

## DIFFUSIVE-THERMAL INSTABILITY AND FLAME EXTINCTION IN NONPREMIXED COMBUSTION

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A systematic study of cellular instabilities in nonpremixed flames is performed by examining shapes, heights, and extinction conditions of over- and under-ventilated slot-jet flames. A variety of fuels and diluents are used to vary Lewis numbers of fuel and oxidizer. Only nonpremixed flames near extinction having sufficiently low effective  $Le$  were found to exhibit cellular instability. It is proposed that near-extinction conditions are required so that reactant intermixing occurs before combustion takes place and that the effective  $Le$  is that of the more completely consumed reactant according to Liñán's criterion for fuel or oxidizer leakage in diffusion flames, which must be  $\leq 0.8$  for cellular instability. It is shown that all known observations of cellular nonpremixed flames can be described this way. Mechanisms proposed in previous work, such as preferential diffusion of light reactants and stoichiometric considerations, are disputed. Flame-height and extinction limit scalings suggest that flame height may be controlled by either buoyancy or jet momentum and extinction by blowoff or heat-loss mechanisms. The characteristics of the observed cellular structures are consistent with the proposed mechanisms. Temperatures at cellular fronts are found to be higher than the calculated equilibrium values, but extinction limits appear not to be extended by diffusive-thermal effects as much as in premixed flames. It is concluded that diffusive-thermal instability of nonpremixed flames occurs in a manner similar to that of premixed flames, regardless of the mechanism leading to reactant intermixing.

### Introduction

Premixed gas flames are known<sup>1-6</sup> to exhibit cellular structures due to imbalances between heat and/or mass diffusion at the flame front. Cellularity is often thought<sup>1-3</sup> to result from diffusive-thermal instabilities which occur when  $Le < 1-2/\theta$ , where  $Le$  is the Lewis number, defined as the ratio of bulk thermal diffusivity to the mass diffusivity of the deficient reactant and  $\theta$  is the nondimensional activation energy of the heat release reaction. For near-stoichiometric mixtures, a weighted  $Le$  ( $Le_{eff}$ ) depending on both reactants can be defined.<sup>4</sup> Others<sup>7,8</sup> argue that cellular instabilities are due to "preferential diffusion" of light reactants: if the deficient reactant diffuses more rapidly than the abundant one, the planar front is unstable.

While experimental evidence of cellular instability of premixed flames abound in the literature,<sup>5,9</sup> much less attention has been paid to cellular flames in *nonpremixed* combustion.<sup>10</sup> Garside and Jackson<sup>11</sup> reported polyhedral structures in  $H_2$  jet diffusion flames when  $H_2$  was sufficiently diluted. The mechanism of formation of polyhedral structures was proposed to be related to the high thermal diffusion rate of  $H_2$ . High flow velocities (causing low

Damköhler number [ $Da \equiv \{\text{chemical reaction rate}\} / \{\text{flow or mixing rate}\}$ ]) were necessary to observe cellularity. Dongworth and Melvin<sup>12</sup> reported cellular flames in diluted  $H_2$ -air mixtures on splitter-plate burners and attributed this to stoichiometric considerations: near extinction, fuel was thought to leak through flame base into the oxidizer stream and forms a composite lean premixed/diffusion flame. How the lean conditions were responsible for cellularity was not clear, although preferential diffusion effects were disputed. "Striped" flames at near-extinction conditions were observed<sup>13</sup> in  $H_2$ -air flames on Tsuji burners when  $H_2$  was heavily diluted with  $N_2$  or Ar. Dilution was argued to move the flame to the fuel side of the stoichiometrically mixed surface, or "mixing layer;" cellularity was attributed to preferential diffusion of  $H_2$  relative to  $O_2$ . When  $H_2$  was diluted with He, no cellularity was observed; they argued that the flame moved to the oxidizer side in this case. Recent experiments<sup>14</sup> on downward flame spread over thin paper strips near extinction (low  $Da$ ) revealed cellular structures when the oxidant Lewis number ( $Le_O$ ) was sufficiently low. It was hypothesized that low  $Da$  created a partially premixed region, then cellularity

occurred at low  $Le_O$  in a manner similar to that in premixed flames.

In all these investigations, nonpremixed cellular flames were reported only near extinction and were always attributed to preferential diffusion effects. We now discuss how these effects influence nonpremixed flame structure.

Liñán<sup>15</sup> showed that in nonpremixed flames, intermixing of fuel and oxidant occurs at low  $Da$  because when mixing outpaces chemical reaction, one or both reactants partially "leak" through the chemical reaction zone without burning completely. Reaction occurs at temperatures lower than the equilibrium (infinite  $Da$ ) diffusion-flame temperature<sup>15</sup> ( $T_{eq}$ ). Either a "diffusion flame" structure, in which a portion of both reactants of order  $1/\theta$  leaks through the reaction zone, or a "premixed-burning" structure, in which a portion of one reactant of order unity leaks, may exist. As  $Da$  is further decreased (by dilution to reduce  $T_{eq}$  and thus reaction rate, or by increasing the local flow or mixing rate), leakage eventually causes extinction.

