Incomplete combustion in nonadiabatic premixed gas flames

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The inward propagating spherical flame and burner stabilized Bunsen-type flame of low-Lewis-number premixtures are studied numerically. It is shown that reduction of the reaction rate induced by the flame stretch makes the flame vulnerable to the radiative heat losses which may well result in a partial or complete extinction of the flame. [S1063-651X(96)09505-0]

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I. INTRODUCTION

It is well known that the structure of the Bunsen flame is highly sensitive to the deficient reactant's Lewis number, Le. If thermal diffusivity of the premixture sufficiently exceeds the reactant's molecular diffusivity (Le$>$1) the flame assumes a continuous luminous conical shape. However, in the opposite case, when molecular diffusivity is high enough (Le $<$1), the flame appears as if it were open at the tip (Lewis and Elbe [1], Law *et al.* [2], Mizomoto *et al.* [3], Mizomoto and Yoshida [4]). In lean hydrogen-air mixtures the unburned hydrogen was found to pass through the tip, provided its content in the fresh mixture is low enough [1].

The analytical description based on slowly varying flame formulation succeeded in capturing many basic features of the phenomenon (Sivashinsky [5]). At Le$<$1 the temperature, reaction rate, and flame speed were found to suffer a dramatic drop as the reaction zone approaches the tip.

In order to maintain the flame at the tip its speed should be equal to that of the incoming gas flow. In light of the observed tendencies it was plausible to expect that near the tip the flame simply goes out. Yet, the above analysis being asymptotic in nature was, strictly speaking, not valid at the extremity of the tip. To obtain a more comprehensive picture a direct numerical simulation of the system was undertaken (Kozlovsky and Sivashinsky [6]). Despite a considerable depletion of the reaction rate (quite in accord with analytical predictions) the flame was found to survive along the whole interface. Moreover, there was no leakage of the deficient reactant through the reaction zone. Similar observations were reported in the recent study by Katta and Roquermore [7] where sophisticated numerical simulations of lean hydrogen-air Bunsen flames accounting for the detailed chemistry and the fluid dynamics were conducted.

For all its seemingly counterintuitive nature the results obtained did not appear as completely unexpected. A few years earlier a similar observation was made for a physically related but geometrically simpler case of the inward propagating cylindrical or spherical flames (Buckmaster and Crowley [8], Fernandez *et al.* [9]). It was found that while at Le$<$1 the reacting rate of the converging flame suffers a significant reduction, there was no residue of the unburned fuel. Note that Buckmaster and Crowley [8] were actually concerned with the problem of the highly elongated axisymmetrical Bunsen flame, which appears to be formally identical to that of inward propagating cylindrical flame.

Since both Bunsen and converging flames at Le$<$1 may be rather strongly stretched, the negative result with regards to quenching and leakage posed a serious challenge to the common belief in the flame extinction by merely enhancing its local stretch [1]. In these circumstances the well known quenching of the counterflow, high-Lewis-number planar flame seems to be rather a nongeneric case, likely to be attributed to the convective heat losses imposed by the diverging flow field. The importance of heat losses has recently also been recognized studying the phenomenon of flame quenching by turbulence (Brailovsky and Sivashinsky [10]). Without heat losses the extinction does not seem feasible no matter how strong the flame distortion and stretch. In light of these observations it is interesting to ascertain whether incorporation of heat losses may also promote the incomplete combustion in low-Lewis-number Bunsen and inward propagating flames, two of the most basic systems of premixed combustion involving stretch. The inward propagating flames are known to occur in turbulent combustion where the highly distorted flame sometimes ejects pockets of unburned gas [11–13].

II. INWARD PROPAGATING FLAME

We employ the following conventional framework for the model: one-step irreversible exothermic reaction with Arrhenius's kinetics, reactant composition is far from stoichiometry, constant density and transport properties, and the radial symmetry of the pertinent temperature-reactant fields (Fig. 1). In the present model we also include the effect due to radiative heat losses. With these assumptions the appropriately nondimensionalized set of equations for temperature and the deficient reactant reads as

$$\frac{\partial T}{\partial t}=\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial T}{\partial r}\right)+(1-\sigma)\Omega(C,T)-Q(T), \qquad (1)$$

