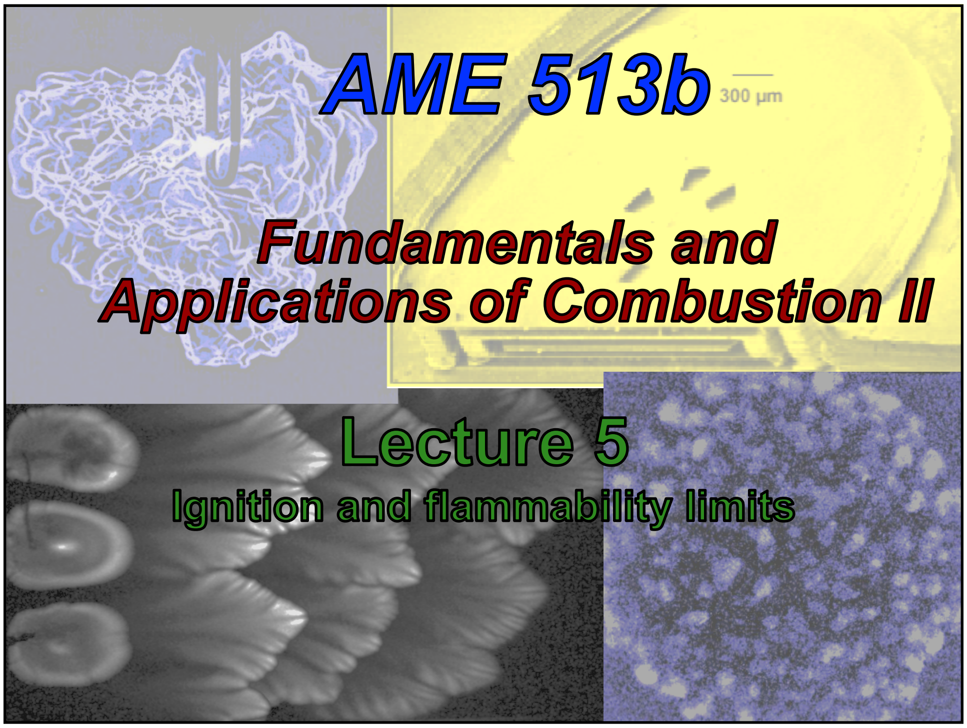


AME 513b

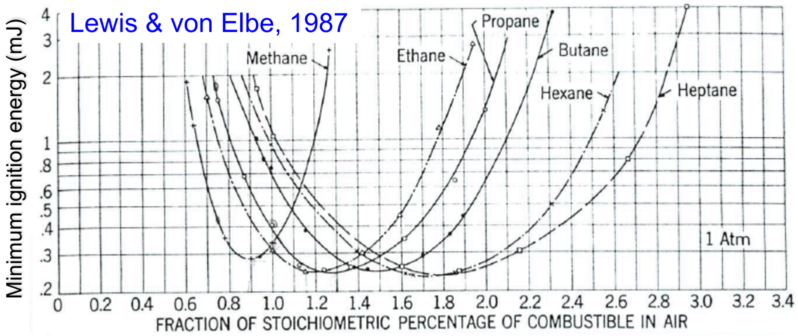
Fundamentals and Applications of Combustion II

Lecture 5

Ignition and flammability limits

Flame ignition - basic concepts

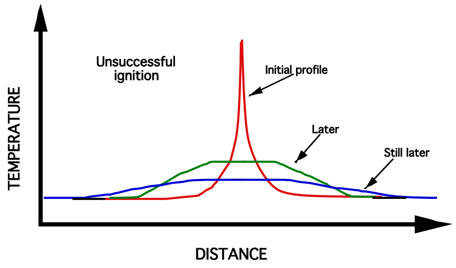
- Experiments (Lewis & von Elbe, 1987) show that a minimum energy (E_{min}) (not just minimum T or volume) required for ignition
- E_{min} lowest near stoichiometric (typically 0.2 mJ) but minimum shifts to richer mixtures for higher HCs (why? Stay tuned…)
- Prediction of E_{min} relevant to energy conversion and fire safety

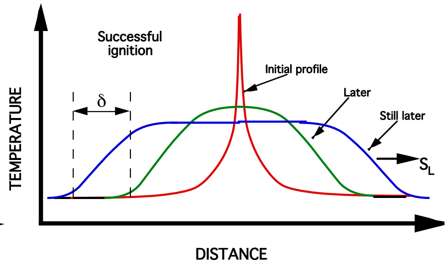

Lewis & von Elbe, 1987

Flame ignition - basic concepts

- E_{min} related to need to create flame kernel with dimension (δ) large enough that chemical reaction (ω) can exceed conductive loss rate (α/δ^2), thus $\delta > (\alpha/\omega)^{1/2} \sim \alpha/(\alpha\omega)^{1/2} \sim \alpha/S_L \sim \delta$
- E_{min} ~ energy contained in volume of gas with $T \approx T_{ad}$ and radius $\approx \delta \approx 4\alpha/S_L$

$$\Rightarrow E_{\min} \approx \frac{4\pi}{3}\delta^3\bar{\rho}C_p\left(T_{ad}-T_\infty\right) \approx 0.3\frac{4\pi}{3}\delta^3\rho_\infty C_p\left(T_{ad}-T_\infty\right) \approx 34\alpha_\infty^2\frac{k_\infty\left(T_{ad}-T_\infty\right)}{S_L^3}$$