According to Liñán, in either the diffusion-flame or premixed-burning regime, which reactant is more completely consumed depends on a parameter  $\alpha + 2\beta$ , where  $\alpha = Y_{O,-\infty}/\nu Y_{F,+\infty}$  is the ratio of oxidizer mass fraction in the oxidizer stream to fuel mass fraction in the fuel stream normalized by the stoichiometric oxidizer-to-fuel mass ratio ( $\nu$ ) and  $\beta$  is the nondimensional difference between temperatures of oxidizer and fuel streams. When  $2\beta < 1$ , the oxidizer/fuel is more completely consumed if  $\alpha + 2\beta$  is less than/greater than unity. When  $|\alpha + 2\beta - 1|$  is sufficiently large, one reactant is almost completely consumed and the structure of the nonpremixed flame is similar to a premixed flame with heat loss in the burned gases (since in the nonpremixed flame with  $2\beta < 1$  the temperature drops on both sides of the reaction zone.) While Liñán assumed  $Le = 1$ , these results are probably qualitatively similar when  $Le \neq 1$ , though we know of no formal analysis.

Since reactant intermixing probably occurs in nonpremixed flames near extinction, the diffusive-thermal mechanism, well known in premixed flames, could potentially cause cellular non-premixed flames. Which reactant is more completely consumed (hereafter called the "deficient reactant") would be critical, since cellular instabilities in premixed flames depend on the diffusive properties of the deficient reactant. Thus we expect  $Le_{eff} = Le_F$  if  $\alpha + 2\beta > 1$  and  $Le_{eff} = Le_O$  if  $\alpha + 2\beta < 1$ , with transition over a region near  $\alpha + 2\beta \approx 1$ .

Our objective is to systematically study the mechanisms of cellular instabilities of nonpremixed flames and their influence on flame extinguishment. Toward this goal, we study (1) various fuels and diluents to test  $Le$  effects, (2) various stoichiometries to test the effects of preferential diffusion and the

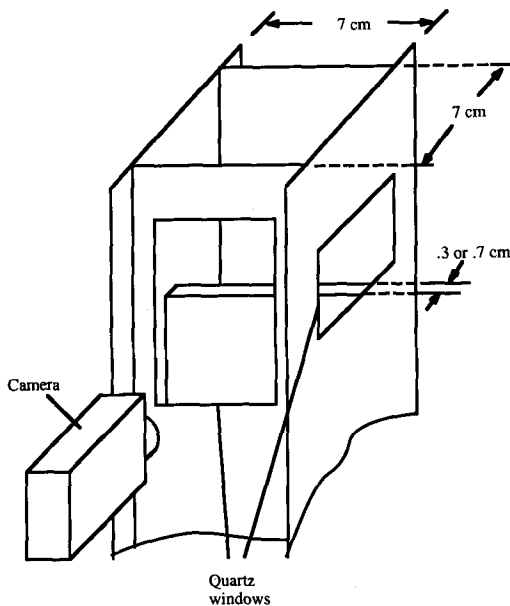


FIG. 1. Schematic of the experimental apparatus.

parameter  $\alpha + 2\beta$ , and (3) dilution and flow velocity effects to test  $Da$  influences. A two-dimensional Wolfhard-Parker burner was employed because opposed-jet configurations induce mean strain which inhibits cellularity (at least in premixed flames)<sup>16</sup> and because axisymmetric Burke-Schumann burners induce curvature effects and azimuthal restrictions on instabilities. Wolfhard-Parker burners also enable observation of a large number of cells in a row.

### Experimental Apparatus and Approach

Slot burners with widths of 7 mm and 3 mm widths (Fig. 1) were employed. Honeycomb flow straighteners in both inner and outer streams minimized transverse velocity gradients. Velocities of inner (fuel + diluent) and outer (oxygen + same diluent) streams were matched to minimize shear-flow instabilities. All flows were laminar and initial temperatures of both streams were 298 K ( $\beta = 0$ ). Computer-commanded commercial mass-flow controllers were employed to ensure consistent flow velocities and compositions. An infrared-sensitive video camera aided visualization of  $H_2$ - $O_2$  flames.

Table I shows the fuels and diluents we employed. The oxidizer was always  $O_2$ . Transport properties shown in Table I were calculated using standard techniques.<sup>17</sup> Compositions employed in these calculations corresponded to experimentally-observed extinction conditions and consisted of stoi-

TABLE I  
Summary of Transport Coefficients, Relevant Parameters, and Flame Behavior Near Extinction

