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# Effect of stoichiometric mixture fraction on nonpremixed H<sub>2</sub>–O<sub>2</sub>–N<sub>2</sub> edge-flames

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#### Abstract

The influences of stoichiometric mixture fraction  $(Z_{st})$  and global strain rate ( $\sigma$ ) on the shapes and propagation rates ( $U_{edge}$ ) of nonpremixed edge-flames in  $H_2-N_2/O_2-N_2$  mixtures were investigated using a counterflow slot-jet apparatus. Both positive and negative  $U_{edge}$  were observed depending on dilution level,  $Z_{st}$  and  $\sigma$ . At low  $Z_{st}$  only continuous flames were observed whereas at sufficiently high  $Z_{st}$ , where a shift from more oxygen-deficient to more fuel-deficient conditions at the reaction zone occurs, broken structures characteristic of low Lewis number premixed flames were observed which enabled combustion under conditions where no flames could be sustained at lower  $Z_{st}$ , even for the same dilution level. At sufficiently high  $\sigma$  these broken structures could transition from advancing edge-flames to isolated, stationary flames, particularly for highly-diluted mixtures. These findings were in surprisingly good agreement with theoretical predictions. Appropriate scalings of these behaviors for different mixtures based on computed 1D extinction strain rates were identified. Nonpremixed  $H_2-N_2/O_2-N_2$  edge-flames have profoundly different responses to  $Z_{st}$  than corresponding hydrocarbon edge-flames, which is shown to be due to differences in the chemistry and Lewis numbers of the two fuels.

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Keywords: Edge-flames; Lewis number; Mixture fraction; Nonpremixed flames; Counterflow

#### 1. Introduction

"Edge-flames" [1,2] are transition regions between burning and non-burning portions of flame sheets that may exist in situations such as flames stabilized near a cold wall or splitter plate, leading edges of flames spreading across condensedphase fuel surfaces, or flame sheets in highly tur-

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bulent flows where "holes" may open or re-seal [3]. Edge-flame behavior also strongly affects flame dynamics in forced unsteady Burke-Schumann configurations [4] and other flame stabilization scenarios [5]. The most significant property of an edgeflame is its propagation speed ( $U_{edge}$ ), defined as the speed of the edge moving from the burned gases towards the unburned gases in the direction parallel to the flame sheet. Previous theoretical studies in premixed [6–8] and nonpremixed [9–11] configurations predict that edge-flames may propagate from

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the burning region into the unburned region, forming a "ignition front" with  $U_{edge} > 0$  or retreat from the burning region into the burned region, forming an "extinction front" with  $U_{edge} < 0$ .  $U_{edge}$  is affected by factors including global strain rate ( $\sigma$ ), Lewis numbers (*Le*, ratio of mixture thermal diffusivity to reactant mass diffusivity) of fuel and oxidant, heat losses, mixture strength and for nonpremixed edge-flames the stoichiometric mixture fraction  $Z_{st} \equiv 1/(1 + mX_f/X_o)$  where *m* is the stoichiometric oxygen-to-fuel mass ratio and  $X_f$  and  $X_o$ are the mass fractions of fuel and oxygen in the reactant mixture streams.

While pure fuel burning with highly-diluted oxygen (air) corresponds to low  $Z_{st}$  (typically 0.06 for hydrocarbon-air mixtures) is prevalent in traditional nonpremixed combustion, new fuels and combustion technologies (biofuels, oxyfuel combustion, massive exhaust gas recirculation, etc.) lead to much broader  $Z_{st}$  ranges (up to 0.8 for pure O<sub>2</sub> burning with highly-diluted hydrocarbon fuel and 0.93 for highly-diluted  $H_2$ .) In a counterflow, as  $Z_{st}$  increases the flame location moves from the oxidizer side of the stagnation plane to the fuel side, resulting in significant differences in the reactant temperature/composition/time history which in turn substantially affects burning rates as characterized by  $U_{edge}$  or extinction strain rate  $(\sigma_{ext})$  [12– 14]. For hydrocarbons, there are two competing factors. One is a chemical effect: at low  $Z_{st}$  the flame location is on the oxygen side of the stagnation plane and radicals produced primarily by the fuel must diffuse upstream to the reaction zone whereas at high  $Z_{st}$  the flame resides on the fuel side and radicals are readily convected downstream to the reaction zone, thereby strengthening the flame, consequently,  $\sigma_{ext}$  increases monotonically with  $Z_{st}$ . The other is a transport effect: at low  $Z_{st}$  reaction is O<sub>2</sub>limited whereas at high  $Z_{st}$  reaction is fuel-limited, leading to a shift in effective  $Le(Le_{eff})$  from that of  $O_2$  (Le<sub>o</sub>) to fuel (Le<sub>f</sub>) as  $Z_{st}$  increases. Hydrogen fuel has much lower  $Le_f$  than any hydrocarbon fuel and vastly different chemistry (we shall show hydrogen does not exhibit the same monotonic increase in  $\sigma_{ext}$  with  $Z_{st}$ , moreover, the very low  $Le_f$ of H<sub>2</sub> in O<sub>2</sub>-N<sub>2</sub> mixtures may lead to diffusivethermal instability (DTI) [15], particularly at high  $Z_{st}$ , which could lead to broken or cellular flames.