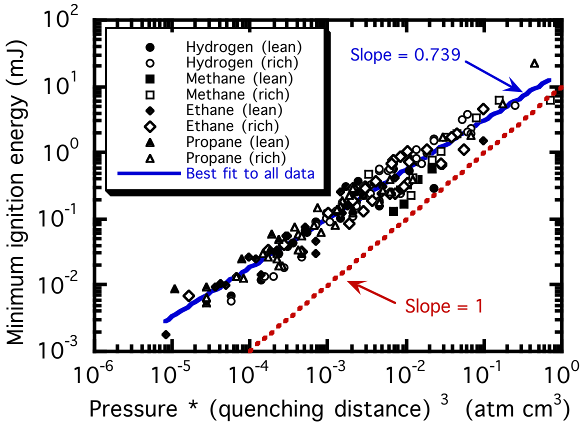


Flame igniton - simple E_{min} formula

- Since $\alpha \sim P^{-1}$, $E_{min} \sim P^{-2}$ if S_L is independent of P
- $E_{min} \approx$ 100,000 times larger in a He-diluted than SF_6-diluted mixture with same S_L, same P (due to α and k [thermal conductivity] differences)
- Stoichiometric CH_4-air @ 1 atm: predicted $E_{min} \approx$ 0.010 mJ ≈ 30x times lower than experiment (due to chemical kinetics, heat losses, shock losses …)
- … but need something more (Lewis number effects):
  - 10% H_2-air ($S_L \approx$ 10 cm/sec): predicted $E_{min} \approx$ 0.3 mJ = 2.5 times higher than experiments
  - Lean CH_4-air ($S_L \approx$ 5 cm/sec): $E_{min} \approx$ 5 mJ compared to ≈ 5000mJ for lean C_3H_8-air with same S_L - but prediction is same for both

Flame igniton - simple E_{min} formula

- $E_{min} \sim \delta^3\rho_\infty$
- δ hard to measure, but quenching distance (δ_q) (min. tube diameter through which flame can propagate) should be ~ δ since Peclet number at extinction [to be discussed later] $Pe_{lim} = S_{L,lim}\delta_q/\alpha \sim \delta_q/\delta \approx 40 \approx$ constant, thus should have $E_{min} \sim \delta_q^3 P$
- Correlation so-so

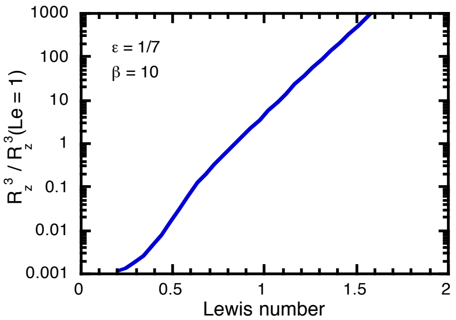


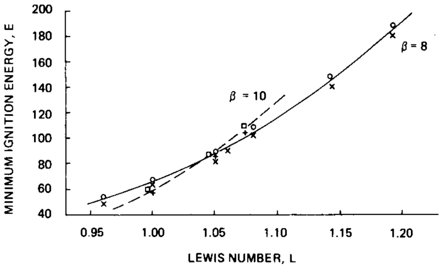
Flame igniton - Lewis number effects

- Recall flame ball solution – use R_z instead of δ to capture Le effects?

$$\frac{R_z}{\delta} = \frac{1}{Le} exp\left(\frac{\beta}{2}\left(\frac{1}{\theta^*} - 1\right)\right)$$

- Energy requirement very strongly dependent on Lewis number!
- 10% increase in Le: 2.5x increase in E_{min} (R_Z; 2.2x (Tromans & Furzeland)





Computations by Tromans and Furzeland, 1986

Flame igniton - Lewis number effects

- Why does minimum MIE shift to richer mixtures for higher HCs?
- $Le_{effective} = \alpha_{effective}/D_{effective}$
- D_{eff} = D of stoichiometrically limiting reactant, thus for lean mixtures $D_{eff} = D_{fuel}$; rich mixtures $D_{eff} = D_{O2}$
- Lean mixtures - $Le_{effective} = Le_{fuel}$
  - Mostly air, so $\alpha_{eff} \approx \alpha_{air}$; also $D_{eff} = D_{fuel}$
  - CH_4: $D_{CH4} > \alpha_{air}$ since $M_{CH4} < M_{N2\&O2}$ thus $Le_{CH4} < 1$, thus $Le_{eff} < 1$
  - Higher HCs: $D_{fuel} < \alpha_{air}$, thus $Le_{eff} > 1$ - much higher MIE
- Rich mixtures - $Le_{effective} = Le_{O2}$
  - CH_4: $\alpha_{CH4} > \alpha_{air}$ since $M_{CH4} < M_{N2\&O2}$, so adding excess CH_4 INCREASES Le_{eff}
  - Higher HCs: $\alpha_{fuel} < \alpha_{air}$ since $M_{fuel} > M_{N2\&O2}$, so adding excess fuel DECREASES Le_{eff}
  - Actually adding excess fuel decreases both α and D, but decreases α more

$$\alpha_{eff} = \alpha_{mix} \sim \sqrt{\frac{Const_1}{M_{mix}}}; D_{O_2} \sim \sqrt{\frac{Const_2}{M_{mix}} + \frac{Const_3}{M_{O2}}}$$