Fuel	Inert	$Le_F$	$Le_O$	$D_F/D_O$	$\phi$	$\alpha$	Flame Behavior near extinction
CH <sub>4</sub>	He	1.83	1.64	0.89	2.00	0.22	noncellular
CH <sub>4</sub>	N <sub>2</sub>	0.96	1.01	1.06	0.50	0.41	noncellular
					2.00	0.30	noncellular
					4.00	0.142	noncellular
					6.67	0.051	noncellular
					10.0	0.047	noncellular
CH <sub>4</sub>	CO <sub>2</sub>	0.73	0.80	1.10	2.00	0.27	marginally cellular
CH <sub>4</sub>	SF <sub>6</sub>	0.39	0.48	1.25	0.50	3.3	cellular
					0.67	1.90	cellular
					2.00	0.31	cellular
					4.00	0.089	cellular
C <sub>3</sub> H <sub>8</sub>	N <sub>2</sub>	1.79	0.99	0.56	2.00	0.51	noncellular
C <sub>3</sub> H <sub>8</sub>	CO <sub>2</sub>	1.39	0.80	0.57	2.00	0.55	noncellular
C <sub>3</sub> H <sub>8</sub>	SF <sub>6</sub>	0.70	0.53	0.76	2.00	0.71	cellular
					10.0	0.071	cellular
H <sub>2</sub>	N <sub>2</sub>	0.35	1.00	2.9	0.25	3.4	cellular
					0.50	1.67	cellular
					1.00	0.80	cellular
					1.50	0.51	cellular near heat loss limit
					2.00	0.35	noncellular
H <sub>2</sub>	SF <sub>6</sub>	0.21	0.36	1.73	0.50	1.76	cellular
					2.00	0.55	cellular
(H <sub>2</sub> + CO <sub>2</sub> )-Air		0.34	0.88	2.6	2.70	1.84	cellular (Ref. 11)
H <sub>2</sub>	N <sub>2</sub>	0.35	1.00	2.9	0.36	2.9	cellular (Ref. 12)
H <sub>2</sub>	N <sub>2</sub>	0.34	1.02	2.9	1.09	3.24	cellular (Ref. 13)
H <sub>2</sub>	Ar	0.33	1.10	3.4	0.77	5.5	cellular (Ref. 13)
H <sub>2</sub>	He	1.02	2.1	2.0	1.13	1.36	noncellular (Ref. 13)
paper-(O <sub>2</sub> + SF <sub>6</sub> )		?	0.53	?	5.1	0.031	cellular (Ref. 14, $p = 1$ atm.)
paper-(O <sub>2</sub> + CO <sub>2</sub> )		?	0.75	?	8.6	0.045	cellular (Ref. 14, $p = 2.3$ atm.)

chiometrically mixed reactants with their carrier diluents. In a given experiment, the molar ratio of fuel in the inner stream to O<sub>2</sub> in the outer stream was held fixed as velocity at burner exit,  $V(0)$ , or dilution was varied. This molar ratio, divided by the stoichiometric molar ratio, is denoted by  $\phi$ , which is a known function of  $\nu$ ,  $Y_{O,-\infty}$ ,  $Y_{F,+\infty}$ , and the component molecular weights. In the limit  $Y_{O,-\infty} \ll 1$  and  $Y_{F,+\infty} \ll 1$ , or when all molecular weights are equal, then  $\alpha = 1/\phi$ . Extinction, corresponding to flame liftoff, was obtained by increasing  $V(0)$  or dilution.

### Results

Figure 2 shows three representative extinction limit curves. At the highest  $V(0)$ , limiting reactant concentrations increase with  $V(0)$ , while at the lowest  $V(0)$  they decrease with increasing velocity. Theory<sup>18,19</sup> suggests that the high  $V(0)$  limit may be a low- $Da$  blowoff limit while the low  $V(0)$  limit

may result from heat losses. At intermediate  $V(0)$ , limiting compositions are frequently almost independent of  $V(0)$ .

Figure 3 shows flame height ( $y_F$ ) measurements, for mixtures 5 mole percent O<sub>2</sub> away from extinction, corresponding to the conditions shown in Fig. 2.  $y_F$  scales with  $V(0)^{1.3 \pm 0.1}$  at low  $V(0)$  and  $V(0)^{1.0 \pm 0.1}$  at high  $V(0)$ , though the transition  $V(0)$  varies and apparently was sometimes beyond the capabilities of our flow controllers. The  $V(0)$  at transition corresponds closely to the  $V(0)$  at transition from nearly-vertical extinction limit behavior to positively-sloped limit behavior seen in Fig. 2. This suggests an additional change in the extinction mechanism beyond the blowoff to heat-loss transition just proposed.

Far from extinction, no mixtures exhibited cellular flames. Cellular flames were only observed close to extinction in mixtures with sufficiently low  $Le_{eff}$  (Table I). In all but one case, cells were observed at all values of  $\phi$  studied. In H<sub>2</sub>-O<sub>2</sub>-N<sub>2</sub> mixtures, cells were observed only when the fuel stream

