

# Lecture 12

## The Structure of diffusion flames Mixture fraction formulation

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### Governing Equations

$$\rho c_p \left( \frac{\partial T}{\partial t} + \mathbf{v} \cdot \nabla T \right) - \nabla \cdot (\lambda \nabla T) = Q \varpi$$

$$\rho \left( \frac{\partial Y_F}{\partial t} + \mathbf{v} \cdot \nabla Y_F \right) - \nabla \cdot (\rho \mathcal{D}_F \nabla Y_F) = -\nu_F W_F \varpi$$

$$\rho \left( \frac{\partial Y_O}{\partial t} + \mathbf{v} \cdot \nabla Y_O \right) - \nabla \cdot (\rho \mathcal{D}_O \nabla Y_O) = -\nu_O W_O \varpi$$

$$\varpi = B \frac{\rho Y_F}{W_F} \frac{\rho Y_O}{W_O} e^{-E/RT}$$

+ mass and momentum equations

$$\rho T = P_0 W / \mathcal{R}$$

For simplicity of presentation we will treat the properties constant, but the results can be easily generalized if they are temperature-dependent.

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Non-dimensionalization.

Choose,  $U$  as a unit of speed,  $l_D = \lambda/\rho_0 c_p U$  and  $l_D/U$  as units of length and time. Use  $Q/c_p v_F w_F$  and  $P_0$  to nondimensionalize temperature and pressure and scale  $\rho$  accordingly.

$$\begin{aligned}\rho\left(\frac{\partial T}{\partial t} + \mathbf{v} \cdot \nabla T\right) - \nabla^2 T &= \varpi \\ \rho\left(\frac{\partial Y_F}{\partial t} + \mathbf{v} \cdot \nabla Y_F\right) - Le_F^{-1} \nabla^2 Y_F &= -\varpi \\ \rho\left(\frac{\partial Y_O}{\partial t} + \mathbf{v} \cdot \nabla Y_O\right) - Le_O^{-1} \nabla^2 Y_O &= -\nu \varpi \\ \varpi &= D \rho^2 Y_F Y_O e^{\beta/T_a - \beta/T}\end{aligned}$$

$$\rho T = 1$$

We are already familiar with all the parameters appearing in these equations. Note, in particular, that

$$D \sim \frac{D_{th}/U^2}{(Be^{-E/RT_a})^{-1}} \quad \beta = \frac{Q/c_p}{\mathcal{R}^2/E} \quad Ta \text{ is the (dimensionless) adiabatic temperature}$$

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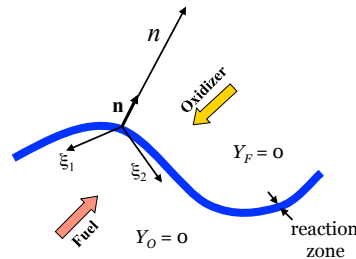
**The structure of the Burke-Schumann solution**

$$D \rightarrow \infty \Rightarrow Y_F Y_O \sim 0$$

The chemical reaction is confined to a sheet  $F(\mathbf{x}, t) = 0$

on either side of the reaction sheet

$$\begin{aligned}\rho\left(\frac{\partial T}{\partial t} + \mathbf{v} \cdot \nabla T\right) - \nabla^2 T &= 0 \\ \rho\left(\frac{\partial Y_F}{\partial t} + \mathbf{v} \cdot \nabla Y_F\right) - Le_F^{-1} \nabla^2 Y_F &= 0 \\ \rho\left(\frac{\partial Y_O}{\partial t} + \mathbf{v} \cdot \nabla Y_O\right) - Le_O^{-1} \nabla^2 Y_O &= 0\end{aligned}$$



Introduce an orthogonal intrinsic coordinate system  $(\xi_1, \xi_2, n)$ , then

$$\nabla = \mathbf{n} \frac{\partial}{\partial n} + \nabla_{\perp}$$

where  $n$  is a coordinate along the normal, and  $\mathbf{n}$  is a unit normal to the surface, given by

$$\mathbf{n} = \frac{\nabla F}{|\nabla F|}$$

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The solution inside the reaction zone is slightly modified from the values along the sheet and, on is obtained by stretching the normal coordinate and determining the appropriate balance between the various terms in the conservation laws.

$$n = \varepsilon \eta$$

$$Y = \varepsilon y_F(\eta) + \dots, \quad Y_O = \varepsilon y_O(\eta) + \dots$$

$$T = T_a + \varepsilon \tau(\eta) + \dots$$

$$\frac{1}{\varepsilon} \frac{\partial^2 \tau}{\partial \eta^2} \sim D \varepsilon^2 y_F y_O \quad \Rightarrow \quad \frac{\partial^2 \tau}{\partial \eta^2} \sim \frac{D \varepsilon^3 e^{-\beta/T_a}}{O(1)} y_F y_O$$

$$\frac{\partial^2}{\partial \eta^2} (\tau + Le_f^{-1} y_f) = \frac{\partial^2}{\partial \eta^2} [\tau + (vLe_o)^{-1} y_o] = 0 \quad \varepsilon \sim D^{-1/3}$$