While normally associated with premixed flames, DTI does occur in nonpremixed flames at nearextinction conditions when the more-consumed reactant has sufficiently low *Le* [16] and thus might be expected in  $H_2-N_2/O_2-N_2$  edge-flames.

Since no systematic experimental study of nonpremixed hydrogen edge-flames has been conducted, this work examines nonpremixed H<sub>2</sub>-N<sub>2</sub>/O<sub>2</sub>-N<sub>2</sub> edge-flame shapes, propagation rates, stability limits and extinction limits for varying  $Z_{st}$  (adjusted by changing the portion of  $N_2$  on the  $H_2$  side versus the  $O_2$  side), varying  $\sigma$  (by changing flow rates) and varying mixture strengths (by changing N, defined as the moles of inert in the  $H_2:O_2:N_2 = 2:1:N$  mixture formed when fuel and oxidant streams are combined in stoichiometric proportions. For example, for N = 18 (H<sub>2</sub>:O<sub>2</sub>:N<sub>2</sub> = 2:1:18), if all N<sub>2</sub> flows from the fuel side then  $H_2:N_2 = 2:18$  thus the fuel-side mixture is 10% H<sub>2</sub>/90% N<sub>2</sub> and the oxygen-side mixture is 100% O<sub>2</sub>, resulting in  $Z_{st} = 0.9407$ , whereas if all the  $N_2$  flows from the  $O_2$  side then  $H_2:N_2 = 2:0$  thus the fuel-side mixture is 100%  $H_2$  and the oxygen-side mixture is  $O_2:N_2 = 1:18$  $(5.26\% O_2/94.74\% N_2)$ , resulting in  $Z_{st} = 0.0074$ . For these extreme  $Z_{st}$  cases and all intermediate  $Z_{st}$ , combining fuel and oxidant streams in stoichiometric proportions results in premixtures with  $H_2:O_2:N_2 = 2:1:18$ , all having the same premixedflame properties). Following prior work [13,14] a counterflow slot-jet apparatus is employed which provides extensional strain orthogonal to the slot plane yet little convection along the slot length, thus edge-flame propagation speeds in the laboratory frame are essentially equal to the propagation speed relative to the cold unburned gas far ahead of the edge-flame (or behind, for retreating edge-flames).

#### 2. Experimental apparatus, procedures and scaling

The counterflow slot-jet apparatus and procedures were similar to those employed previously [13,14]. Figure S1 shows a schematic of this apparatus. Slot-jet exit velocities ( $U_f$  and  $U_o$  for fuel and oxidizer respectively) were regulated by thermal mass flow controllers to achieve specified global strain rate ( $\sigma = (U_f/d)[1 + (U_o/U_f)(\rho_o/\rho_f)^{1/2}]$ , where

Table 1

Properties of mixtures tested.  $S_L$  is calculated using CHEMKIN with Li et al. [22] kinetics. For all cases  $Le_f \approx 0.33$  and  $Le_o \approx 1.07$ .