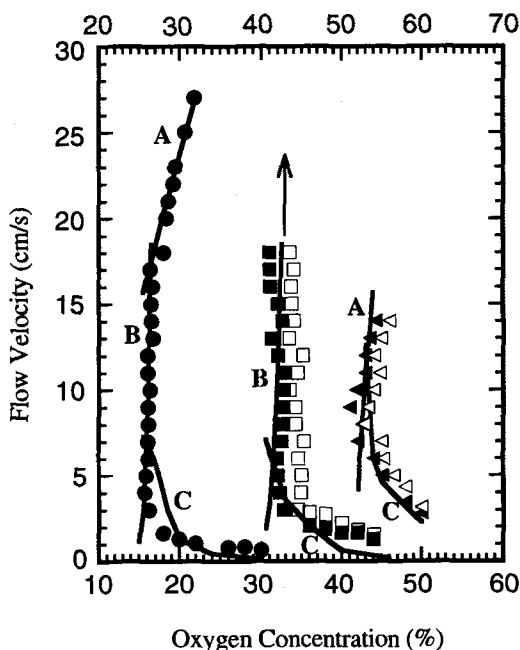


FIG. 2. Mole percent  $O_2$  in the outer stream corresponding to the onset of extinction and cellular initiation. Circles:  $N_2$ -diluted  $C_3H_8$ - $O_2$  diffusion flames (slot width = 7 mm,  $\phi = 2$ ). Squares:  $SF_6$ -diluted  $CH_4$ - $O_2$  diffusion flames (slot width = 7 mm,  $\phi = 2$ ). Triangles:  $SF_6$ -diluted  $CH_4$ - $O_2$  diffusion flames (slot width = 3 mm,  $\phi = 2$ ). Solid symbols represent extinction conditions; empty symbols represent conditions at the onset of cellularity. Note squares use upper abscissa. Curves represent predictions of scaling models (see text). A:  $(RR)/V(0) = \text{constant}$  (momentum-controlled blowoff limit); B:  $(RR)V(0)^{-1/3} = \text{constant}$  (buoyancy-controlled blowoff limit); C:  $(RR)V(0)^{4/3} = \text{constant}$  (buoyancy-controlled heat-loss limit).

was sufficiently diluted (Fig. 4), corresponding to low  $\phi$  or high  $\alpha$ . For one near-critical case ( $\phi = 1.50$ ) cellularity occurred only at low  $V(0)$ .

For brevity, only representative cases are shown in Figs. 2-4, but similar extinction limit, flame height, and cellular structure characteristics were observed in all other cases tested (Table I).

Fig. 5 shows photographs of cellular fronts which reveal different structures depending on  $V(0)$ . At high  $V(0)$ , cellular structures occurred only at the flame base (Fig. 5a). At intermediate  $V(0)$  (Fig. 5b), cellular structure occurred all along the flame length. In both cases blowoff occurred following cellular flame lift-off. While lifting off, the cellular structure was maintained. At low  $V(0)$ , flame lengths were short and the cellular instability broke the flame into

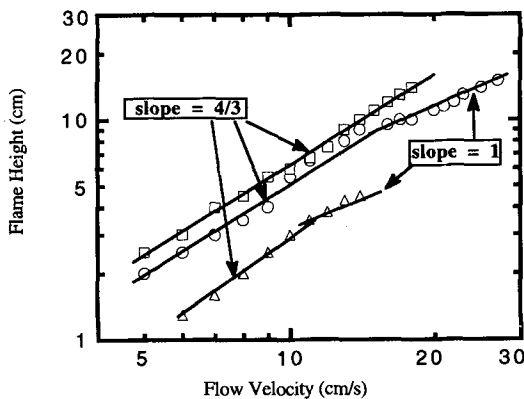


FIG. 3. Heights of flames for the same mixtures and slot widths as in Figure 2 at 5 mole percent  $O_2$  above the limiting values. Symbols same as in Figure 2.

patches (Fig. 5c); extinction occurred when the cells disappeared. Underventilated flames near extinction also exhibited cellularity when  $Le$  was sufficiently low (Fig. 5d). The cellular structure was qualitatively similar to overventilated cases. For all types of structures cells were frequently unsteady, especially very near extinction limits.

Maximum temperatures found in cellular fronts of  $\phi = 2.0$   $CH_4$ - $O_2$ - $SF_6$  and  $\phi = 1.0$   $H_2$ - $O_2$ - $N_2$  flames near extinction were located and measured using fine-wire thermocouples. The radiation-corrected values were  $1,900 \pm 100$  K and  $1,450 \pm 100$  K, while calculated equilibrium temperatures ( $T_{eq}$ ) were 1,490 K and 1,120 K, respec-

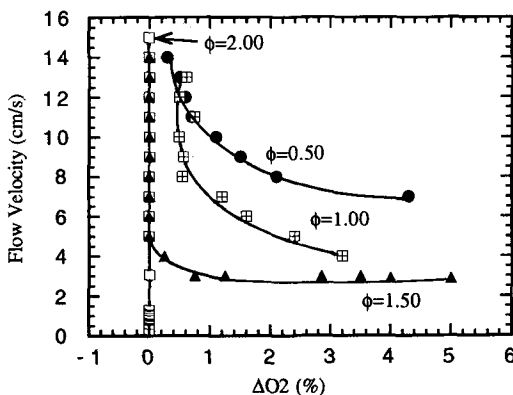


FIG. 4. Difference between mole percent  $O_2$  at the onset of cellularity and mole percent  $O_2$  at extinction for  $N_2$ -diluted  $H_2$ - $O_2$  mixtures at various  $\phi$  and flow velocities. Where the difference is zero, no cells were observed.