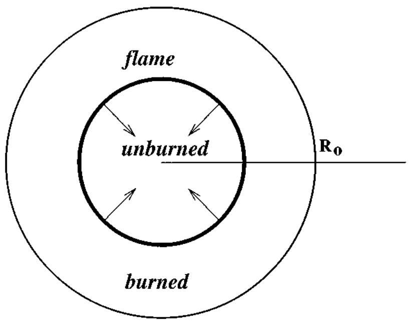

FIG. 1. Diagram of inward propagating spherical flame. The bold line corresponds to the flame interface, arrows indicate direction of flame motion.

$$\frac{\partial C}{\partial t} = \frac{1}{\text{Le}} \frac{1}{r^2} \frac{\partial}{\partial r}\left(r^2 \frac{\partial T}{\partial r}\right) - \Omega(C,T), \tag{2}$$

where

$$\Omega(C,T) = \tfrac{1}{2}(1-\sigma)^2 \text{Le}^{-1} N^2 C \exp[N(1-1/T)]. \tag{3}$$

Here, T is the nondimensional temperature in units of T_b, the adiabatic temperature of combustion products; C is the nondimensional concentration of the deficient reactant in units of C_0, its value in the fresh mixture; r is the nondimensional spatial coordinate in units of $l_{\text{th}} = D_{\text{th}}/U_b$, the thermal width of the flame; D_{th} is the thermal diffusivity of the mixture; U_b is the speed of a planar adiabatic flame; t is the nondimensional time in units of l_{th}/U_b, $\sigma = T_0/T_b$, T_0 being the fresh mixture temperature; $N = T_a/T_b$ is the nondimensional activation energy, T_a being the activation temperature; $\text{Le} = D_{\text{th}}/D_{\text{mol}}$ is the Lewis number, D_{mol} being the molecular diffusivity of the deficient reactant; $Q(T) = h(T^4 - \sigma^4)$ is the term responsible for the radiative heat losses with h being the scaled Stefan-Boltzmann constant in units of $\rho_b c_p l_p U_b / 4T_b^3 l_{\text{th}}$, where l_p is the Plank mean absorption length, c_p is the specific heat, and ρ_b is the burned gas density, and $\Omega(C,T)$ is the appropriately normalized reaction rate to ensure that at large N the nondimensional speed of a well-settled planar adiabatic flame will be close to unity.

Equations (1) and (2) are considered in the interval $0 < r < R_0$, and subject to the insulating boundary conditions

$$\frac{\partial T}{\partial r} = 0, \quad \frac{\partial C}{\partial r} = 0 \quad \text{at } r = 0, R_0. \tag{4}$$

The initial conditions are defined as

$$C(r,0) = C_p(r - R_1), \quad T(r,0) = T_p(r - R_1) \tag{5}$$

where C_p, T_p are the numerically calculated concentration and temperature profiles in a *planar* nonadiabatic flame at $h = h_q(N,\text{Le},\sigma)$ corresponding to the planar flame quenching threshold, and $r = R_1$ is the point where the reaction rate $\Omega[C(r,0),T(r,0)]$ reaches its maximum. In all subsequent numerical runs R_1 is taken as $R_0 - 60$.

In order to check whether radiative heat losses may indeed lead to incomplete combustion one may follow the temporal evolution of the integral:

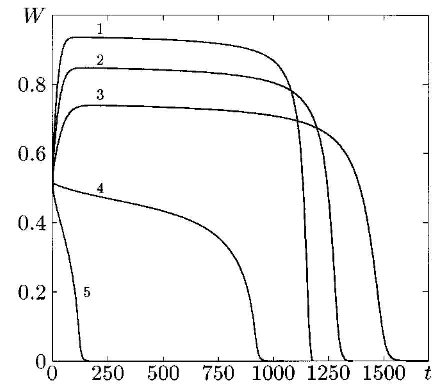

FIG. 2. Temporal evaluation of the reaction rate integral [Eq. (7)] for different levels of heat losses: curve 1, $h=0$; curve 2, $h=0.01$; curve 3, $h=0.02$; curve 4, $h=0.0325$; curve 5, $h=0.04$ ($\sigma=0.2$, Le=0.5, $N=10$, $R_0=1120$).

$$M(t) = 4\pi \int_0^{R_0} C(r,t) r^2 dr, \tag{6}$$

corresponding to the total amount of the deficient reactant available in the system at a given instant of time.