Ν	$\rho_u/\rho_b$	$S_L$ (cm/s)	$\sigma_{ext} (1/s) Z_{st} = 0.15$	$\sigma_{ext} (1/s) Z_{st} = 0.5$	$\sigma_{ext} (1/s) Z_{st} = 0.9$
15	3.75	4.40	176.7	230.3	182.9
16	3.62	2.90	111.2	153.8	95.8
17	3.50	1.78	70.0	102.7	50.2
18	3.39	1.04	44.1	68.5	26.3
19	3.29	0.49	27.7	45.8	13.7
20	3.20	0.35	17.4	30.6	7.2



Fig. 1. False-color direct images of edge-flames in mixtures with N = 18. H<sub>2</sub>-N<sub>2</sub> flows from the bottom upwards, O<sub>2</sub>-N<sub>2</sub> from the top downwards. Slot spacing (*d*) = 7.5 mm. White dashed lines indicate the stagnation plane location. (a)  $Z_{st} = 0.6$ ,  $\sigma = 80/s$ , advancing continuous-flame (mode I); (b)  $Z_{st} = 0.4$ ,  $\sigma = 80/s$ , retreating continuous-flame (mode II); (c)  $Z_{st} = 0.9$ ,  $\sigma = 80/s$ , advancing broken-flame (mode II); (d)  $Z_{st} = 0.70$ ,  $\sigma = 110/s$ , stationary broken-flame (mode III) (d)  $U_{st} = 0.33$  [23].

 $(\rho_0, \rho_f)$  are the gas densities at the jet exits and d the jet spacing [17]) and mixtures (H<sub>2</sub>-N<sub>2</sub> and O<sub>2</sub>-N<sub>2</sub>) at values of  $X_o$  and  $X_f$  required to obtain desired values of  $Z_{st}$ . We employed equal jet exit velocities  $(U_f = U_o)$  and since  $\rho_o \approx \rho_f$ , the stagnation plane location was essentially at the midplane between the jets. For all cases shown  $H_2-N_2$  issued from the lower jet and O<sub>2</sub>-N<sub>2</sub> from the upper jet. Reversing these flows had no significant effect on the results, hence buoyancy effects were insignificant for the conditions tested. Honeycomb inserts at the jet exits provided uniform flow across the jets' width (5 mm) and length (130 mm). Nitrogen sheath flows with the same exit velocities as the reactive jets were employed on both sides of both reactant streams to prevent secondary flames. The jets were maintained at room temperature by water-cooling. The edgeflames were recorded with high-speed intensified video using a camera sensitive to near-IR emissions near 823 nm where  $H_2O$  has a weak emission band.  $U_{edge}$  and general flame behavior (broken vs. continuous, burning vs. extinguished) was cross-checked with shadowgraph images and no differences were found but only direct video images are reported here because of the challenges associated with interpreting shadowgraph images. Because the slotjet aspect ratio is finite, there is a slight extensional



Fig. 2. False-color direct images of broken-flames. (a) N = 18,  $Z_{st} = 0.8$ ,  $\sigma = 100/s$ ; (b) N = 18.5,  $Z_{st} = 0.8$ ,  $\sigma = 100/s$ ; (c) N = 19,  $Z_{st} = 0.8$ ,  $\sigma = 100/s$ ; (d) N = 18,  $Z_{st} = 0.7$ ,  $\sigma = 100/s$ ; (e) N = 18,  $Z_{st} = 0.7$ ,  $\sigma = 120/s$ ; (f) N = 17,  $Z_{st} = 0.9$ ,  $\sigma = 20/s$ ; (g) N = 18,  $Z_{st} = 0.85$ ,  $\sigma = 240/s$ ; (h) computed reaction rate contours for a mode IV flame with Le = 0.33 [23]. Images (a) and (f) are advancing (Mode III), all others are stationary (Mode IV).

flow along the slot length which influences  $U_{edge}$  in the laboratory frame slightly; following prior work [13,14], this bias was nullified by interpolating  $U_{edge}$ vs. position along the slot to the jet centerline. In some cases broken flame structures were observed which left nearly stationary flame "islands" behind the leading edge; we defined  $U_{edge}$  as the propagation speed of the leading flame island regardless of the behavior of the trailing flame islands. For retreating edge-flames this issue did not arise because broken flames were never observed. At jet Reynolds numbers  $Re = U_{jet}d/\nu > 500$ , where  $\nu$  is the cold-gas kinematic viscosity, unsteady flames were observed, apparently indicating transition to turbulent flow, consequently, only data for Re < 500 are presented.

