. Figure 2 shows some representative velocity profiles of the lower flame for the adiabatic, twin flame configuration. These velocity profiles allowed the determination of the velocity gradient, dV/dx, which is the negative of the stretch rate, K. The extinction stretch rate for a lean mixture was determined by measuring the flow velocity corresponding to the state beyond which the flame would extinguish with a slight increase in the air flow. In the present investigation the stretch rates were varied between 150 ~ 350/s, while the initial mixture temperatures were between 348 and 398 K and at atmospheric pressure.

Three types of experiments were performed to allow for various degrees of downstream heat transfer. In a symmetrical, twin flame configuration, properties of the combustibles issuing from the two nozzles were identical. Thus upon ignition two identical flames were produced on the two sides of the stagnation surface. Due to the symmetry the extent of heat loss downstream of the flame is minimal and the flames are approximately adiabatic. In the second arrangement the upper flow of combustibles was replaced by room-temperature nitrogen, such that only one flame was produced. Furthermore, since the downstream boundary temperature for this flame is the room temperature, this nitrogen-opposed flame not only is subadiabatic through downstream heat loss, but the extent of downstream heat loss can be considered to be a maximum. In the third arrangement, the upper combustible was replaced by room-temperature air, so that with a lean flame the opposed air flow is reasonably inert, again providing the maximum amount of downstream heat loss. However, for a rich flame the excess combustible species downstream of the flame can further react with the oxygen in the air, releasing additional heat. The flame therefore possibly becomes superadiabatic, receiving heat from downstream. It is therefore clear that the propensity to flame extinction with increasing stretching could vary quite substantially for these three types of arrangements.

RESULTS AND DISCUSSIONS

Determination of Laminar Burning Velocities

Determination of the laminar burning velocities utilizes the twin flame configuration because these flames are nearly adiabatic. Figures 3 and 4 plot the experimental values of the minimum velocity or the burning velocity, V_{min}, of the lower flame, versus the stretch rate K for the lean and rich mixtures, respectively, at the mixture temperature of 363 K. In comparison with experiments using gaseous fuels, it is much more difficult to obtain stable flames of vaporized liquid fuels. Specifically, liquid fuels must not only be prevaporized, they must also be heated to prevent condensation. Furthermore, the temperature of the mixture must be monitored and adjusted continually. Hence, much effort was required to obtain each datum shown in the figures. Stretch rates were obtained over as wide and as low a range as possible. These results clearly show the flame propagation velocity to be affected by flame stretch. The influence of K also exhibits a systematic trend, in that for very lean mixtures, such as with an equivalence ratio, ϕ, of 0.7, V_{min} decreases with increasing K. However, this trend is moderated as ϕ increases, and there is almost no effect of K on V_{min} at $\phi = 0.8$. The trend is reversed with further increase in ϕ, whereupon V_{min} increases with increasing K, and this becomes more pronounced as the mixture becomes richer.

The above results are consequences of stretch in the presence of thermal expansion and mixture nonequidiffusion. It has been shown analytically [6] that, for a mixture with equal thermal and mass diffusivities such that the Lewis number, Le, is unity, the combined effects of flow divergence and thermal expansion will lead to an increase in the reference, minimum velocity, V_{min}, with increasing stretch rate. Here Le is defined as $\alpha_m/D_{k,N_2}$ where α_m is the thermal diffusivity of the mixture and D_{k,N_2} is the mass diffusivity of the deficient species relative to the abundant species, N_2 in the present case. It is also well established [4] that, for a positively stretched counterflow flame, the burning intensity increases with increasing stretch rate when Le is less than unity and decreases when it is greater than unity. One would therefore anticipate that, except for mixtures for which Le is much larger than unity, V_{min} would increase with increasing stretch.

The results of Figs. 3 and 4 can be understood in the light of the above factors. For the lean mixtures, the controlling species is the relatively heavy benzene whose diffusivity in the mixture

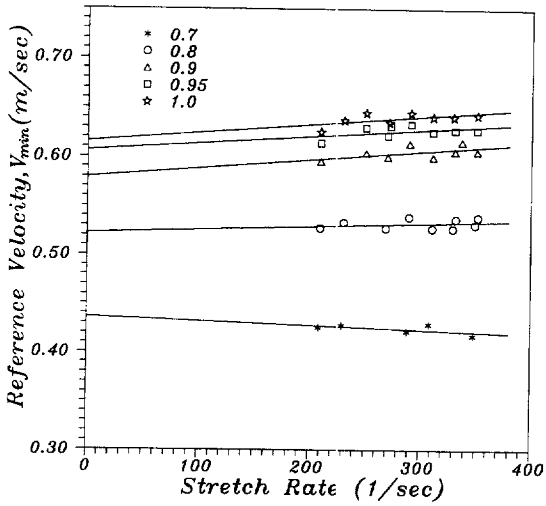

Fig. 3. Minimum velocity vs stretch rate, lean mixtures.