It is also instructive to see the temporal evolution of the reaction rate bulk intensity W, defined as

$$W(t) = \int_0^{R_0} \Omega(C,T) dr. \tag{7}$$

The problem (1)–(5) was solved for Le=0.5; $\sigma=0.2$; $N=5,10,20$; $R_0=1120$; $0<h<0.1$ and $0<t<1700$. Note that for the initial condition (4) $M(t)$ may never exceed $4\pi R^3/3 \simeq 4.99\times 10^4$. In the absence of heat losses ($h=0$), in spite of the strong decay of the reaction rate [see Fig. 2 (curve 1)], by the end of the process all fuel is found to be used up [Fig. 3 (curve 1)], see also [9]. The incorporation of heat losses ($h>0$) changes the picture significantly.

Figures 2 and 3 show the temporal evolution of the integrals (6) and (7) at $N=10$, Le=0.5, $\sigma=0.2$, $R_0=1120$, and $h=0.01$ (curve 2), $h=0.02$ (curve 3), $h=0.0325$ (curve 4), $h=0.04$ (curve 5). In this case, in contrast to the adiabatic one, as the flame approaches the center the amount of uncon-

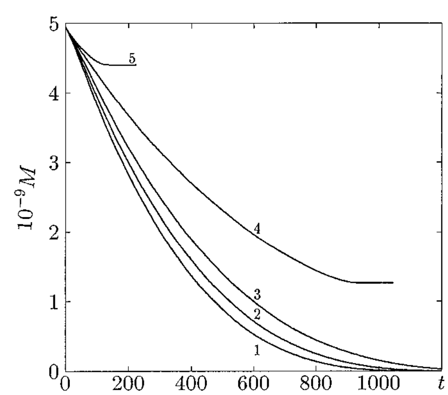

FIG. 3. Temporal evolution of the deficient reactant integral [Eq. (6)] for different levels heat losses: curve 1, $h=0$; curve 2, $h=0.01$; curve 3, $h=0.02$; curve 4, $h=0.0325$; curve 5, $h=0.04$ ($\sigma=0.2$, Le=0.5, $N=10$, $R_0=1120$).

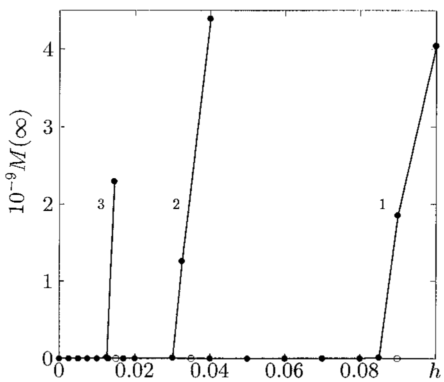

FIG. 4. Total amount of unburned fuel vs heat loss intensity for different levels of activation energy: curve 1, $N=5$, curve 2, $N=10$, and curve 3, $N=20$. Open circles (○) on the h axis correspond to the flammability limits of the associated planar flames.

sumed fuel, M, does not vanish but rather comes to a certain finite value. With increasing the level of heat losses, as one might expect, the effect becomes more pronounced.

Speaking of the unconsumed fuel one of course presumes the time duration (nondimensional) of the process to be comparable with R_0, i.e., when combustion actually occurs as a flame. For $t \gg R_0$, since the reaction rate Ω is never identically zero, all the fuel will be eventually used up regardless of the heat loss intensity.

As has long been observed by Joulin and Clavin [14] for any heat loss however small there is a nonzero level of fuel leakage. In planar high activation energy flames this effect is extremely weak up to very flammability point, $h=h_q$. Beyond h_q the flame goes out and thereupon reaction occurs at such a low rate that the fuel consumption may safely be regarded as terminated. For Le=0.5, $\sigma=0.2$, and $N=5,10,20$ as was verified numerically, the planar flame quenches at $h=h_q=0.090$, 0.035, and 0.015, respectively.

A similar development clearly takes place in inward propagating flames as well. In these systems, however, the flame goes out at a level of heat losses, $h=h_l$, noticeably lower than h_q, the quenching point of the associated planar flame.

Figure 4 and Table I yield the total amount of the unburned fuel, M, near the flammability threshold versus the heat loss intensity, h, evaluated for Le=0.5, $\sigma=0.2$, and $N=5$ (curve 1), $N=10$ (curve 2), $N=20$ (curve 3). Figures 5 and 6(a)–(c) depict the temperature, concentration, and the reaction rate profiles for the intermediate and the final stage of the process where the flame is nearly quenched.

TABLE I. Total amount of unburned fuel M_∞ and heat loss intensity h for Le=0.5 and $\sigma=0.2$.