For conditions resulting in  $U_{edge} > 0$ , an N<sub>2</sub> jet was used to "erase" the established flame from one end of the slot-jet to nearly the other end and was then retracted, enabling the edge-flame to propagate. For conditions resulting in  $U_{edge} < 0$ , first a mixture with  $U_{edge} > 0$  was introduced, then electrically-heated wires at both slot ends were activated and  $X_o$  and  $X_f$  were slowly reduced to the required values. (The heating wires anchored flames by enhancing local flame temperature and thus reaction rates at the flame ends under conditions which they would retreat without localized heating.) The N<sub>2</sub> jet was momentarily introduced at one slot end to separate the flame from its anchoring hot-wire, thereby triggering an extinction front.

Theories [9–11] predict that for adiabatic nonpremixed edge-flames with  $Le_f = Le_o = 1$ , constant density and low  $\sigma$ ,  $U_{edge}/S_L = 1$ , where  $S_L$  is the laminar burning velocity of a stoichiometric mixture of fuel and oxidizer streams. Fundamentally this scaling is appropriate because for  $Le_f \approx Le_o \approx 1$ the nonpremixed flame temperature is close to that of an adiabatic premixed flame and thus the over-



Fig. 3. Effect of  $Z_{st}$  and  $\sigma$  on scaled edge-flame speeds: (a) N = 17; (b) N = 18. Filled symbols indicate continuous flames (Mode I), open symbols indicate broken flames (Mode III).

all reaction rate  $\omega$  can be estimated as  $S_L^2/\alpha$  [15], where  $\alpha$  is the gas thermal diffusivity. Other theory [18] predicts that when thermal expansion effects are incorporated,  $U_{edge}/S_L$  (in the unburned gas reference frame far upstream of the edge) increases in proportion to  $(\rho_u/\rho_b)^{1/2}$ , where  $\rho_u$  and  $\rho_b$  are the unburned and burned gas densities, respectively. Consistent with these theories, prior experiments [13,14] have shown that values of the scaled propagation speed  $\tilde{U} = (U_{edge}/S_L)(\rho_u/\rho_b)^{-1/2}$ are near unity for nearly-adiabatic edge-flames with  $Le_f \approx Le_o \approx 1$ . In contrast, we found this scaling to be unsatisfactory for H2-O2-N2 edge-flames because the low  $Le_f$  significantly enhances flame temperatures [19] and thus  $\omega$ ; in fact, for the mixtures we tested (Table 1) values of  $S_L$  are so low that they cannot burn as plane premixed flames in our apparatus (Peclet numbers  $Pe \equiv S_L d/\alpha$ are below the typical extinction limit criterion Pe = 40, even for the least-diluted mixture). Park et al. [20] asserted that for weak  $H_2$ - $O_2$ - $N_2$  flames the global extinction strain rate  $\sigma_{ext}$  rather than  $S_L$  may be used to characterize global reaction rates; following this approach  $U_{edge}$  will scale with  $(\alpha \omega)^{1/2} \sim (\alpha \sigma_{ext})^{1/2}$  rather than  $S_L$ , thus, in this work we define  $\tilde{U} = U_{edge} / [\alpha \sigma_{ext} (\rho_u / \rho_b)]^{1/2}$ . We also scale  $\sigma$  with  $\sigma_{ext}$ , leading to a dimensionless strain rate  $\varepsilon \equiv \sigma / \sigma_{ext}$ . We shall show that  $\tilde{U}$  and  $\varepsilon$  are indeed appropriate scalings; we also note they can be used for  $Le_f \approx Le_o \approx 1$  since in that case  $\sigma_{ext} \sim S_L^2 / \alpha \sim \omega$ [15].

While low- $S_L$  stretch-free flames can be extinguished by radiative loss, using the method described in [21], the estimated characteristic radiative loss rate is less than 0.35/s for all mixtures, which is far smaller than the smallest  $\sigma$  examined (16/s). Consequently, radiative transport is negligible compared to convective transport for our flames.