is rather low. The value of Le for the very lean mixtures should therefore be much larger than unity. For example, for $\phi = 0.7$, Le is estimated to be 2.31 by the Sandia Code [12]. This, therefore, yields the decreasing trend as shown. However, for rich mixtures the controlling species is oxygen and Le is less than unity, for $\phi = 1.3$, Le is 0.95. Thus the decreasing trend should be reversed as ϕ increases from the ultralean values, in agreement with experimental results. The value of $V_{\min}$ is quite insensitive to stretch for the near-stoichiometric mixtures. Furthermore, the slope of the plot of $V_{\min}$ versus K

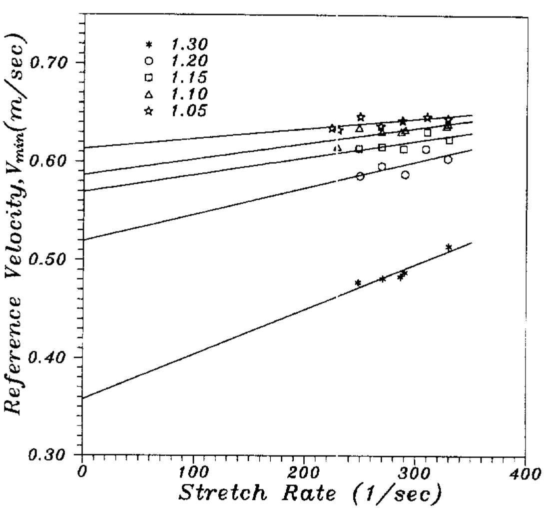

Fig. 4. Minimum velocity vs stretch rate, rich mixtures.

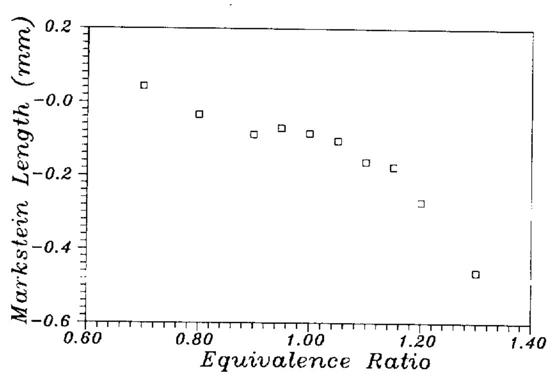

Fig. 5. Markstein length vs equivalence ratio.

gives the Markstein length [13], and Fig. 5 then shows these for different equivalence ratios, obtained from Figs. 3 and 4. The Markstein length varies from $(4.28) \times 10^{-2}$ to $(-4.61) \times 10^{-1}$ mm as the equivalence ratio changes from 0.7 to 1.3.

Linear extrapolation to zero stretch rate, $K = 0$, as shown in Figs. 3 and 4, yields the laminar burning velocities for these mixtures, S_u^o [3, 4]. Values from this extrapolation have a standard deviation of 1%. These values are shown in Fig. 6 as a function of the equivalence ratio. Since the freestream is heated to effect prevaporization, the laminar burning velocities shown in Fig. 6 are uniformly higher than room temperature literature values for initially unheated benzene/air mixtures. In the reference of Brad-

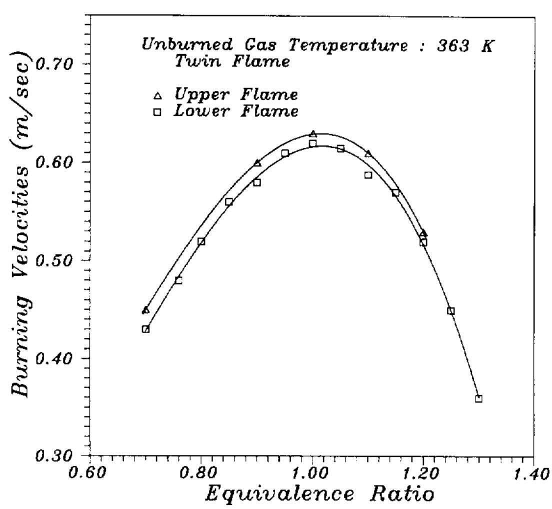

Fig. 6. Upper and lower flame burning velocity vs equivalence ratio at 363 K and 1 at. pressure.

ley et al. [14], the maximum burning velocities are within the range of 0.39 to 0.49 m/s at room temperature, compared with a value of 0.61 m/s for the present preheated mixture at 363 K. However, a direct comparison is not possible because of the different freestream temperatures.