$N=5$		$N=10$		$N=20$	
h	M_∞	h	M_∞	h	M_∞
0	0	0	0	0	0
0.02	0.0039	0.01	0.216	0.0025	0.591
0.04	1.9	0.013	34.1	0.05	5520
0.05	89	0.017	1920	0.0075	71700
0.06	1740	0.02	12900	0.01	77600
0.07	27900	0.03	1.13e+07	0.0125	1.61e+07
0.08	794000	0.0325	1.25e+09	0.0145	2.29e+09
0.085	1.29e+07	0.04	4.29e+09		
0.09	1,85e+09				
0.1	4.04e+09				

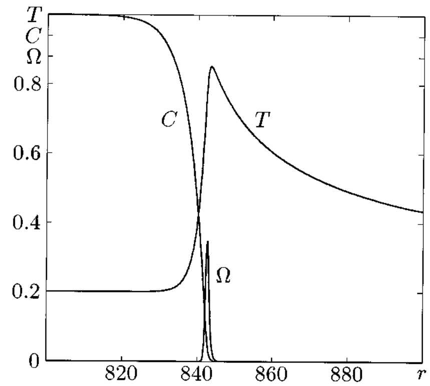

FIG. 5. Intermediate distribution of the concentration (C), temperature (T), and reaction rate (Ω) at $t=450$ ($h=0.0325$, Le=0.5, $\sigma=0.2$, $N=10$, $R_0=1120$).

III. BURNER STABILIZED FLAME

Despite its conceptual simplicity the Bunsen burner flame constitutes quite a nontrivial system for the adequate mathematical description. Even for the constant-density approximation the pertinent hydrodynamical problem appears to be rather involved. For the limited objective of the present study, however, it will be sufficient to consider a related yet geometrically simpler system, where flame is stabilized not at the burner rim but rather in the Poiseuille flow between two parallel plates (Fig. 7). To hold the flame on such a burner the lower half of the channel wall ($y<0$) is maintained at a fixed temperature $T=\sigma$, while the upper half ($y>0$) is regarded as thermally insulated. Adopting the basic assumptions of the preceding section the appropriately modified set of reaction-diffusion equations [(1), (2)] may be written as

$$\frac{\partial T}{\partial t}+v(x)\frac{\partial T}{\partial y}=\frac{\partial^2 T}{\partial x^2}+\frac{\partial^2 T}{\partial y^2}+(1-\sigma)\Omega(C,T)-Q(T), \tag{8}$$

$$\frac{\partial C}{\partial t}+v(x)\frac{\partial C}{\partial y}=\frac{1}{\mathrm{Le}}\left(\frac{\partial^2 C}{\partial x^2}+\frac{\partial^2 C}{\partial y^2}\right)-\Omega(C,T), \tag{9}$$

where $v(x)=V[1-(x/d)^2]$ is the flow velocity profile.

Equations (8) and (9) are considered within a rectangular domain $(-d<x<d, -m<y<l)$ with the boundary conditions:

$$\begin{aligned}
&T=\sigma, \quad C=1 \quad \text{at } y=-m, \quad -d<x<d\\
&T_y=0, \quad C_y=0 \quad \text{at } y=1, \quad -d<x<d\\
&T=\sigma, \quad C_x=0 \quad \text{at } x=\pm d, \quad -m<y<0\\
&T_x=0, \quad C_x=0 \quad \text{at } x=\pm d, \quad 0<y<l.
\end{aligned} \tag{10}$$