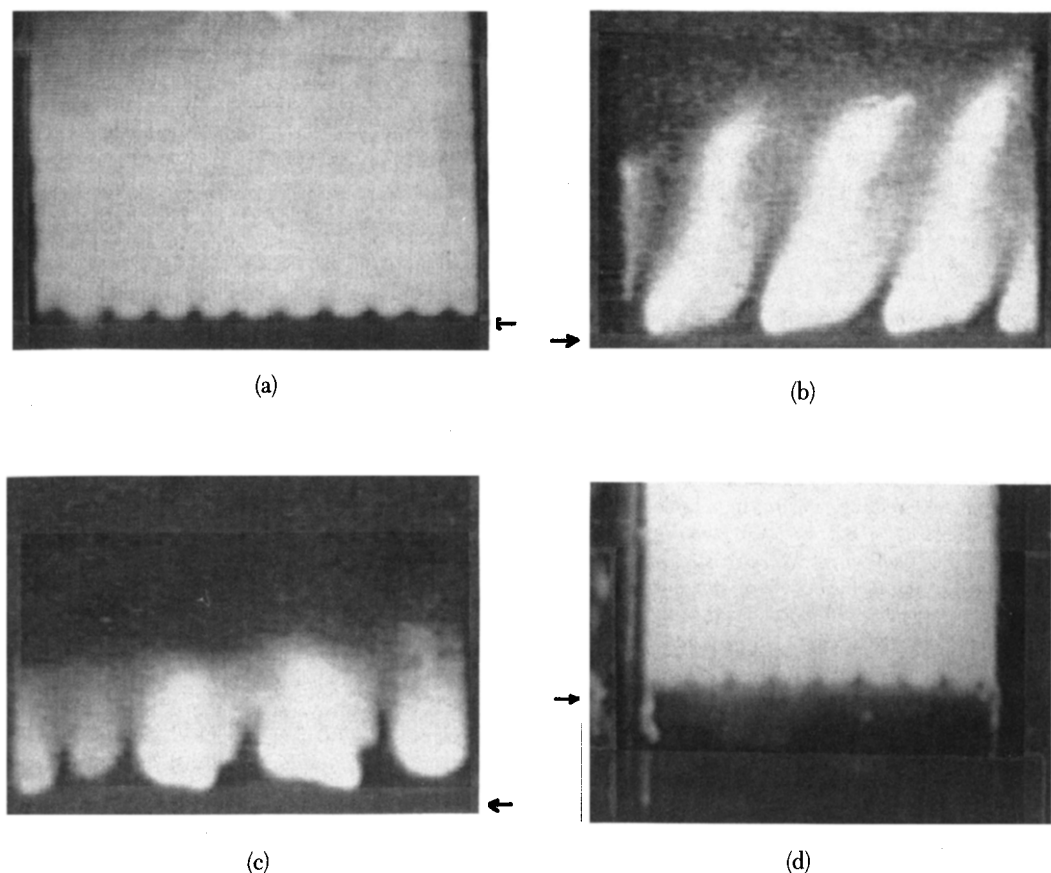


FIG. 5. Direct photographs of cellular nonpremixed flame structures. Arrows indicate elevation of the burner rim. (a) overventilated  $\text{SF}_6$ -diluted  $\text{CH}_4$ - $\text{O}_2$  flame near the momentum-dominated blowoff limit. (b) overventilated  $\text{SF}_6$ -diluted  $\text{CH}_4$ - $\text{O}_2$  flame near the buoyancy-dominated blowoff limit. (c) overventilated  $\text{SF}_6$ -diluted  $\text{CH}_4$ - $\text{O}_2$  flame near the low-velocity heat-loss limit. (d) underventilated  $\text{SF}_6$ -diluted  $\text{C}_3\text{H}_8$ - $\text{O}_2$  flame near the momentum-dominated blowoff limit.

tively. Values of  $T_{eq}$  were calculated by determining the composition at the flame front in the usual way,<sup>10,15</sup> and using a chemical equilibrium program to account for variable specific heat and dissociation effects. By comparison, the maximum measured flame temperature for the same  $\phi = 2.0$   $\text{CH}_4$ - $\text{O}_2$ - $\text{SF}_6$  mixture under *noncellular* conditions (obtained by varying  $V(0)$ , see Fig. 2) was  $2,000 \pm 100$  K. These results suggest that, as in premixed flames,<sup>1-3</sup> Lewis number effects increase flame temperatures above the equilibrium,  $Le = 1$  predictions. In the nonpremixed case, however, it seems that cellularity does not provide an increase in maximum flame temperature, but rather the increase occurs via one-dimensional Lewis number effects, probably in the manner described by Law and Chung.<sup>20</sup>