Flame igniton - dynamic analysis

- R_Z is related (but not equal) to an ignition requirement
- Joulin (1985) analyzed unsteady equations for Le < 1

$$\chi(\sigma) ln(\chi(\sigma)) + \frac{q(\sigma)}{2} = \chi(\sigma)\int_0^{\sigma} \frac{d\chi(s)}{d\sigma}\frac{ds}{\sqrt{\sigma - s}}$$

$$\chi \equiv \frac{R(\sigma)}{R_z}; \sigma \equiv 4\pi\left(\frac{(\theta^*)^2}{1-\varepsilon}\frac{Le}{1-\sqrt{Le}}\right)^2 \frac{\alpha t}{R_z^2}; q \equiv \frac{\Theta}{4\pi\lambda R_z T_{ad}(\theta^*)^2}$$

(χ, σ and q are the dimensionless radius, time and heat input)
and found at the optimal ignition duration

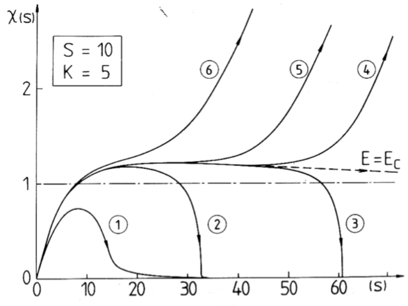
$$E_{min} \approx 14\beta\left(\frac{1-\varepsilon}{\varepsilon}\right)\left(\frac{1-\sqrt{Le}}{\theta^* Le}\right)^2 \rho_{ad} C_p (T_{ad} - T_\infty) R_z^3$$

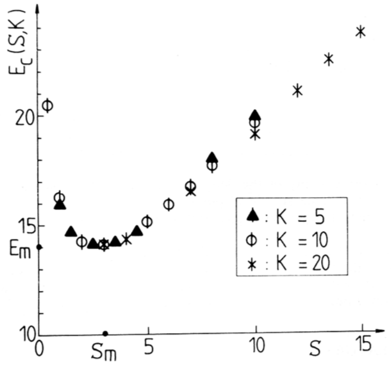
which has the expected form

E_{min} ~ {energy per unit volume} x {volume of minimal flame kernel} ~ $\{\rho_{ad} C_p(T_{ad} - T_\infty)\}$ x $\{R_z^3\}$

Flame igniton - dynamic analysis

- Joulin (1985)



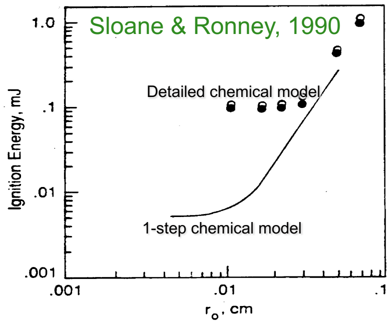


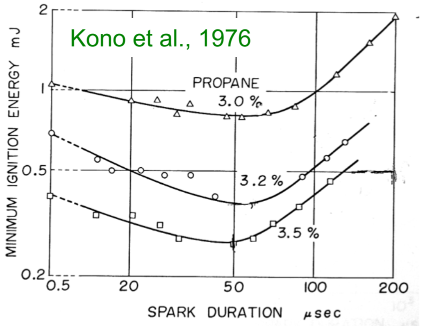
Radius vs. time

Minimum ignition energy vs. ignition duration

Flame igniton - effect of spark gap & duration

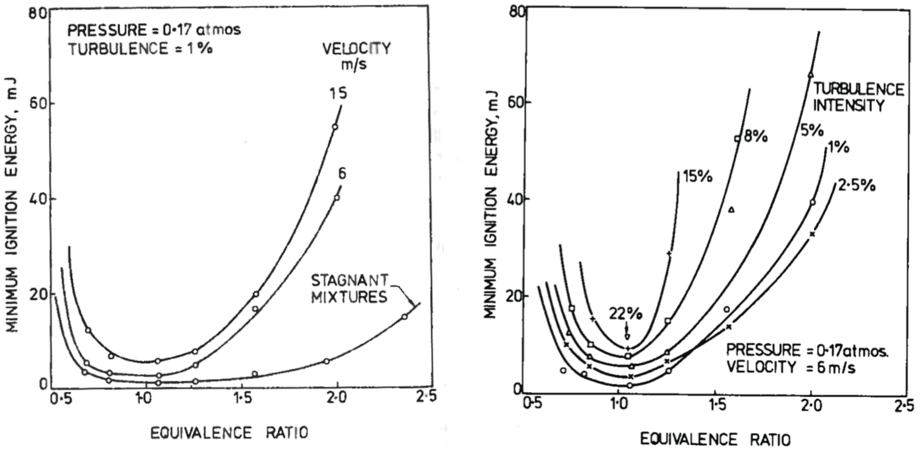
- Expect "optimal" ignition duration ~ ignition kernel time scale ~ R_Z^2/α
  - Duration too long - energy wasted after kernel has formed and propagated away - $E_{min} \sim t^1$
  - Duration too short - larger shock losses, larger heat losses to electrodes due to high T kernel
- Expect "optimal" ignition kernel size ~ kernel length scale ~ R_Z
  - Size too large - energy wasted in too large volume - $E_{min} \sim R^3$
  - Size too small - larger heat losses to electrodes