- The reaction zone is diffusive-reactive, with convection negligibly small
- The thickness of the reaction zone for large  $D$  is  $\sim D^{-1/3}$
- The reaction proceeds at a rate that depends on the local fuel and oxidizer concentrations, but is independent of temperature (nothing has been assumed about the Zel'dovich number, so that  $\exp(\beta/T_a - \beta/T) \sim 1$  when  $T \sim T_a$ )

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$$\frac{\partial^2}{\partial \eta^2} (\tau + Le_f^{-1} y_f) = \frac{\partial^2}{\partial \eta^2} [\tau + (vLe_o)^{-1} y_o] = 0$$

$$\tau + v^{-1} Le_o^{-1} y_o = a\eta + b$$

$$\tau + Le_f^{-1} y_f = -c\eta + d$$

$$\frac{\partial^2 \tau}{\partial \eta^2} = C(a\eta - \tau)(c\eta + \tau)$$

$$\tau \rightarrow a\eta \quad \text{as } \eta \rightarrow -\infty$$

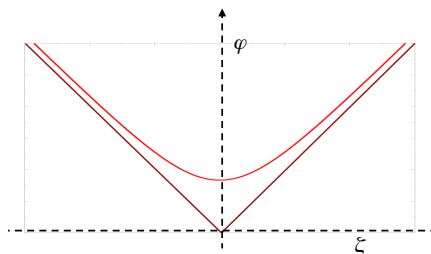
$$\tau \rightarrow -c\eta \quad \text{as } \eta \rightarrow \infty$$

Matching conditions with the solution on either side of the reaction sheet, yields  $b = d = 0$  and expressions for  $a, c$

A transformation of the coordinate  $\eta$  and the dependent variable  $\tau$  yields, after some algebraic manipulations, the canonical problem

$$\frac{\partial^2 \varphi}{\partial \zeta^2} = (\varphi - \zeta)(\varphi + \zeta)$$

$$\frac{\partial \varphi}{\partial \zeta} \rightarrow \pm 1 \quad \text{as } \zeta \rightarrow \pm \infty$$



The solution always exist!

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## Activation Energy Asymptotics

*Linan, Acta Astro. 1974; Cheatham & Matalon, JFM 2000*

The previous discussion did not exhibit the extreme sensitivity of the reaction rate to small changes in temperature. The proper expansion parameter is the Zel'dovich, or activation energy parameter  $\beta$ , or  $\varepsilon \sim 1/\beta$ .

$$\begin{aligned} n &= \varepsilon \eta \\ Y &= \varepsilon y_f(\eta) + \dots, \quad Y_o = \varepsilon y_o(\eta) + \dots \\ T &= T_a + \varepsilon \tau(\eta) + \dots \end{aligned}$$

$$\omega = D \rho^2 Y_f Y_o e^{\beta/T_a - \beta/T} = D \rho^2 Y_f Y_o e^{\beta(T - T_a)/T_a} \sim D \rho_a^2 (\varepsilon y_f)(\varepsilon y_o) e^{\beta \varepsilon \tau / T_a^2}$$

$$\text{Let } \varepsilon = T_a^2 / \beta$$

$$-\frac{1}{\varepsilon} \frac{\partial^2 \tau}{\partial \eta^2} = \hat{D} \varepsilon^2 \rho_a^2 y_f y_o e^{\tau / T_a^2} \qquad -\frac{\partial^2 \tau}{\partial \eta^2} = \underbrace{D \varepsilon^3 \rho_a^2}_{\hat{D} = O(1)} y_f y_o e^{\tau}$$

$$D = (\beta^3 / T_a^4) \hat{D} \quad \text{distinguished limit}$$

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$$\frac{\partial^2}{\partial \eta^2} (\tau + L e_f^{-1} y_f) = \frac{\partial^2}{\partial \eta^2} [\tau + (v L e_o)^{-1} y_o] = 0$$

$$\tau + v^{-1} L e_o^{-1} y_o = a \eta + b$$

$$\tau + L e_f^{-1} y_f = -c \eta + d$$

The coupling functions provide linear relations between  $y_f, y_o$  and  $\tau$  which, when substituted into the equation for the temperature yields, as before, after an appropriate transformation of the variables and some manipulations, a canonical problem