## Discussion

### Flame Height:

Our flame-height scalings are consistent with Roper's<sup>21</sup> predictions for buoyancy-controlled and momentum-controlled regimes. At high  $V(0)$ , the residence time is proportional to  $y_F/V(0)$  and the transverse diffusion time to  $d(0)^2/D$ , where  $d(0)$  is the slot width and  $D$  is the fuel diffusivity. Equating residence and diffusion times yields  $y_F \sim V(0)d(0)^2/D$ . At low  $V(0)$ , the flow velocity  $V(y)$  depends on elevation ( $y$ ) because of buoyancy according to  $V(y) \sim (gy)^{1/2}$ , where  $g$  is the gravitational acceleration. Thus the residence time  $\sim (y_F/g)^{1/2}$ . Continuity dictates that the stream tube width  $d(y) \sim d(0)V(0)/V(y)$ . Then since diffusion time is

$\sim d(y)^2/D$ ,  $y_F \sim [V(0)^4 d(0)^4 / g D^2]^{1/3}$ . Fig. 3 shows that transition from buoyancy-controlled to momentum-controlled flame height did not seem to occur in one 7 mm slot-width case up to the largest  $V(0)$  our flow controllers could provide, whereas the 3-mm slot did show transition. This is consistent with the notion that buoyancy is more important in larger flames.

#### Extinction Behavior:

Velocities where transitions from apparently buoyancy-controlled to apparently momentum-controlled flame height occur (Fig. 3) correspond approximately to those of extinction mechanism transitions (Fig. 2). Thus, the blow-off limits of the flames studied may consist of momentum-dominated and buoyancy-dominated mechanisms. In the momentum-dominated regime, mixing time scales as  $d(0)/V(0)$ , hence  $Da \sim (RR)d(0)/V(0)$ , where  $RR$  is the characteristic reaction rate  $\sim \exp(-\theta)$ .  $Da$  would be lowest at the flame base, where concentration gradients are largest. In the buoyancy-dominated regime,  $Da(y) \sim (RR)d(y)/V(y)$ ; this leads to  $Da(y) \sim (RR)V(0)d(0)/gy$ , which decreases with increasing  $y$ ; at the highest relevant  $y$ , namely  $y_F$ ,  $Da \sim (RR)[D^2/V(0)d(0)g^2]^{1/3}$ . Thus  $Da$  would be low near the flame tip and also low at the flame base (for the same reasons as before). If  $Da$  at extinction ( $Da_{ext}$ ) remains constant when  $V(0)$  is changed, these scalings indicate that  $V(0)$  will affect  $RR$  (thus  $T_f$  and limit oxygen concentration) much more in the momentum-controlled regime than in the buoyancy-controlled regime, consistent with our findings (Fig. 2).

At the lowest  $V(0)$ , heat-loss may be the dominant extinction mechanism.<sup>18,19</sup> Theory<sup>10</sup> predicts that the ratio of heat-loss rate ( $Q_{loss}$ ) to heat-generation rate ( $Q_{gen}$ ) is a constant ( $\sim 1/\theta$ ) at the limit. We expect  $Q_{gen} \sim \Sigma(RR)\rho C_p \Delta T$  and  $Q_{loss} \sim kA\Delta T/d(0)$ , where  $\rho$  = density,  $\Delta T$  = temperature rise due to combustion,  $k$  = thermal conductivity,  $\Sigma$  = flame volume  $\sim y_F d(0)L$ ,  $A$  = slot burner area =  $d(0)L$ , and  $L$  = slot burner length. Thus we propose  $Q_{gen}/Q_{loss} \sim \{V(0)^4 d(0)^7 / g D^5\}^{1/3} Le^{-1}(RR) \approx$  constant for buoyancy-controlled flame heights and  $Q_{gen}/Q_{loss} \sim \{V(0)d(0)^3/D^2\} Le(RR) \approx$  constant for momentum-controlled flame heights. Unlike the blow-off limits, these requirements imply that the reactant concentrations should decrease with increasing  $V(0)$ , consistent with our findings at low  $V(0)$  (Fig. 2).

These predictions were quantitatively tested by inferring  $RR$  from the calculated  $T_{eq}$  at the limit concentrations (see Introduction) using a representative  $E_a \approx 40$  kcal/mole for hydrocarbon oxidation<sup>7</sup> and holding  $D$ ,  $d(0)$ , and  $g$  constant. The proper flame-height scaling for each extinction was determined from the results of Fig. 3. Since numerical

values of  $Da_{ext}$  are not predicted by our scaling analysis, we "anchor" each predicted extinction curve to experimental values at one point. Fig. 2 shows that the model predictions are generally consistent with experiments, though only modestly so at the low-velocity limits.

#### Mechanisms of Cellular Instability:

Table I shows that all cellular flames have  $Le_{eff}$  sufficiently smaller than unity. Marginal cellularity seems to correspond to  $Le_{eff} \approx 0.8$  ( $\text{CH}_4\text{—O}_2\text{—CO}_2$  flames). The  $\text{H}_2\text{—O}_2\text{—N}_2$  flames have  $Le_F \approx 0.35$  and  $Le_O \sim 1.0$ , thus cellularity is expected when the fuel is the deficient reactant ( $\alpha > 1$ ) but not under oxygen-deficient conditions ( $\alpha < 1$ ). Since  $Le_{eff}$  is a specially weighted average<sup>4</sup> of  $Le_f$  and  $Le_O$ , the threshold is expected to occur at  $\alpha < 1$ , consistent with the experimental results. Perhaps coincidentally, the threshold  $\phi$  for cellularity ( $\approx 1.5$ ) is nearly the same as that found in premixed flames.<sup>5</sup> Also, near the threshold  $\phi$ , cells were found only at low  $V(0)$ , which we have shown is likely a heat-loss extinction. This is consistent with premixed-flame theory<sup>6</sup> which predicts that heat-loss promotes cellularity by raising the critical  $Le_{eff}$ .