Sloane & Ronney, 1990


Kono et al., 1976

Flame igniton - effect of flow environment

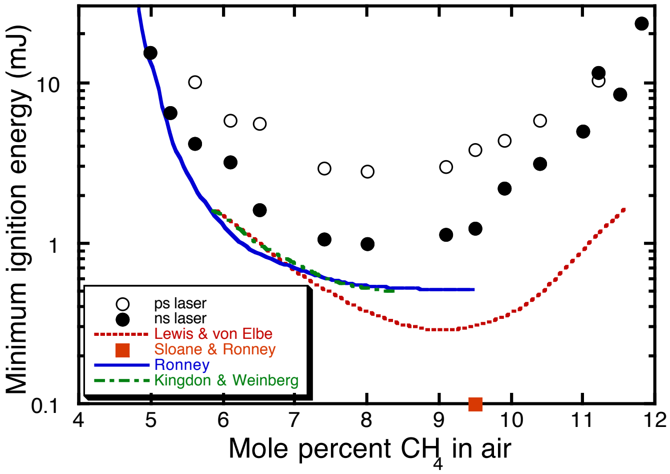
- Mean flow or random flow (i.e. turbulence) (e.g. inside IC engine or gas turbine) increases stretch, thus E_{min}



Ballal and Lefebrve, 1975

Flame igniton - effect of ignition source

- Laser ignition sources higher than sparks despite lower heat losses, less asymmetrical flame kernel - maybe due to higher shock losses with shorter duration laser source?

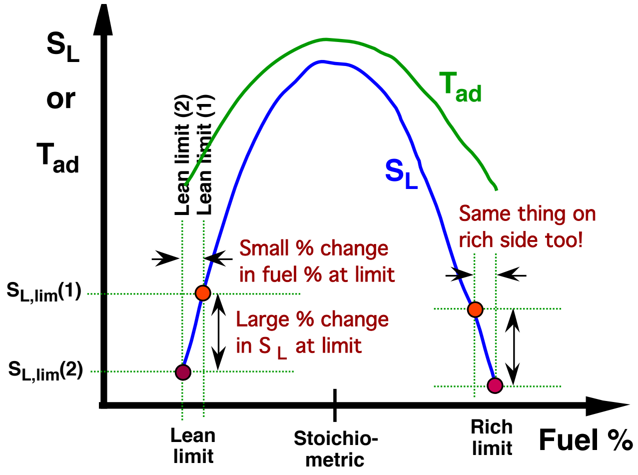


Lim et al., 1996

Flammability and extinction limits

- Too lean or too rich mixtures won't burn - *flammability limits*
- Even if mixture is flammable, still won't burn in some environments
  - Small diameter tubes
  - Strong hydrodynamic strain or turbulence
  - High or low gravity
  - High or low pressure
- Understanding needed for combustion engines & industrial combustion processes (leaner mixtures $\Rightarrow$ lower T_{ad} $\Rightarrow$ lower NO_x); fire & explosion hazard management, fire suppression, ...
- Limits occur for mixtures that are *thermodynamically flammable* - theoretical adiabatic flame temperature (T_{ad}) far above ambient temperature (T_∞)
- Limits characterized by *finite (not zero) burning velocity at limit*
- Models of limits due to losses - most important prediction: burning velocity at the limit ($S_{L,lim}$) - better test of limit predictions than composition at limit

Premixed-gas flames – flammability limits



2 limit mechanisms, (1) & (2), yield similar fuel % and T_{ad} at limit but very different $S_{L,lim}$

Flammability limits in vertical tubes

- Most common apparatus - vertical tube (typ. 5 cm in diameter)
- Ignite mixture at one end of tube, if it propagates to other end, it's "flammable"
- Limit composition depends on orientation - *buoyancy effects*

Upward propagation

Downward propagation

Chemical kinetics of flammability limits

- Lean hydrocarbon-air flames: recall main branching reaction (promotes combustion) is (in units of moles, cm^2, s, cal)

$$H + O_2 \rightarrow OH + O;\ \frac{d[H]}{dt} = -10^{16.7}T^{-0.8}[H][O_2]e^{-16500/\Re T}$$

Depends on P^2 since [ ] ~ P, strongly dependent on T

- Why important? Only energetically viable way to break O=O bond (120 kcal/mole), even though [H] is small
- Main H consumption reaction

$$H + O_2 + M \rightarrow HO_2 + M;\ \frac{d[H]}{dt} = -10^{15.2}T^{0}[H][O_2][M]e^{+1000/\Re T}$$

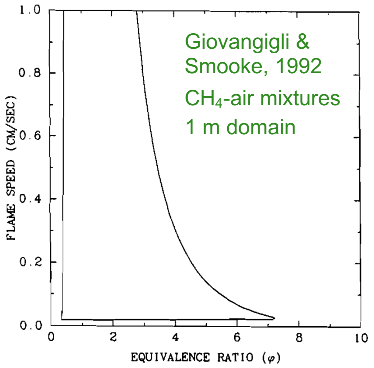
for M = N_2 (higher rate for CO_2 and especially H_2O)

Depends on P^3, nearly independent of T

- Why important? Inhibits combustion by replacing H with much less active HO_2
- Branching or inhibition may be faster depending on T and P

Chemical kinetics of flammability limits

- Rates equal ("crossover") when
  $[M] = 10^{1.5}T^{-0.8}e^{-17500/RT}$
- Ideal gas law: P = [M]RT thus
  $P = 10^{3.4}T^{0.2}e^{-17500/RT}$ (P in atm)
  $\Rightarrow$ crossover at 950K for 1 atm, higher T for higher P
- …but this only indicates that chemical mechanism may change and perhaps overall reaction rate ω will drop rapidly
- Computations show no limits without losses – no purely chemical criterion (Lakshmisha *et al.*, 1990; Giovangigli & Smooke, 1992) - for steady planar adiabatic flames, S_L decreases smoothly to zero as fuel conc. decreases (domain sizes up to 10 m, S_L down to 0.02 cm/s)
- …but as S_L decreases, δ increases - need larger computational domain or experimental apparatus
- Also more buoyancy & heat loss effects as S_L decreases ….