#### 3. Results and discussion

#### 3.1. Flame structures

Figure 1 shows false-color direct images of the four types of structures observed, denoted modes I-IV: (I) advancing continuous-flames, (II) retreating continuous-flames, (III) advancing broken-flames and (IV) stationary broken-flames. (No retreating broken flames were observed under any condition tested.) As expected, for  $Z_{st}$  less (greater) than 0.5, the flame lies on the O<sub>2</sub> (H<sub>2</sub>) side of the stagnation plane. The broken flames are nearly flat, probably because the reaction zone must remain close to the stoichiometric mixing location, unlike low-Le cellular premixed flames which may be highly curved. The broken-flame structures are remarkably similar to those predicted theoretically (Fig. 1e) [23]. For Mode III, individual cells formed behind the leading edge-flame and remained stationary rather than splitting apart after formation of a continuous-flame, as predicted in [23]. A splitting sequence is shown in the Supplemental Data, Fig. S2. While advancing broken-flames (Mode III) would recover after being "erased" by the N<sub>2</sub> jet, stationary broken-flames did not recover after erasure.

Figure 2a–h show false-color direct images of broken-flames at high  $Z_{st}$ . Figure 2a-c show a sequence with increasing N where  $Z_{st}$  and  $\sigma$  are



Fig. 4. Effect of scaled strain rate  $\varepsilon$  on scaled  $U_{edge}$ : (a)  $Z_{st} = 0.15$ ; (b)  $Z_{st} = 0.5$ ; (c)  $Z_{st} = 0.9$ . Filled symbols indicate continuous flames (Modes I, advancing and II, retreating), open symbols indicate broken flames (Modes III, advancing).

held constant; as N increases thus the mixture becomes weaker, the flame "void fraction" increases. Figure 2d,e show the same behavior with increasing  $\sigma$  where N and  $Z_{st}$  are held constant. Figure 2f shows a case with very low  $\sigma$  (near the heat-loss induced limit) with many irregular cells. Finally, Fig. 2g shows a case with very high  $\sigma$ , where the flow as seen by shadowgraph images (not shown) is clearly turbulent ( $Re_{jet} > 500$ ) but the cells remain almost stationary. Stationary broken-flame structures (mode IV) are again remarkably similar to those predicted theoretically (Fig. 2h) [23].

#### 3.2. Propagation rates

Figure 3 shows effects of  $Z_{st}$  and  $\sigma$  on scaled edge-flame speeds  $\tilde{U}$  for two mixture strengths. It should be emphasized that for each plot, every point corresponds to exactly same mixture when fuel and oxidant streams are combined in stoichiometric proportions (thus every point on the plot has the same  $T_{ad}$ ,  $\rho_u / \rho_b$ and  $S_L$  (see Introduction), yet  $\tilde{U}$  (thus  $U_{edge}$ ) varies drastically depending on  $Z_{st}$  and  $\sigma$ . As with nonpremixed hydrocarbon edge-flames [13],  $\tilde{U}$  is maximum at intermediate  $\sigma$  with heat-loss induced extinction at low  $\sigma$  in addition to the high- $\sigma$  extinction limit. Except at very high  $Z_{st}$ ,  $\tilde{U}$  increases with increasing  $Z_{st}$  which is consistent with the notion of decreasing  $Le_{eff}$  as  $Z_{st}$ increases (see Introduction).  $\tilde{U}$  may exceed unity, consistent with the expectation of low Leeff at high  $Z_{st}$ . It is somewhat surprising, however, that upon transition from advancing continuous (filled symbols, Mode I) to broken (open symbols, Mode III) edge-flames there is no significant change in Uvalues. Figure 4 shows effects of  $\varepsilon$  on  $\tilde{U}$  for several dilution levels (N) for three fixed  $Z_{st}$  values. For low (0.15) and intermediate (0.5)  $Z_{st}$ , as  $\varepsilon$  increases  $\tilde{U}$  first decreases slowly then decreases drastically until extinguishment. For high (0.9)  $Z_{st}$ , the trend is quite different, though in all cases have similar maximum scaled  $\tilde{U}$  (1.5~2.5). The reason for this difference is discussed in the following section. In Fig. 4, all curves overlap except for near-extinction and broken-flame cases, demonstrating that  $\varepsilon$  and  $\tilde{U}$  are proper scaling parameters for strain rate and edge-flame speed, respectively.