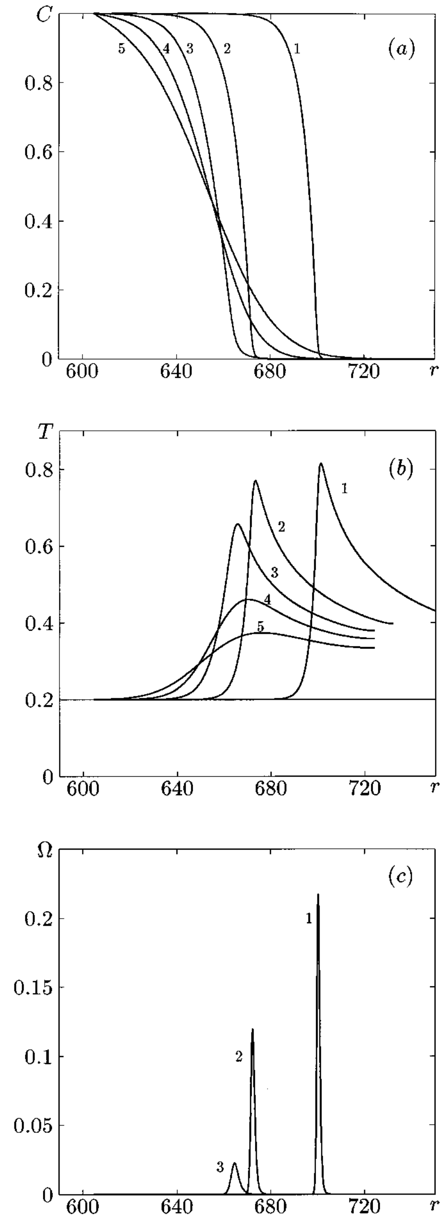

FIG. 6. Final distributions of the concentration (a), temperature (b), and reaction rate (c) at several consecutive instants of time: (1) $t=750$; (2) $t=800$; (3) $t=950$; (4) $t=1000$; (5) $t=1070$ ($h=0.0325$, Le=0.5, $\sigma=0.2$, $N=10$, $R_0=1120$).

To produce the equilibrium solution the following initial conditions were used:

$$C(x,y,0)=1$$

$$T(x,y,0)=\sigma+(1-\sigma)\exp[-(x^2+y^2)]. \quad (11)$$

The problem (8)–(11) was solved for the following set of parameters:

$$\text{Le}=0.5, \quad (12)$$

$$N=5,$$

$$\sigma=0.2,$$

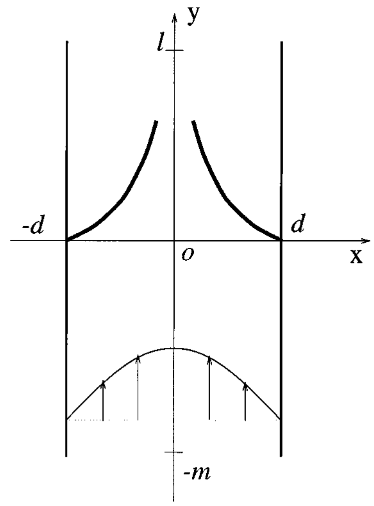

FIG. 7. Diagram of Bunsen-burner–type flame stabilized in the Poiseuille flow between two parallel plates. Bold line and arrows correspond to the flame interface and the flow-field velocity profile, respectively.

$$d=30,$$

$$l=450,$$

$$m=50,$$

$$V=2,4,$$

$$h=0,0.2,0.3,0.4.$$

As a numerical method an alternative directions finite-difference scheme on a uniform orthogonal grid was employed.

Figure 8(a) depicts distribution of the temperature, concentration, and reaction rate in the absence of radiative heat losses ($h=0$). While the flame visually appears here as if it were open at its tip, there is actually no leakage. All the fuel supplied to the reaction zone is fully consumed [6,7].

The introduction of heat losses alters the picture considerably. Figures 8(b)–8(e) show the temperature, concentration, and reaction rate distributions at $h=0.2$, 0.3, 0.4, and $V=2$, 4. The tip opening appears here to be even more prominent. Moreover, this time the fuel indeed leads through the flame. Figures 9 and 10 demonstrate temperature, concentration, and reaction rate distributions along the lines $x=0$ (curve 1), $x=d/3$ (curve 2), $x=2d/3$ (curve 3), and $x=d$ (curve 4). Shown in Fig. 11 is the fuel concentration at infinity, C_∞, versus the heat loss intensity, h.

Note that in this case the flame survives (partially) at the level of heat losses markedly exceeding the flammability limit ($h_q=0.09$) of the planar flame of the same premixture (see Sec. II). The reason for such an outcome is easily explained. It is well known that in low-Lewis-number premixtures the freely propagating flame spontaneously assumes a cellular structure whose low-temperature ridges are pointed toward the burned gas. In this sense the low-Lewis-number Bunsen flame may be regarded as a portion of a cellular flame attached to a fixed position at the burner's wall ($y=0$,