Preferential diffusion models<sup>7,8</sup> are inconsistent with our observations because near-extinction  $\text{CH}_4\text{—O}_2\text{—SF}_6$  and  $\text{H}_2\text{—O}_2\text{—SF}_6$  mixtures were cellular at all  $\alpha$ , that is, for both fuel-deficient and oxygen-deficient conditions. For these cases the deficient reactant changes from more-diffusive fuel to less-diffusive  $\text{O}_2$ , hence preferential diffusion models predict transition from cellular to noncellular behavior. Similarly, no cellularity was observed in  $\text{CH}_4\text{—O}_2\text{—He}$  flames with  $\alpha < 1$  (oxygen-deficient burning) where  $D_O/D_F > 1$  and  $Le_{eff} = Le_O = 1.64$ . This is consistent with diffusive-thermal but not preferential-diffusion hypotheses. Additionally, similar results were found for underventilated flames (Fig. 5d), indicating that stoichiometric variations alone are not the predominant factor.

While cellularity was observed for low- $Le$  mixtures regardless of the apparent extinction and flame-height mechanisms, these mechanisms affect the visible flame structures. In the momentum-dominated liftoff regime (Fig. 5a)  $Da$  is lowest, thus intermixing is greatest, at the flame base, hence cellular structures would be most prevalent there. In the buoyancy-dominated liftoff regime,  $Da$  may be low both at the flame base and the flame tip (according to our scaling analysis), hence cells appear in both regions even when the flame is many slot-widths tall. In the heat-loss regimes (Fig. 5c), flame-heights are small and according to our scaling analysis losses are significant throughout the flame volume, thus cells appear throughout the flame. These observations concerning cellular structures are

consistent with the extinction-limit scaling analyses discussed above.

#### *Discussions of Previous Work:*

Some previous work on cellular instability of diffusion flames (also listed in Table I) is now discussed in the context of low  $Da$ , low  $Le$ , and the role of  $Le_{eff}$ .

Garside and Jackson's<sup>11</sup> polyhedral diluted hydrogen flame had  $\alpha = 1.84$ . Thus  $H_2$  is the more completely consumed reactant, thus  $Le_{eff} \approx 0.34$ . Accordingly, our criterion predicts cellular instability.

The role of dilution in Dongworth and Melvin's  $H_2$  flames<sup>12</sup> is also consistent with our model. Unlike their explanation, we believe  $Le$ , not stoichiometric considerations alone, govern these phenomena.

Ishizuka and Tsuji's<sup>13</sup>  $N_2$ - and Ar-diluted  $H_2$ - $O_2$  flames, having  $\alpha = 3.2$  and 5.0, respectively, are cellular. Rather than preferential diffusion effects we believe cellularity occurs because  $H_2$  is the more completely consumed reactant, therefore  $Le_{eff} = Le_F \approx 0.3$ . If a premixed-flame structure existed, it would lie on the oxidizer side, contrary to their suggestion that the flame sits on the fuel side. Since  $\alpha > 1$  for their  $H_2$ - $O_2$ -He flames, again  $Le_{eff} = Le_F$ . Since  $D_F/D_O > 1$ , preferential diffusion arguments would dictate cellularity, however, the flame was not cellular because  $Le_{eff} > 1$ . (Note that even for  $H_2$ ,  $Le_F$  can be greater than unity depending on diluent type).

For the paper-burning experiments,<sup>14</sup> the pyrolysis products are not known, hence  $Le_F$  cannot be determined, but since  $Y_{F,+ \infty} \approx 1$  close to the fuel surface and because the overall stoichiometry of cellulose combustion is known, we can infer  $\alpha \ll 1$ . Then, since  $Le_O \approx 0.53$  and 0.75 in  $SF_6$ - and  $CO_2$ -diluted atmospheres, respectively, cellular instability is expected to occur and is experimentally observed. Cellular flames were not observed with He, Ne, Ar, or  $N_2$  dilution. Since  $Le_O > 0.8$  in these cases, cellularity is not expected by our criterion.