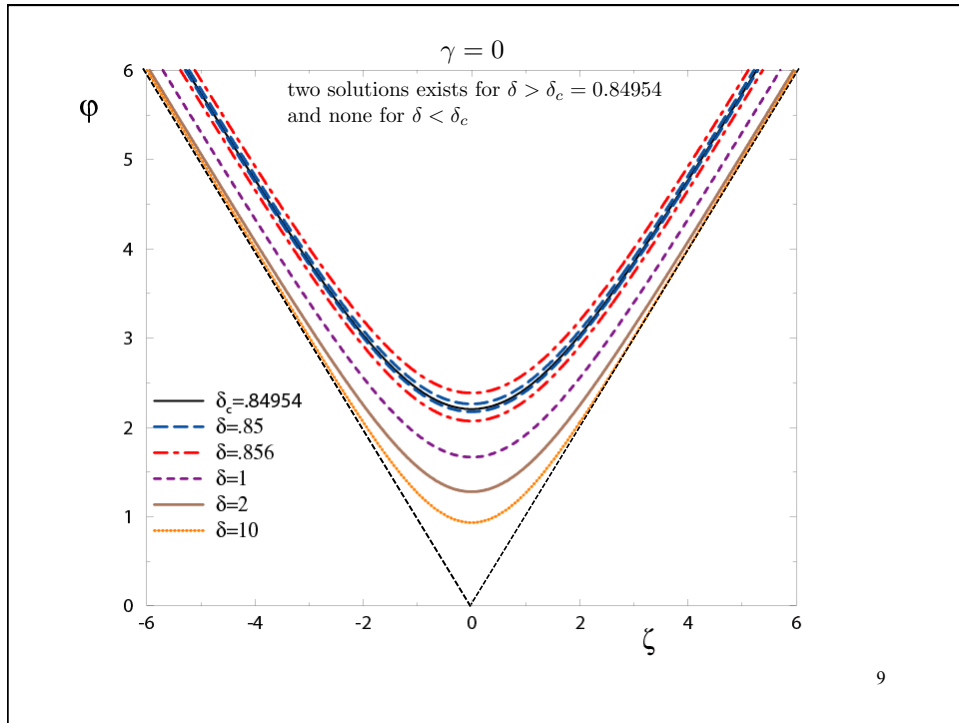
$$\frac{\partial^2 \varphi}{\partial \xi^2} = (\varphi - \zeta)(\varphi + \zeta) \exp\{\delta^{-1/3}(\varphi + \gamma \zeta)\}$$

$$\frac{\partial \varphi}{\partial \xi} \rightarrow \pm 1 \quad \text{as } \zeta \rightarrow \pm \infty$$

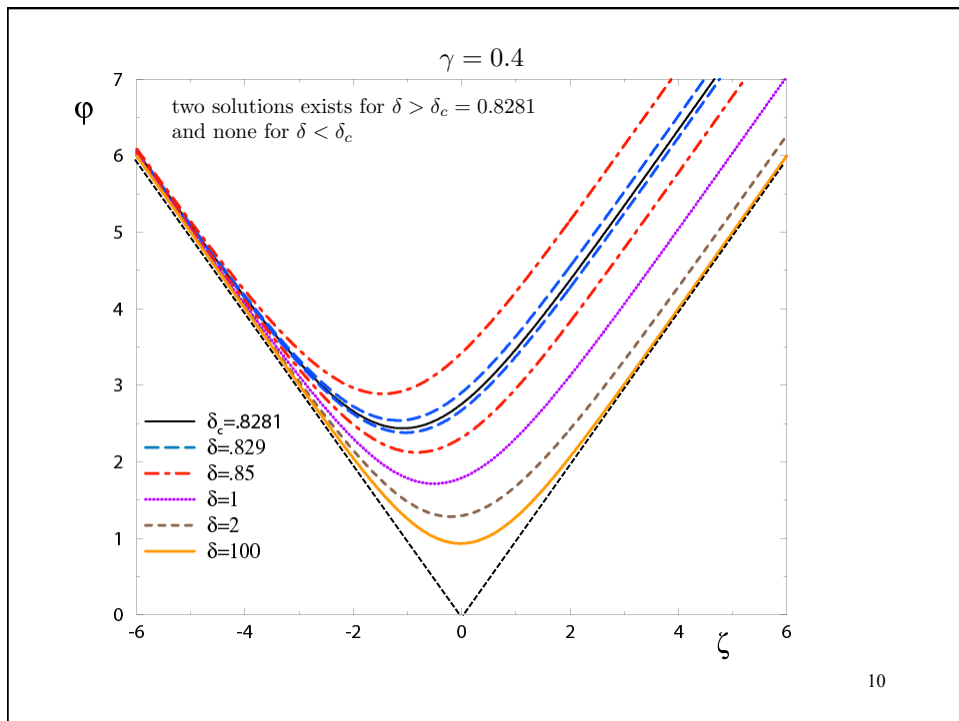
but with two auxiliary parameters  $\gamma$  and  $\delta$  determined from the “outer solution”, namely the solution outside the reaction zone.

Note that  $\delta \sim D$  is effectively a “reduced Damköhler number”, which extend the solution to large but not necessarily infinite  $D$ . It is evident that when  $\delta = \infty$  the problem reduces to the structure of the BS solution.

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- The solution of the BVP determines

$$\lim_{\xi \rightarrow \infty} (\varphi - \zeta) = \delta^{1/3} S_1 \quad \text{and} \quad \lim_{\xi \rightarrow -\infty} (\varphi + \zeta) = \delta^{1/3} S_2$$

where  $S_1$  and  $S_2$  are the leakage functions, that determine the degree of leakage of fuel and/or oxidizer ( $S_F$  and  $S_O$ ) through the reaction zone.