Giovangigli & Smooke, 1992

CH_4-air mixtures

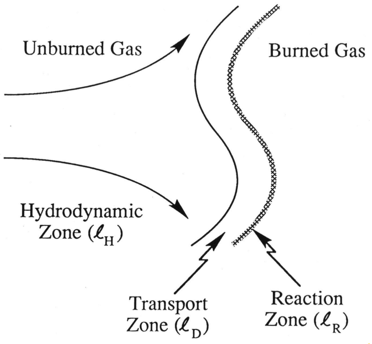
1 m domain

Aerodynamic effects on premixed flames

- Aerodynamic effects occur on a large scale compared to the transport or reaction zones but affect S_L and even existence of the flame
- Why only at large scale?
  - Re on flame scale ≈ $S_L\delta/\nu$ (ν = kinematic viscosity)
  - Re = $(S_L\delta/\alpha)(\alpha/\nu) = (1)(1/Pr) \approx 1$ since Pr ≈ 1 for gases
  - $Re_{flame} \approx 1 \Rightarrow$ viscosity suppresses flow disturbances
- Key parameter: stretch rate (Σ)

$$\Sigma \equiv \frac{1}{A}\frac{dA}{dt} \quad (A \;=\; \text{flame area})$$

- Generally Σ ~ U/d
  U = characteristic flow velocity
  d = characteristic flow length scale

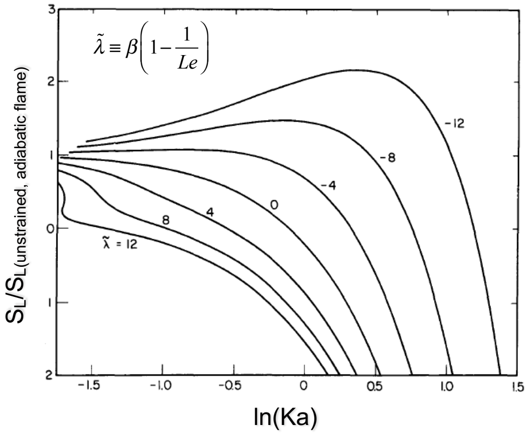


Aerodynamic effects on premixed flames

- Strong stretch ($\Sigma \geq \omega \sim S_L^2/\alpha$ or *Karlovitz number* Ka $\equiv \Sigma\alpha/S_L^2 \geq 1$) extinguishes flames
- Moderate stretch strengthens flames for Le < 1

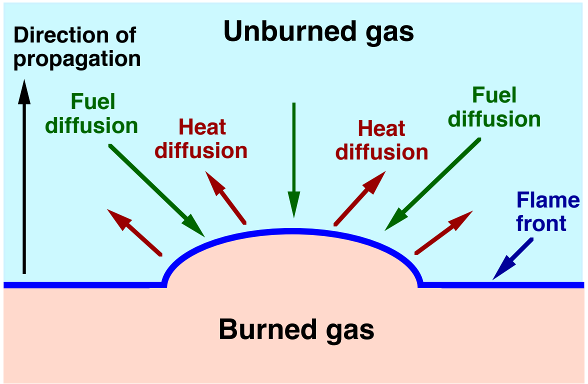
$$\text{Le} \equiv \frac{\text{Thermal diffusivity of the bulk mixture } (\alpha)}{\text{Mass diffusivity of scarce reactant into the bulk mixture (D)}}$$

Buckmaster & Mikolaitis, 1982a, cold reactants against adiabatic products



Lewis number tutorial

- Le affects flame temperature in curved (shown below) or stretched flames
- When Le < 1, additional thermal enthalpy loss in curved/stretched region is less than additional chemical enthalpy gain, thus local flame temperature in curved region is higher, thus reaction rate increases drastically, local burning velocity increases
- Opposite behavior for oppositely curved flames



Time scales - premixed-gas flames

- Chemical time scale

  $t_{chem} \approx \delta/S_L \approx (\alpha/S_L)/S_L \approx \alpha/S_L^2$
- Conduction time scale

  $t_{cond} \approx T_{ad}/(dT/dt) \approx d^2/16\alpha$

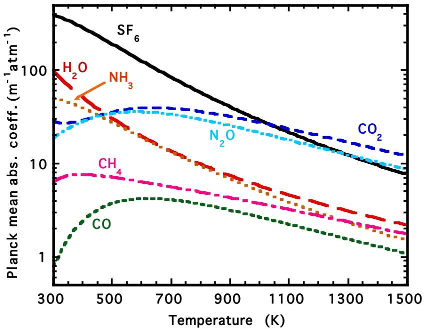
  d = tube or burner diameter
- Buoyant transport time scale

  $t \sim d/V;\ V \approx (gd(\Delta\rho/\rho))^{1/2} \approx (gd)^{1/2}$

  (g = gravity, d = characteristic dimension)

  Inviscid: $t_{inv} \approx d/(gd)^{1/2} \approx (d/g)^{1/2}$ $(1/t_{inv} \approx \Sigma_{inv})$

  Viscous: $d \approx \nu/V \Rightarrow\ t_{vis} \approx (\nu/g^2)^{1/3}$



- Radiation time scale

  $t_{rad} \approx T_{ad}/(dT/dt) \approx T_{ad}/(\Lambda/\rho C_p)$

  (Λ = radiative heat loss per unit volume)