#### *Effects of Instabilities on Extinction Limits:*

In premixed flames, cellular instabilities in low- $Le$  mixtures enable flames to exist under conditions where the adiabatic flame temperature ( $T_{ad}$ ) is below the explosion limit temperature. For example,<sup>5,7</sup> the lean  $H_2$ -air premixed flammability limit is about 4.0%  $H_2$ , corresponding to  $T_{ad} \approx 620$  K. No compositions with corresponding-low  $T_{eq}$  were flammable in our nonpremixed flames. This might be because the structure of nonpremixed flames with large  $|\alpha + 2\beta - 1|$  is very similar to that of premixed flames with heat loss *but with a temperature*

*gradient in the thermal-decay zone comparable to that of the zone upstream of the reaction zone.*<sup>15</sup> In premixed flames near extinction, the ratio of these gradients is much smaller, namely of order  $1/\theta$ .<sup>1,5</sup> Possibly this inherently-strong heat loss in nonpremixed flames prevents low- $T_{eq}$  mixtures from being flammable, but no theoretical description of nonpremixed cellular flames is available to test this hypothesis.

## Conclusions

Studies of cellular nonpremixed flames reveal that these phenomena are closely associated with the nature of structures within nonpremixed flames similar to those of premixed flames. It is proposed that cellular structures occur when (1) near-extinction conditions are present, so that intermixing of fuel and oxidant occurs and (2) within the framework of Liñán's criterion, burning occurs under conditions where the more completely consumed reactant has a Lewis number sufficiently less than unity ( $\approx 0.8$ ). The proposed criterion is independent of the means by which near-extinction conditions are obtained, that is, whether heat-loss or limited residence time (low  $Da$ ) causes extinction and whether jet momentum or buoyancy controls flame height and residence time. Simple flame-height scaling laws<sup>21</sup> and constant Damköhler-number models seem to qualitatively describe the various extinction regimes.

All current and previous experimental observations are consistent with the proposed cellularity criterion. Additional support for the role of premixed cellular flame structures within nonpremixed flames are our observations that heat-loss promotes instability and that cellular flame structures are consistent with considerations of where low- $Da$  (and thus intermixing) is most likely. Previously-proposed preferential diffusion and stoichiometry criteria are unable to predict our experimental results.

Probably because of differences in premixed versus nonpremixed flame structure, flammability enhancement via diffusive-thermal mechanisms is less than in premixed flames. No analysis of diffusive-thermal instability in nonpremixed flames has been reported; such an analysis would be more difficult than stability analyses of planar premixed flames<sup>1-6</sup>, since in our geometry the stationary state is inherently two-dimensional.

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#### COMMENTS

A. Ghoniem, *Massachusetts Institute of Technology, USA*. Can you explain what do you mean by saying that the flame behavior is premixed-flame-like? What does lead to premixing in this diffusion flame and by what mechanism?

*Author's Reply.* According to Liñán's analysis, at low  $Da$  (i.e., when the flow time scale is sufficiently rapid compared to the chemical time scale) there is insufficient residence time for complete chemical reaction to occur. Consequently, both reactants leak through the reaction zone. Since the activation energy is assumed large, the temperature outside this zone is too low for significant quantities of leaked reactants to be consumed on a time scale commensurate with the flow time scale. The resulting low- $Da$  temperature and concentration profiles are much like that of a (lean, for  $\alpha > 1$ ) premixed flame with heat loss because (1) on the fuel side, the temperature rises as the reaction zone is approached, (2) the temperature falls on the other side of the reaction zone, (3) the fuel is almost completely consumed in the reaction zone, and (4) the oxidant is in stoichiometric excess throughout the reaction zone (outside the reaction zone, the composition is irrelevant because the composition affects only the reaction rate). The only difference between this

structure and a lean premixed flame with heat loss is that the oxidant profile slopes the opposite direction. However, since oxidant is plentiful (compared to fuel) throughout this zone, this difference is not significant.

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F. A. Williams, *University of California at San Diego, USA*. It seems to me that for dilute, strongly diffusing reactants there is an instability mechanism in which the reactants can diffuse through the inerts to flame stripes that become effectively less dilute, leaving mixtures between stripes too weak to burn. Describing this requires consideration of diffusion and finite-rate chemistry but *not* any premixed convective-diffusive zone. From this viewpoint I question your claim that premixing is essential for the cellular instability of the diffusion flame. Someone should do a simple AEA theory to test this!!

*Author's Reply.* From your description it is not clear to us by what mechanism small disturbances would be amplified to generate the nonuniform flame you propose. The term "strongly diffusing" could imply either strongly diffusing with respect to other



reactants or strongly diffusing with respect to thermal energy. The former is characterized by the ratio of reactant diffusivities (relevant to the preferential diffusion model of instability) and the latter by the Lewis numbers of the reactants (relevant to the diffusive-thermal model). The experiments support the latter model.

We did not mean to imply that a premixed-like *convective-diffusive* zone (where both reactants are present in significant quantities far removed from the reaction zone on one side) is required for instability. What Liñán's analysis predicts (when  $\alpha$  is

not close to unity) is a premixed-like *reactive-diffusive* zone (which is what you suggest is necessary). As we mention in the answer to Prof. Ghonheim's question, the composition outside the reaction zone is irrelevant. Since a premixed-like convective-diffusive zone may not exist, probably only the initial instability mechanism is similar to that of premixed cellular flames; the final cellular flame structure is probably not like that of a premixed flame.

We agree that someone should do an analysis of diffusion-generated instabilities in nonpremixed flames.