- The parameter  $-1 < \gamma < 1$  fixes the amount of heat needed for given conditions and depends strongly on the mixture strength  $\phi$ . Specifically it is the excess of heat conducted to one side of the reaction sheet, and is given by

$$\gamma = -\frac{1}{\Delta} \left\{ (\partial T / \partial n)^{n=0^+} + (\partial T / \partial n)^{n=0^-} \right\} \quad \text{where} \quad \Delta = -[\partial T / \partial n] > 0$$

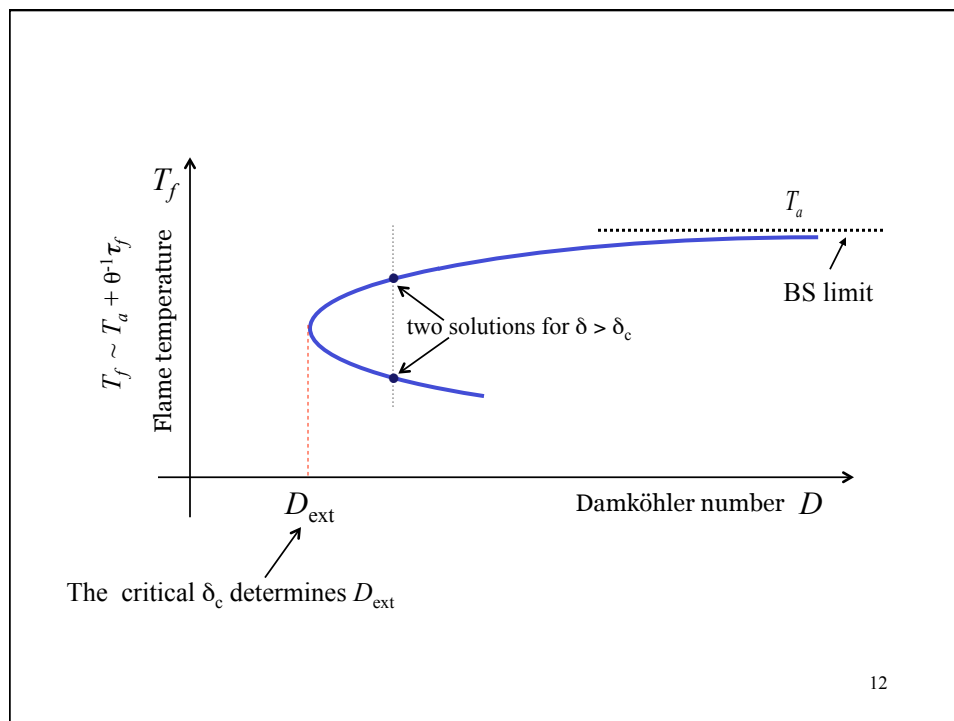
- The parameter  $\delta$  measures the intensity of the reaction; it controls how much heat can be generated for given conditions, and depends on the excess/deficiency in available enthalpy in the reaction zone.

$$\delta = \frac{4Le_o Le_F T_a^4 D}{\beta^3 \Delta^2} e^{h_f/2} \quad h_f = (T_f - T_a)\beta + \frac{1-\gamma}{2} S_F + \frac{1+\gamma}{2} S_O$$