  Optically thin radiation: $\Lambda = 4\sigma a_p(T_{ad}^4 - T_\infty^4)$

  a_p = Planck mean absorption coefficient [typ. 2 m^{-1} at 1 atm]

  $\Rightarrow \Lambda \approx 10^6$ W/m^3 for HC-air combustion products

  $\Rightarrow t_{rad} \sim P/\sigma a_p(T_{ad}^4 - T_\infty^4) \sim P^0$, P = pressure

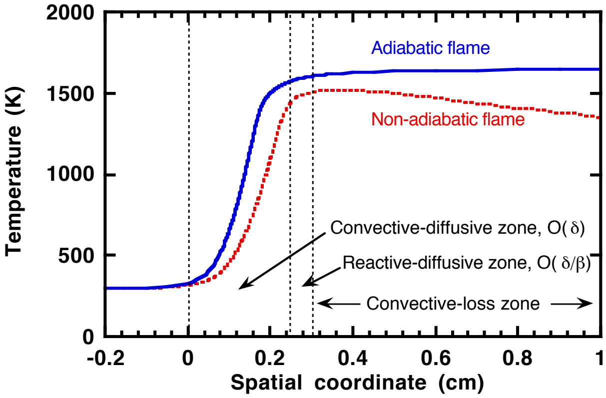
Time scales (hydrocarbon-air, 1 atm)

Time scale	Stoich. flame	Limit flame
Chemistry (t_{chem}) or diffusion (t_{diff})	0.00094 sec	0.25 sec
Buoyant, inviscid (t_{inv})	0.071 sec	0.071 sec
Buoyant, viscous (t_{vis})	0.012 sec	0.010 sec
Conduction (t_{cond}), d = 5 cm	0.95 sec	1.4 sec
Radiation (t_{rad})	0.13 sec	0.41 sec

- Conclusions
  - Buoyancy unimportant for near-stoichiometric flames (t_{inv} & $t_{vis} >> t_{chem}$)
  - Buoyancy strongly influences near-limit flames at 1g (t_{inv} & $t_{vis} < t_{chem}$)
  - Radiation effects unimportant at 1g ($t_{vis} << t_{rad}$; $t_{inv} << t_{rad}$)
  - \+ Radiation effects dominate flames with low S_L ($t_{rad} \approx t_{chem}$), but only observable at µg
  - Small t_{rad} (a few seconds) - drop towers useful
  - Radiation > conduction only for d > 3 cm
  - $Re \sim Vd/\nu \sim (gd^3/\nu^2)^{1/2} \Rightarrow$ turbulent flow at 1g for d > 10 cm

Flammability limits due to losses

- Golden rule: at limit $\dfrac{\text{Heat loss rate per unit volume}}{\text{Heat generation rate per unit volume}} \approx \dfrac{1}{\beta}$
- Why $1/\beta$ not 1? T can only drop by $O(1/\beta)$ before extinction - O(1) drop in T means exponentially large drop in reaction rate ω, thus exponentially small S_L (could also say heat generation occurs only in δ/β region whereas loss occurs over δ region)



Flammability limits due to losses

- Heat loss to walls
  - $t_{chem} \sim t_{cond} \Rightarrow S_{L,\lim} \approx (8\beta)^{1/2}\alpha/d$ at limit
    or $Pe_{\lim} \equiv S_{L,\lim}d/\alpha \approx (8\beta)^{1/2} \approx 9$
  - Actually $Pe_{\lim} \approx 40$ **(USE $Pe_{\lim} \approx 40$ NOT 9)** due to temperature averaging - consistent with experiments (Jarosinsky, 1983)
- Upward propagation in tube
  - Rise speed at limit $\approx 0.3(gd)^{1/2}$ due to buoyancy alone (same as air bubble rising in water-filled tube (Levy, 1965))
    $\Rightarrow Pe_{\lim} \approx 0.28\, Ra_d^{1/2}$; Ra_d = Rayleigh number $\equiv gd^3/\alpha\nu$
  - Causes stretch extinction (Buckmaster & Mikolaitis, 1982b):
    $t_{chem} \approx t_{inv}$ or $1/t_{chem} \approx \Sigma_{inv}$

$$\Rightarrow S_{L,\lim} = f(Le)\left(\frac{g\alpha^2}{d}\right)^{1/4}; f(Le) = \exp\left(\frac{\beta}{4}\left(1-\frac{T_\infty}{T_{ad}}\right)\left(1-\frac{1}{Le}\right)\right)$$

Note f(Le) < 1 for Le < 1, f(Le) > 1 for Le > 1 - flame can survive at lower S_L (weaker mixtures) when Le < 1

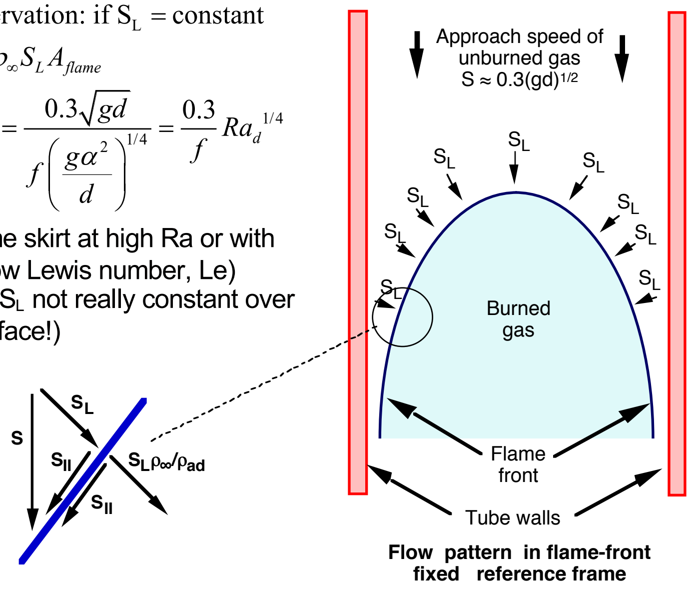
Difference between S and S_L

Mass conservation: if S_L = constant

$$\rho_\infty S A_{tube} = \rho_\infty S_L A_{flame}$$

$$\frac{A_{flame}}{A_{tube}} = \frac{S}{S_L} = \frac{0.3\sqrt{gd}}{f\left(\frac{g\alpha^2}{d}\right)^{1/4}} = \frac{0.3}{f} Ra_d^{1/4}$$