#### 3.3. Regimes of flame behavior

Figure 5 shows maps of flame behavior in  $Z_{st}$ - $\varepsilon$  space for two fixed N values. Note that on each map, every point produces the same stoichiometric mixture of fuel and oxidant streams. For all  $Z_{st}$  the two extinction limits at high and low  $\varepsilon$  are evident. For  $Z_{st} < 0.6$  no broken flames are observed; only for  $Z_{st} > 0.6$  can Modes I-IV all be observed as  $\varepsilon$  increases. For N = 17,  $Z_{st} > 0.85$  and



Fig. 5. Flame response maps in  $Z_{st}$ - $\varepsilon$  space: (a) N = 17; (b) N = 18. Recall mode designations: (I) advancing continuousflames, (II) retreating continuous-flames, (III) advancing broken-flames and (IV) stationary broken-flames. Vertical dashed lines indicate minimum (pure H<sub>2</sub> vs. O<sub>2</sub>–N<sub>2</sub>) and maximum (H<sub>2</sub>–N<sub>2</sub> vs. pure O<sub>2</sub>) values of  $Z_{st}$  attainable. Dashed curves indicate transition to turbulent structures.



Fig. 6. Flame response maps in N- $\varepsilon$  space: (a)  $Z_{st} = 0.3$ ; (b)  $Z_{st} = 0.8$ . Dashed curves indicate transition to turbulent structures.

N = 18,  $Z_{st} > 0.78$  only broken flames are observed, indicating dominance of low-*Le* DTI. For N = 17,  $Z_{st} < 0.08$  and N = 18,  $Z_{st} < 0.15$ , no combustion occurs at any  $\varepsilon$ . Note that for both N = 17 and N = 18, at  $Z_{st} \approx 0.58$ ,  $\varepsilon \approx 1.3$ , a rather remarkable bifurcation occurs - moving radially outward from these points small distances in  $Z_{st}$ - $\varepsilon$  space, depending on the direction one may encounter any of 4 of the observed flame structures or extinction! Figure 6 shows maps of flame behavior in N- $\varepsilon$  space for two fixed  $Z_{st}$  values. Again, the low- $\varepsilon$  and high- $\varepsilon$  extinction limits are evident and only for higher  $Z_{st}$  are broken flames observed.

## 3.4. Computations of $\sigma_{ext}$ and comparison with experiment

Previous work [14] on  $Z_{st}$  influences on nonpremixed edge-flames in hydrocarbon fuels demonstrated that many aspects of multidimensional



Fig. 7. Comparison of measured (solid lines and data points) and computed (dashed curves) values of extinction strain rate ( $\sigma_{ext}$ ) for varying  $Z_{st}$  with N = 17 and N = 18.

edge-flame behavior could be interpreted in terms of the readily-computed  $\sigma_{ext}$ . Such computations (Fig. 7) for  $H_2-N_2/O_2-N_2$  flames were performed with CHEMKIN using the same gap (d) as the experiments. Measured values of  $\sigma_{ext}$  are similar to but mostly higher than the computed values. For  $Z_{st} > 0.6$ , two sets of experimental data are shown, for "extinction" of continuous-flames (transition to broken-flames) and for extinction of broken-flames. Remarkably, computed values of  $\sigma_{ext}$  correspond well with transition to brokenflames observed in experiments. Values of  $\sigma$  at extinction of broken-flames increase rapidly with increasing  $Z_{st}$  and at  $Z_{st} > 0.8$  could not be observed because of transition to turbulent flow (see Fig. 5). Figure 7 again illustrates the bifurcation in flame behavior near  $Z_{st} \approx 0.6$ . This bifurcation in the experiment data is due to transition from one-dimensional continuous to multi-dimensional broken-flame structures, whereas the computations are strictly one-dimensional and cannot exhibit this bifurcation. Ref. [23] reports computations employing schematic single-step chemistry and predicts that that broken-flames exist only when continuous flames cannot, which is precisely what is seen experimentally. It is unclear whether the peak in  $\sigma_{ext}$  that occurs at  $Z_{st} \approx 0.6$  is fundamentally related to this bifurcation or is purely coincidental.