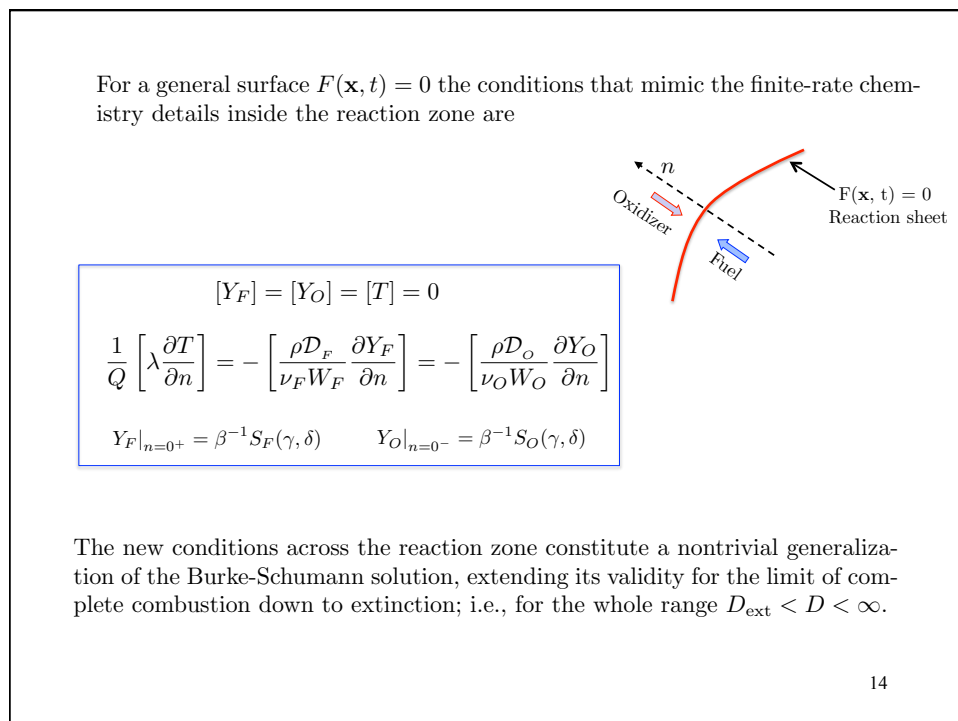
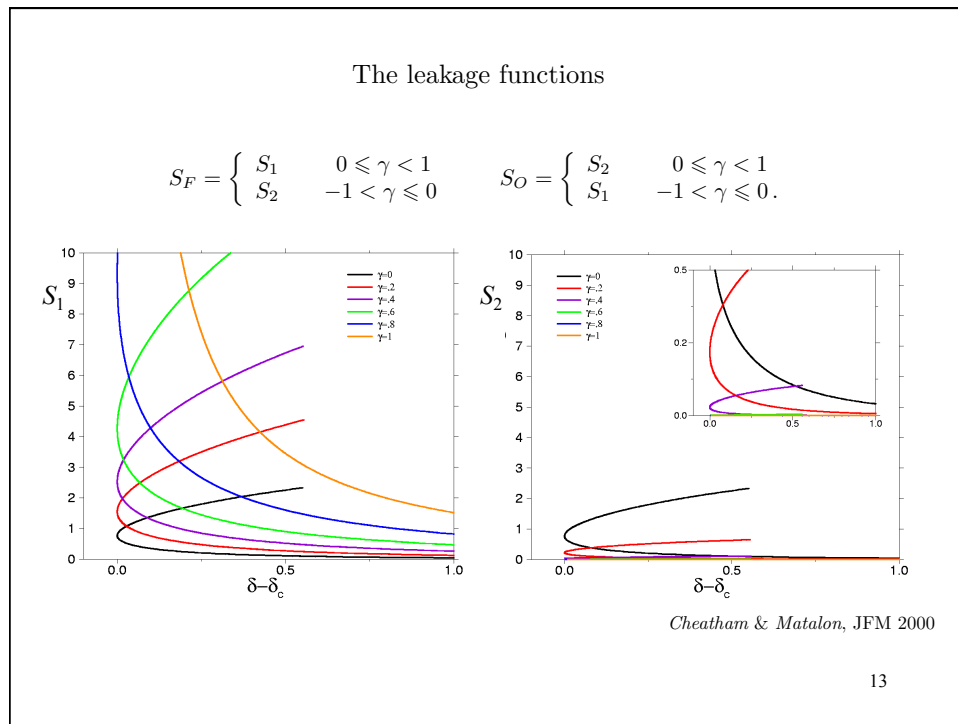
- The nonexistence of solutions for  $\delta < \delta_c$ , or equivalently for  $D$  below a critical value is associated with extinction. Linan (1974) provided the approximation

$$\delta_c \approx e \cdot \left\{ (1-|\gamma|) - (1-|\gamma|)^2 + 0.26(1-|\gamma|)^3 + 0.055(1-|\gamma|)^4 \right\}$$

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### The Mixture Fraction

In premixed combustion the fuel-to-oxidizer ratio, or equivalence ratio, is fixed and remains constant everywhere. Consequently, the progress of combustion can be described by a variable measuring the extent of completion of the reaction. In diffusion flames, the fuel-to-oxidizer ratio is not constant and an additional variable is needed. A convenient one is the mixture fraction defined as  $Z = 0$  in the oxidizer stream and  $Z = 1$  in the fuel stream.

**The mixture fraction**  $Z = Z(\mathbf{x}, t)$  is the fraction of the total amount of material present that came originally from the fuel stream.

Consider a two-stream problem, having uniform properties on one portion of the boundary - the fuel stream, (denoted with subscript 1) and different uniform properties over the rest of the boundary - the oxidizer stream (denoted with subscript 2). There is no fuel in the oxidizer boundary and no oxidizer in the fuel boundary.

$$Z = \frac{\nu Y_F - (Y_O - Y_{O_2})}{\nu Y_{F1} + Y_{O2}} \quad \nu = \frac{\nu_O W_O}{\nu_F W_F}$$

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A more general way, good for more involved chemistry, is to use the local mass fractions of atoms.

For example: suppose the fuel stream contains  $\text{CH}_4$  and the oxidizer stream contains air. Fuel atoms appear in  $\text{CH}_4$ ,  $\text{CO}$ ,  $\text{OH}$ ,  $\text{H}_2\text{O}$ , etc . . .

The element mass fraction of C and H is

$$\frac{16}{16} Y_{\text{CH}_4} + \frac{12}{28} Y_{\text{CO}} + \frac{1}{17} Y_{\text{OH}} + \frac{2}{18} Y_{\text{H}_2\text{O}}$$

which can be normalized such that  $Z = 1$ .