Although the present results were obtained by linear extrapolation of the stretched flame data, it has been suggested that such extrapolation should be nonlinear [6], and this would yield lower values [6, 15]. Nevertheless, Vagelopoulos et al. [7] show that the differences between the linear and nonlinear extrapolation are usually small, and that they can be further minimized at sufficiently large nozzle separation distances. Since the present nozzle separation distance is considered to be large [7], and since the scatter in the present experimental data prevents identification of any nonlinear trend, the linear extrapolation is justified. Finally, we note that the accuracy of the extrapolated results improves with the use of small values of the stretch rate or Karlovitz number, *Ka* [6], where *Ka* is defined as the $K \times \alpha_m/(S_u^o)^2$. The values of *Ka* range from 0.02 to 0.08, sufficiently small to justify linear extrapolation.

Effects of Buoyancy on Burning Velocities

Figure 6 also shows the unstretched laminar burning velocities for the upper flames of the twin flame configuration at different equivalence ratios. Analogous to the steady propagation of free flame surfaces in a flame tube, in which the upwardly moving flame has a higher propagation velocity than the downwardly moving flame, the same buoyancy effect should also be operative for the present stationary flame, albeit with a much weaker influence because forced convection is the dominant mode of convection here. Thus, Fig. 6 shows that the upper yields slightly higher laminar burning velocities than the lower flame, with a maximum difference of about 4%. These results are in agreement with the previous observations of Egolfopoulos and Law [16]. We therefore conclude that in a gravitational field, there are directional differences in the measured flame propagation velocities, and an average might be obtained from the upward and downward oriented flame values in order to minimize such an influence. These buoyancy effects could also become more important for weak flames of limit composition in mixtures.

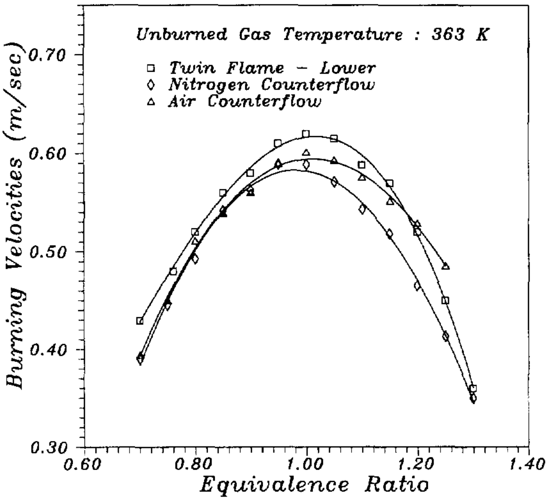

Fig. 7. Burning velocity of twin, nitrogen and air counterflow flame at 363 K and 1 at. pressure.

Effects of Downstream Heat Transfer on Burning Velocities

Downstream heat loss is minimized with the twin flame configuration. In order to assess the extent of the influence of downstream heat transfer, we first experimented with counterflows of combustible mixture against cold nitrogen flow. This yielded a single flame with maximum heat loss. Figure 7 compares the stretch-free laminar burning velocities from the adiabatic twin flame configuration with those determined with the cold downstream flow. The downstream heat loss slightly reduces the laminar burning velocities. The maximum value for the adiabatic laminar burning velocity, 0.61 m/s, is reduced to 0.58 m/s when nitrogen is used as the counterflowing gas. This suggests that downstream heat loss also has some measurable influence on flame propagation.

In the second series of experiments nitrogen was replaced by air as the counterflow gas. Figure 7 shows that the measured burning velocities for the air counterflow agree well with those for the nitrogen counterflow at equivalence ratios less than about 0.85, but assume higher values at higher equivalence ratios. These results confirm the earlier expectation that for lean mixtures the air counterflow has the same role as the nitrogen counterflow in providing a downstream heat sink. However, as the mixture becomes richer, the combustible species from the flame further react with the oxygen in the counterflowing air, resulting in a second (diffusion) flame. This downstream heat release increases the apparent burning velocity. The enhancement is greatly increased when the mixture becomes rich because of the substantial amount of combustible species unreacted through the premixed flame.

The combined effect of heat loss/gain is clearly demonstrated by comparison of the burning velocities for the twin flame and for the air counterflow, shown in Fig. 7. With $\phi < 1.2$, the flame loses heat downstream, while for $\phi > 1.2$ the flame gains heat from downstream. It is of interest to note that even in the presence of a diffusion flame with the air counterflow, the temperature of the diffusion flame is still lower than that of the premixed flame, for ϕ between 1.0 and 1.2, resulting in downstream heat loss.