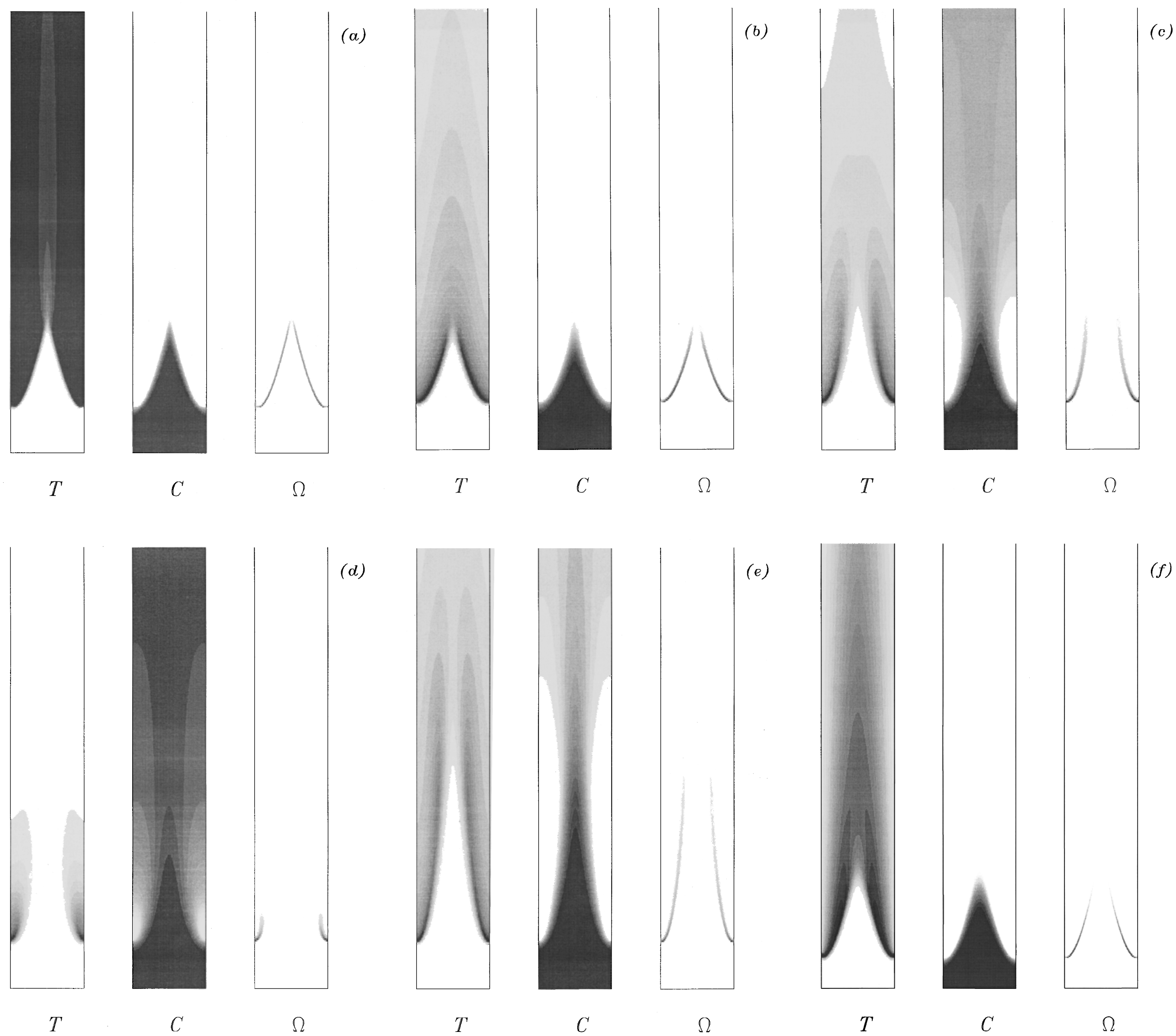

FIG. 8. Distribution of temperature (T), concentration (C), and reaction rate (Ω) for (a) $h=0$, $V=2$; (b) $h=0.2$, $V=2$; (c) $h=0.3$, $V=2$, (d) $h=0.4$, $V=2$, (e) $h=0.3$, $V=4$; (f) $h=0$, $V=2$, conductive heat losses (13).

$x=\pm d$, Fig. 7). It is also known that cellular flames may survive far beyond the planar flame flammability limit and virtually without fuel leakage (e.g., Joulin and Sivashinsky [15], Kagan and Sivashinsky [16]). Yet at sufficiently high heat losses the cellular flame breaks up into separate cup-like—or ball-like—flamelets while a considerable amount of fuel escapes the reaction zone and remains unconsumed (e.g., Buckmaster [17]). The above results for the burner may clearly be seen as a manifestation of similar trends in the context of the anchored cellular flames. The later, however, compared to the freely evolving cells, appear to be markedly stretched by the underlying flow.