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From the species equations

$$\rho \frac{DY_F}{Dt} - \rho \mathcal{D}_F \nabla^2 Y_F = -\nu_F W_F \omega \quad \rho \frac{DY_O}{Dt} - \rho \mathcal{D}_O \nabla^2 Y_O = -\nu_F W_O \omega$$

$$\rho \frac{DZ}{Dt} - \rho \mathcal{D}_F \nabla^2 Z = (\rho \mathcal{D}_F - \rho \mathcal{D}_O) \nabla^2 Y_O$$

we see that only for equal diffusivities,  $Z$  satisfies a reaction-free equation (i.e., it is a conserved scalar, which is a quantity that is neither created or destroyed by chemical reaction)

$$\rho \frac{DZ}{Dt} - \rho \mathcal{D} \nabla^2 Z = 0$$

Expressions relating the mass fractions to  $Z$  are easily obtained. From the definition

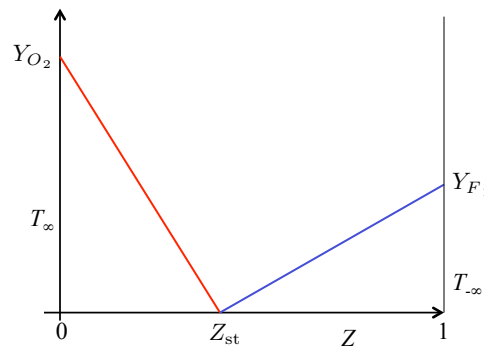
$$Z = \frac{\nu Y_F - (Y_O - Y_{O_2})}{\nu Y_{F_1} + Y_{O_2}} \quad \Rightarrow \quad \begin{aligned} Y_F &= Y_O = 0 \\ Z_{st} &= \frac{Y_{O_2}}{\nu Y_{F_1} + Y_{O_2}} \end{aligned}$$

defines the stoichiometric surface

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Since on one side of the stoichiometric surface  $Y_O = 0$  and on the other side  $Y_F = 0$ , we have

$$\begin{aligned} \text{For } 0 < Z < Z_{st} \quad Y_O &= Y_{O_2} \left(1 - \frac{Z}{Z_{st}}\right) \\ \text{For } Z_{st} < Z < 1 \quad Y_F &= Y_{F_1} \left(\frac{Z - Z_{st}}{1 - Z_{st}}\right) \end{aligned}$$



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$$\rho \frac{DT}{Dt} - \frac{\lambda}{c_p} \nabla^2 T = Q\omega$$

Since in the fast chemistry limit, the temperature also satisfies a reaction-free equation on either side of the stoichiometric surface, i.e.,  $\omega = 0$ , the temperature  $T$  may also be expressed as a linear combination of  $Z$ . For, if  $T = T(Z(\mathbf{x}, t))$

$$\begin{aligned} \frac{DT}{Dt} &= \frac{dT}{dZ} \frac{DZ}{Dt}; & \nabla^2 T &= \frac{dT}{dZ} \nabla^2 Z \\ \rho \frac{DT}{Dt} - \frac{\lambda}{c_p} \nabla^2 T &= \left( \rho \frac{DZ}{Dt} - \frac{\lambda}{c_p} \nabla^2 Z \right) \frac{dT}{dZ} = 0 \end{aligned}$$

which is clearly satisfied, provided, the Lewis number is one ( $\lambda/c_p = \rho D$ ).

$$\begin{aligned} \text{Hence } T &= T(Z) \\ T(0) &= T_2, \quad T(1) = T_1 \end{aligned}$$

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$$\begin{aligned} \rho \frac{DY_F}{Dt} - \rho D_F \nabla^2 Y_F &= -\nu_F W_F \omega \\ \rho \frac{DY_O}{Dt} - \rho D_O \nabla^2 Y_O &= -\nu_F W_O \omega \\ \rho \frac{DT}{Dt} - \frac{\lambda}{c_p} \nabla^2 T &= Q\omega \end{aligned}$$

We note that, for unity Lewis numbers, the functions

$$h_F = T + (Q/c_p \nu_F W_F) Y_F, \quad h_O = T + (Q/c_p \nu_O W_O) Y_O$$

are conserved scalars.

$$h_F = h_F(Z), \quad h_O = h_O(Z)$$

$$\begin{aligned} h_F &= T_2 + [(Q Y_{F_1} / c_p \nu_F W_F) + T_1 - T_2] Z \\ h_O &= T_1 + [(Q Y_{O_2} / c_p \nu_O W_O) + T_2 - T_1] (1 - Z) \end{aligned}$$

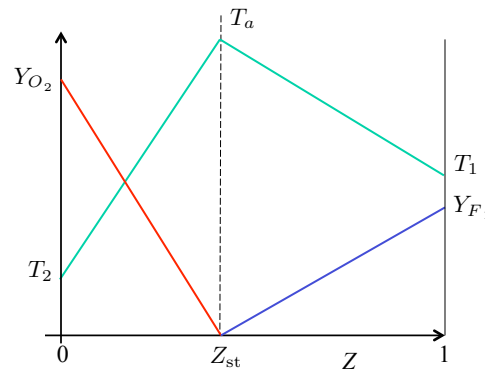
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from which we deduce

$$T = \begin{cases} T_2 + [(QY_{F_1}/c_p\nu_F W_F) + T_1 - T_2] Z & 0 < Z < Z_{st} \\ T_1 + [T_2 + (QY_{O_2}/c_p\nu_O W_O)](1 - Z) & Z_{st} < Z < 1 \end{cases}$$

$$T_a = T_2 + [(QY_{F_1}/c_p\nu_F W_F) + T_1 - T_2] Z_{st}$$

the adiabatic temperature



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We have seen that the temperature and mass fractions expressed in terms of the mixture fraction  $Z$  takes a very simple form. When the combustion field is expressed in terms of one coordinate, it is convenient to make a change of variables and use  $Z$  as the transformed coordinate. This, of course, under the assumption of unity Lewis numbers. For example, in the counterflow configuration, the equation for  $Z$  is (in dimensional form)

$$2\epsilon x \frac{dZ}{dx} + D \frac{d^2 Z}{dx^2} = 0$$

$$\Rightarrow Z = \frac{1}{2} \left( 1 - \operatorname{erf}(\sqrt{\epsilon/D} x) \right)$$