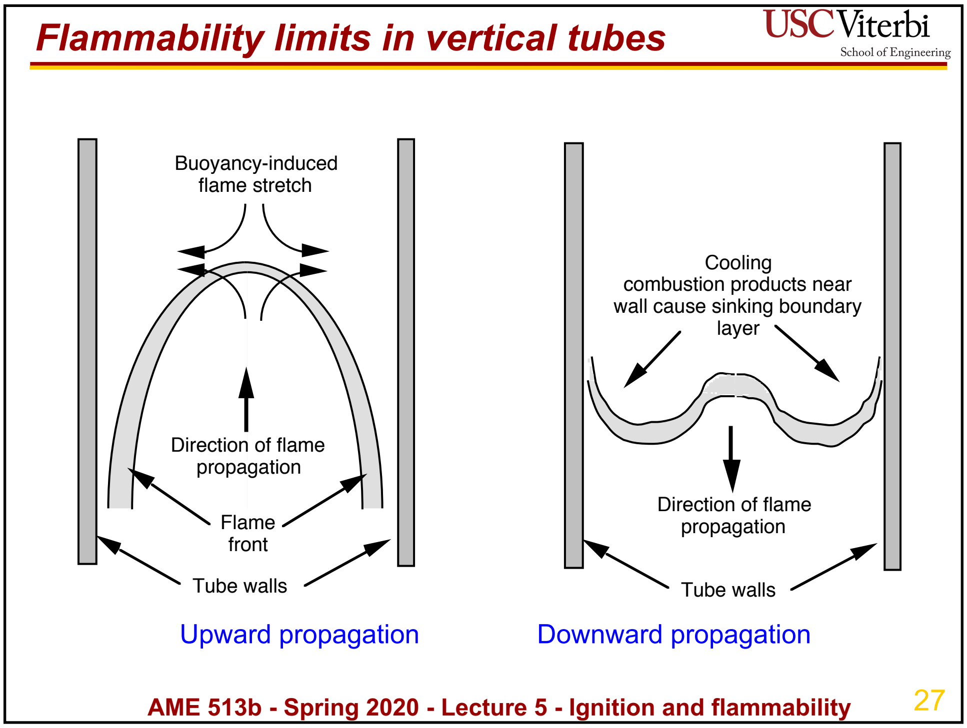
⇒ long flame skirt at high Ra or with small f (low Lewis number, Le) (but note S_L not really constant over flame surface!)



Flow pattern in flame-front fixed reference frame

Flammability limits in vertical tubes

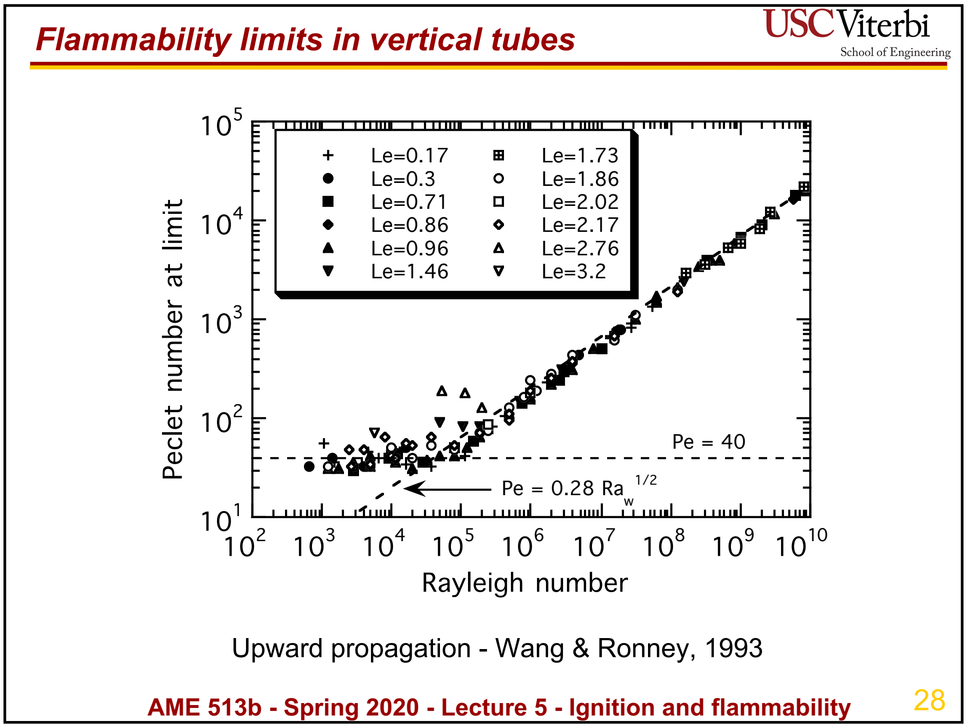
- Downward propagation – sinking layer of cooling gases near wall outruns & "suffocates" flame (Jarosinsky *et al.*, 1982)
  - $t_{chem} \approx t_{vis} \Rightarrow S_{L,lim} \approx 1.3(g\alpha)^{1/3}$
  - $Pe_{lim} \approx 1.7\ Ra_d^{1/3}$
  - Can also obtain this result by equating S_L to sink rate of thermal boundary layer = $0.8(gx)^{1/2}$ for $x = \delta$
  - Consistent with experiments varying d and α (by varying diluent gas and pressure) (Wang & Ronney, 1993) and g (using centrifuge) (Krivulin et al., 1981)