As discussed in the Introduction,  $Z_{st}$  influences on nonpremixed edge-flames in hydrocarbon fuels are affected by both Lewis numbers of fuel and oxygen and the unique chemistry of these fuels [14]. To separate *Le* and chemistry effects for hydrogen, computations of  $\sigma_{ext}$  were performed with Lennard–Jones parameters and molecular masses



Fig. 8. Computed effects of  $Z_{st}$  on  $\sigma_{ext}$  for H<sub>2</sub>– N<sub>2</sub>/O<sub>2</sub>–N<sub>2</sub> mixtures with standard transport and artificial transport causing  $Le_f = Le_o \approx 1$ , along with results for CH<sub>4</sub>–N<sub>2</sub>/O<sub>2</sub>–N<sub>2</sub> ( $Le_f = Le_o \approx 1$ ) and i-CH<sub>4</sub>–N<sub>2</sub>/O<sub>2</sub>–N<sub>2</sub> ( $Le_f > Le_o \approx 1$ ) mixtures.

of H<sub>2</sub> and H set to those of O<sub>2</sub> and O, respectively, in order to force the Lewis numbers of H<sub>2</sub> and O<sub>2</sub> to be equal (and both nearly unity). Comparisons of  $H_2-N_2/O_2-N_2$  flames with standard transport and artificial transport causing  $Le_f = Le_o \approx 1$  are shown in Fig. 8. With  $Le_f = Le_o \approx 1$ ,  $\sigma_{ext}$  decreases by a factor of 4,5 and becomes almost independent of  $Z_{st}$ . By comparing these data with CH<sub>4</sub>–N<sub>2</sub>/O<sub>2</sub>–N<sub>2</sub> flames which also have  $Le_f = Le_o \approx 1$  but the unique hydrocarbon chemistry that leads to  $\sigma_{ext}$  monotonically increasing with  $Z_{st}$ , (also shown in Fig. 8) it can be concluded that (1) the low Le of  $H_2$ causes more robust flames with higher  $\sigma_{ext}$  and (2) Lef effects, not chemistry effects, are responsible for the non-monotonic effect of  $Z_{st}$  on  $\sigma_{ext}$  in  $H_2-N_2/O_2-N_2$  flames. Figure 8 also shows results for i-C<sub>4</sub>H<sub>10</sub>-N<sub>2</sub>/O<sub>2</sub>-N<sub>2</sub> mixtures where (in contrast to  $H_2-N_2/O_2-N_2$ ) the high  $Le_f$  of  $i-C_4H_{10}$  causes a <u>decrease</u> in  $Le_{eff}$  (from  $Le_f$  to  $Le_o$ ) as  $Z_{st}$  decreases, and thus exhibits non-monotonic behavior in contrast to the monotonic trend with  $CH_4-N_2/O_2-N_2$ mixtures.

#### 4. Conclusions

The effects of stoichiometric mixture fraction  $(Z_{st})$  on flame structure and propagation rates in nonpremixed edge-flames  $H_2-N_2/O_2-N_2$  mixtures were studied using a counterflow slot-jet apparatus. Both advancing and retreating edge-flames were characterized in terms of scaled edge-flame speed  $(\tilde{U})$  and strain rate ( $\varepsilon$ ). Due to the low Lewis number of  $H_2$ , scalings of  $\tilde{U}$  and  $\varepsilon$  based on burning

velocity  $S_L$  were inappropriate, instead scalings based on extinction strain rate  $\sigma_{ext}$  were successfully employed. Both high- $\varepsilon$  residence-time limit extinction limits and low- $\varepsilon$  heat loss-induced extinction limits were observed. As in prior work on hydrocarbon edge-flames [14],  $\tilde{U}$  is neither independent of  $Z_{st}$  nor symmetric with respect to  $Z_{st} = 0.5$ , but for very different reasons. For hydrocarbons a chemical effect leads to  $\tilde{U}$  monotonically increasing with  $Z_{st}$ , but this effect is absent in H<sub>2</sub>-N<sub>2</sub>/O<sub>2</sub>-N<sub>2</sub> flames. Instead, the low Lewis number of H<sub>2</sub> and the shift from oxygen-limited to fuel-limited reaction as  $Z_{st}$  increases causes the observed behavior. Above  $Z_{st} \approx 0.6$  a bifurcation leads to the possibility of broken (in many cases still nearly planar) edge-flames which may be advancing or stationary, but were not observed for retreating edges. The broken flames continue to survive under conditions (high strain or heat loss) where continuous plane edge-flames cannot. These results indicate that the behavior of turbulent nonpremixed hydrogen flames and the effects of stoichiometric mixture faction thereupon should not be anticipated based on extrapolation of Lewis number and strain rate effects for flames in hydrocarbons or other fuels.

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#### Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10. 1016/j.proci.2018.05.010.

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Professor Zhenghong Zhou University of Southern California

Dear Prof. Zhou:

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