This transformation contracts the axial coordinate  $x$  from  $(-\infty, \infty)$  to  $(1, 0)$ .

The structure of the reaction zone, which follows the previous discussion provides the limitation  $D > D_{\text{ext}}$  for the existence of steady solutions, or equivalently  $\epsilon < \epsilon_{\text{ext}}$ .

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In more general circumstances, the largest changes in the mixture-fraction field  $Z(\mathbf{x}, t)$  occur along the direction of  $|\nabla Z|$  from the surface  $Z = Z_{st}$ . A coordinate transformation  $(x, y, z) \rightarrow (x, y, Z(x, y, z))$  is then used to describe the reaction zone structure.

This can be easily related to our previous discussion, using that

$$-\frac{\partial}{\partial n} = |\nabla Z| \frac{\partial}{\partial Z} + \frac{\nabla Z \cdot \nabla_T}{|\nabla Z|}$$

where  $\nabla_T = \left( \frac{\partial}{\partial x}, \frac{\partial}{\partial y} \right)$  is the transverse gradient.

The equation describing the reaction zone structure becomes

$$-|\nabla Z|^2 \frac{\partial^2 \tau}{\partial \eta^2} = \hat{D} y_F y_O e^\tau$$

where  $|\nabla Z|^2$  is evaluated here at the stoichiometric surface  $Z_{st}$ , and yields the same canonical problem, as before, with the additional factor  $4(1 - Z_{st})^2 / |\nabla Z|^2$  in  $\delta$ .

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The dimensional quantity  $\chi = 2\mathcal{D}|\nabla Z|^2$ , in units of 1/s is known as the instantaneous scalar dissipation rate. Its value at the stoichiometric surface,  $\chi_{st}$ , can be interpreted as an “inverse diffusion time”. Flame extinction occurs when the scalar dissipation rate  $\chi$  exceeds a critical value  $\chi_{st}^{ext}$ , which may be associated with an extinction time.

The mixture fraction formulation is useful in more complex configurations when the profiles depend weakly on the transverse directions  $x, y$  and could be considered to depend only on  $Z$ . In this case, results of the counterflow problem could be applied to these more complex problem once the mixture fraction field  $Z(\mathbf{x}, t)$  is known.

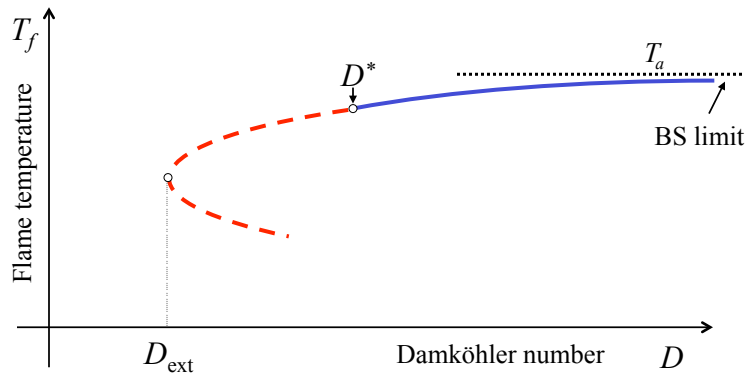
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## Flame instabilities

Instabilities in diffusion flames have been typically observed at high flow rates, or near-extinction conditions. They are predominately diffusive-thermal in nature with hydrodynamics playing a secondary role.