For the same level of heat losses an increase in the flow velocity V results in the leakage rate $[VC_\infty(V)]$ enhancement (Table II). Yet due to the extinction of the highly reactive parts of the flame the final concentration $C_\infty(V)$ appears to be diminished [Figs. 8(c)–8(e)]. Note that above a certain level V_{cr} the attachment of the flame becomes impossible and it is swept away by the incoming flow. For example, in the adiabatic case ($h=0$), other parameters being as in (12), $V_{cr}=25$.

It is interesting that due to the invariably present molecular diffusion, the unconsumed reactant carried through the open tip again finds itself near the undepleted parts of the flame adjacent to the tip. This time, however, the reactant appears on the opposite, combustion products side of the reaction zone. One, as it were, ends up here with a diffusion flame in which the roles of the oxidant and fuel are played by the same reactant.

IV. CONCLUDING REMARKS

The two systems considered in this study give ground to the following general assertion. The stretch, while by itself being unable to destroy the flame, nonetheless may significantly reduce its reaction intensity. This makes the flame vulnerable to the volumetric heat losses, which may well bring it to a partial or complete extinction. To ensure a no-

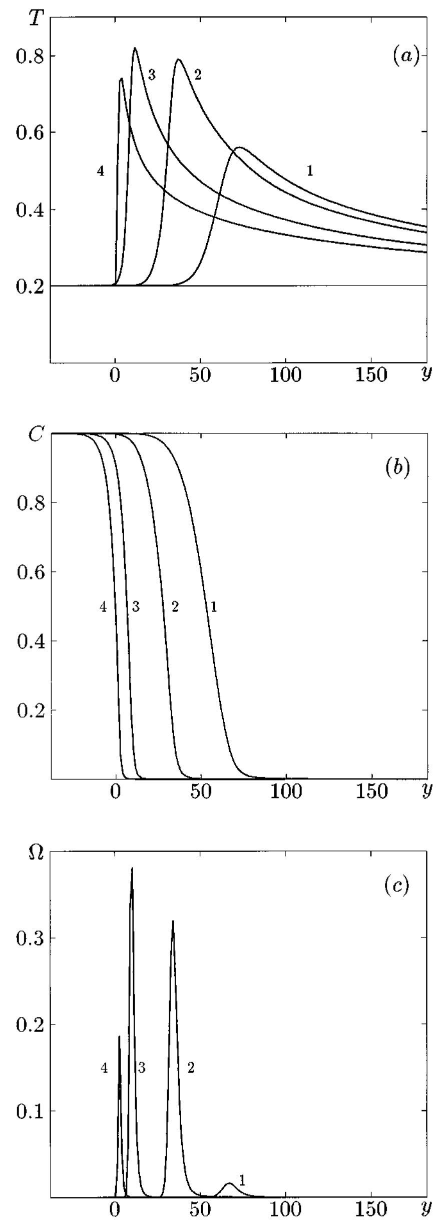

FIG. 9. Temperature (a), concentration (b), and reaction rate (c) profiles along the lines: $x=0$ (curve 1), $x=d/3$ (curve 2), $x=2d/3$ (curve 3), and $x=d$ (curve 4), for $h=0.2$ and $V=2$.

ticeable leakage of the nominally deficient reactant the level of heat losses, however, should exceed a certain threshold, h_l. Depending on the geometry of the problem h_l may well turn out to be higher than h_q (planar flame quenching threshold), as happens in the above considered Bunsen burner flame (Sec. III).

In the adopted formulation the heat loss intensity, h, appears as a prescribed but otherwise arbitrary parameter. The natural question is whether for the real life radiative emission this number may become high enough to ensure the global or local extinction. In the absence of quantitative experimental data on the inward propagating or burner stabilized flames one may look at the physically related and well explored problem of the outward propagating spherical flame of lean CH_4-O_2-N_2 premixture (Ronney [18], Sibulkin and Frendy [19,20]). Similar to the above system the Lewis number of