Lean Extinction and Flammability Limits

Since the Lewis number of lean benzene/air flames is greater than unity, extinction can be achieved by positive stretch alone [4]. A unique extinction stretch rate exists for a given value of ϕ. Figure 8 shows the equivalence ratio at extinction to increase with an increasing stretch rate, as expected. Because the variation is linear, the value of ϕ can be extrapolated to zero stretch to yield the lean flammability limit of the mixture, in accordance with the suggestion of Law and Egolfopoulos [17]. Thus, for a mixture temperature of 348 K, the lean limit concentration is 1.45%, or $\phi = 0.524$.

Increasing the mixture temperature reduces both the equivalence ratio at extinction and the flammability limit, as shown in Fig. 8. Specifically, the lean limit concentrations and equivalence ratios for mixture temperatures of 373 and 398 K are 1.42% (0.516) and 1.40% (0.508), respectively. According to the data of Coward and Jones [18], the lean flammability limit for a mixture temperature of 298 K is 1.4%, determined by either the flame tube or the constant

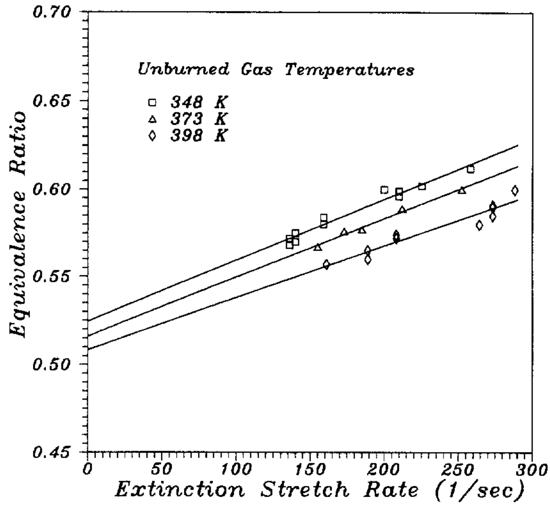

Fig. 8. Equivalence ratio vs extinction stretch rate at different temperatures.

volume bomb method. The present study suggests a value of 1.48% with linear extrapolation of the obtained values with temperature to 298 K. This is a little higher than the value of Coward and Jones [18], but is comparable.

Finally, extinction limits with the maximum downstream heat loss, with counterflowing nitrogen, have also been determined. Figure 9 shows the values of ϕ at extinction to be substantially increased by heat loss, as expected.

The linear behavior of the mixture concentra-

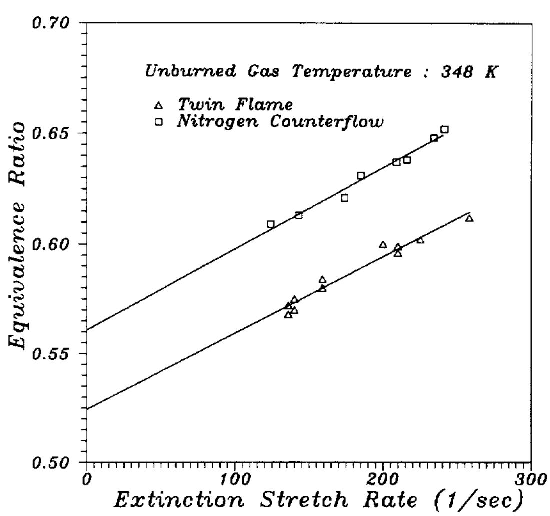

Fig. 9. Extinction stretch rate of twin and nitrogen counterflow flames.

tion with the extinction stretch rate yields useful quantitative information for the assessment of the extinction states. The rate of change of the extinction stretch rate with the volume percent (equivalence ratio) of benzene is about $10^{-3}\%-s$ ($3 \times 10^{-4} - s$), with and without heat loss. The effect of temperature on the flammability limit is about $8.8 \times 10^{-4}\%/K$ ($3.2 \times 10^{-4}/K$).

CONCLUSIONS

The present study has successfully determined the laminar burning velocities and extinction stretch rates of benzene/air flames by using heated prevaporized mixtures and the counterflow technique. These experimental data are useful for the modeling studies of flame chemistry of aromatic fuels. The experimental results also show the strong influence of the Lewis number, primarily due to the large molecular weight of benzene. Furthermore, it is shown that downstream heat loss has some influence on the laminar burning velocity, and that buoyancy exerts a measurable influence. The latter point is of particular interest, suggesting the need for additional fundamental study of the influence of body forces on the structure and response of flames in general. Finally, the observed linearity between the extinction stretch rate and the fuel concentration at extinction provides useful guidelines for empirical estimations of the flammability limits of lean mixtures.

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