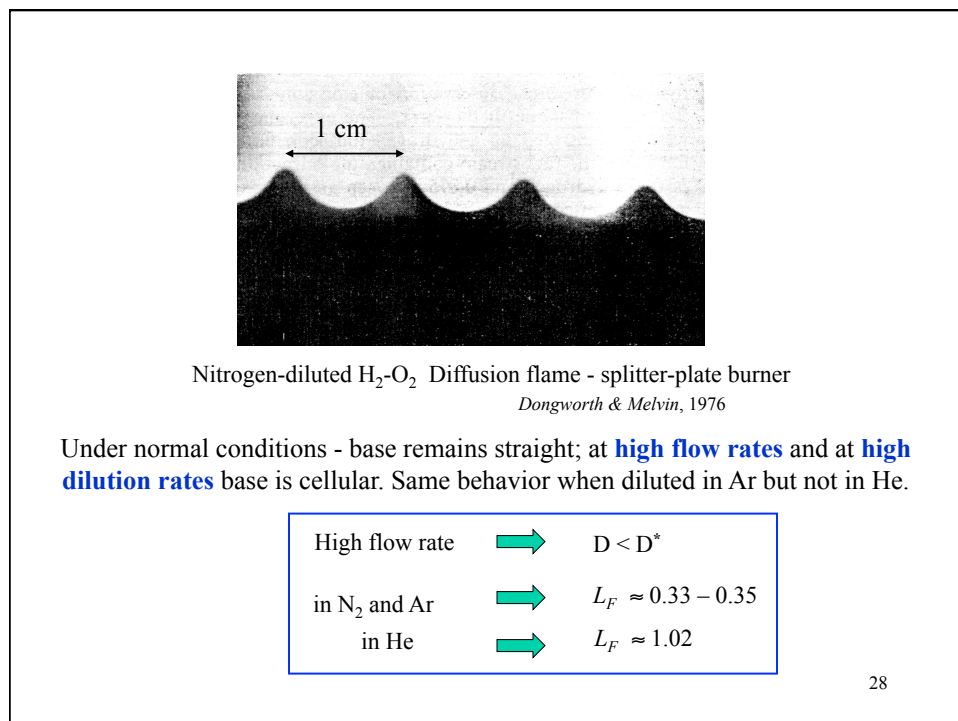
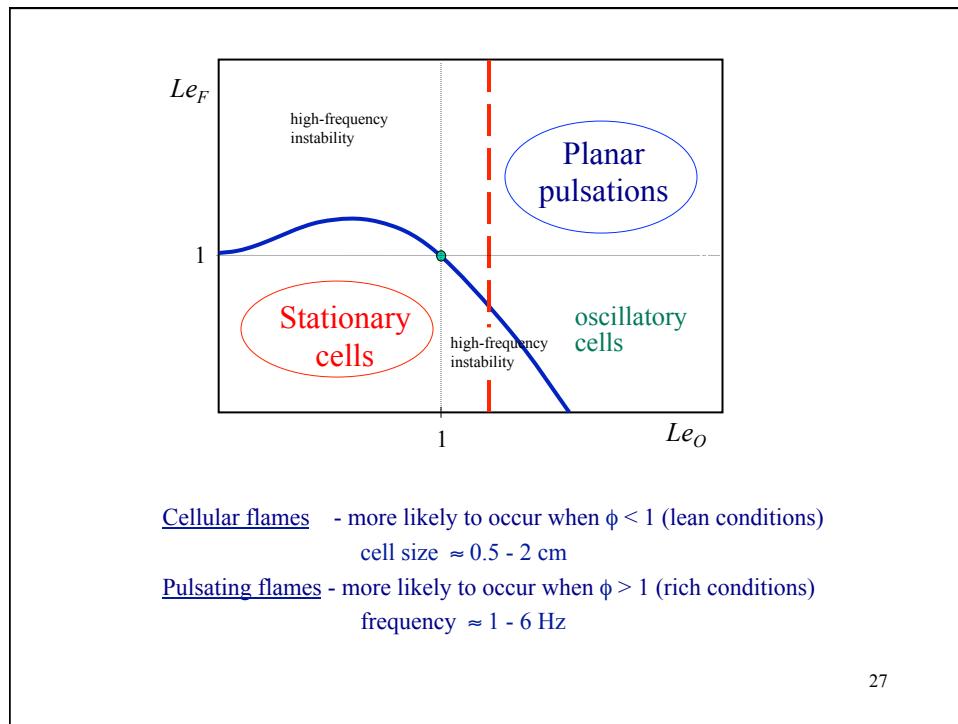
The Burke-Schumann solution of complete combustion (corresponding to the limit  $D \rightarrow \infty$ ) is unconditionally stable. Instabilities result only when there is sufficient leakage of reactants through the reaction sheet with a flame temperature significantly below  $T_a$ .

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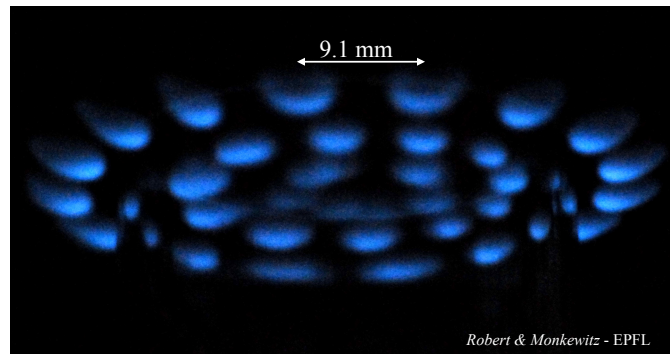
- Instabilities develop when there is significant reactant leakage  
i.e. for  $D_{\text{ext}} < D < D^*$ , namely at [high flow rates](#) or [near-extinction](#) conditions.
- Nature of the instability depends on the parameters  $L_F, L_O, \phi$

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### Cellular Diffusion Flame

H<sub>2</sub> & O<sub>2</sub> diluted in CO<sub>2</sub> ( $Le_F = 0.35$ ,  $Le_O = 0.86$ ,  $\phi = 0.24$ )



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