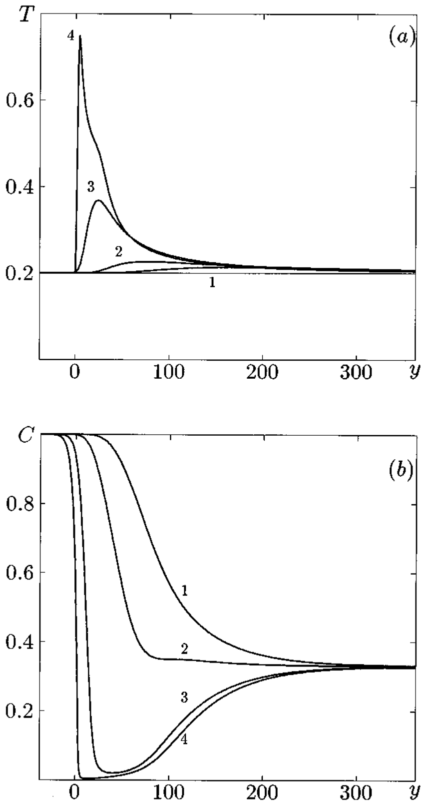

FIG. 10. Temperature (a) and concentration (b) profiles along the lines: $x=0$ (curve 1), $x=d/3$ (curve 2), $x=2d/3$ (curve 3), and $x=d$ (curve 4), for $h=0.2$ and $V=2$.

the deficient reactant, CH_4, is below unity and the flame is under permanent stretch. Comparison of the calculated flammability limit induced by the radiative emission from the products (CO_2, H_2O) with the observations shows quite good agreement.

Theoretical predictions appear to be rather close to observations also for the radiation controlled hydrogen-air flame balls occurring in very lean hydrogen-air premixtures (Buckmaster *et al.* [21]). In light of this one may be quite confident that the radiative heat loss is an effective enough agency to provide incomplete combustion both for inverted spherical and burner stabilized CH_4-H_2–air flames as well.

One should realize, however, that under different external

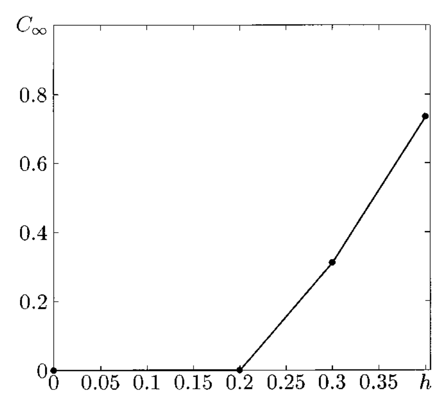

FIG. 11. Fuel concentration on ''infinity'' for various levels of heat losses.

TABLE II. Flow velocity V and leakage rate VC_∞.

V	C_∞	VC_∞
2	0.311	0.622
4	0.207	0.828

conditions similar effects may be achieved not via radiative, but rather via conductive heat losses. Such a situation may arise, for example, in the low-Lewis-number inward propagating cylindrical or wedge-shaped flames evolving between two closely spaced plates that provide the necessary heat losses (cf. Joulin and Sivashinsky [22]). The pertinent mathematical models are readily obtained from those of Secs. II and III by setting $Q(T)=h(T-\sigma)$.

The most interesting issue here is, however, to ascertain what happens in a convenentional Bunsen burner where the flame is in a permanent contact with the low temperature surroundings. Do the far-field conductive heat losses induce the flame tip leakage? To tackle the problem one may still adopt the model of Sec. III where the boundary condition for temperature at $x=\pm d$ is set as

$$T(-\pm d,y)=\sigma+(1-\sigma)\exp(-y^2). \tag{13}$$

The elevated temperature at $y=0$ produces the ‘‘igniting ring’’ that prevents the flame blowoff [Fig. 8(f)]. The numerical simulations conducted for a wide enough range of the flow ratges, V, did not detect any fuel leakage unless the gap, d, falls below some critical value. In the latter case the flame goes out simultaneously over the whole cross section of the channel. The tip leakage occurring in the lean hydrogen-air Bunsen flame [1] is therefore likely to be controlled by radiative rather than conductive heat losses.

The difficulties in earlier analytical descriptions of the adiabatic or nearly adiabatic Bunsen flame tips at Le<1 do not necessarily imply that the flame tip is essentially a nonlocal problem where the flame structure is controlled by the overall flame shape. Indeed, the theory of cellular flames (e.g., Sivashinksy [23]), pertaining precisely to Le<1 system, suggests that in order to regularize the solution near the tip one should address a higher order slowly varying flame formulation incorporating not only the flame stretch but its variation along the flame front as well. Derivation of the corresponding fourth-order differential equation for the flame interface involves some tedious algebra, but otherwise should be rather straightforward. The modified relation between the local geometry of the flame and its speed should elucidate how the adiabatic flame may maintain full consumption of the fuel under low reaction rate simultaneously keeping the local flame speed at a rather high level. The systematic exploration of this issue will be addressed in a future work.

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