Flammability limits in vertical tubes

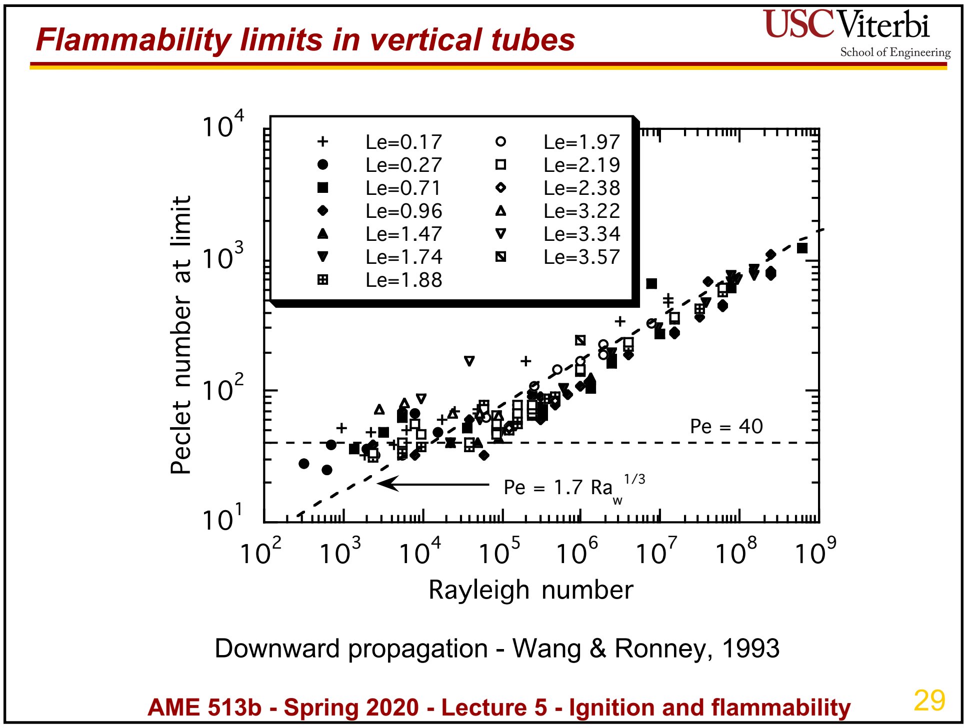
Upward propagation

Downward propagation



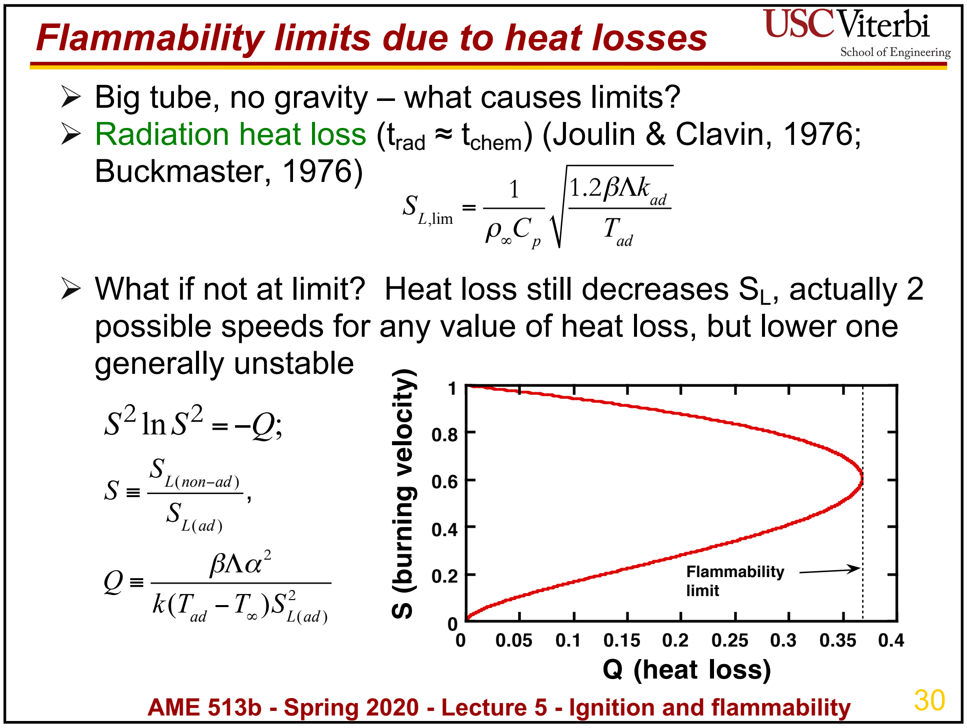
Flammability limits in vertical tubes

Upward propagation - Wang & Ronney, 1993



Flammability limits in vertical tubes

Downward propagation - Wang & Ronney, 1993



Flammability limits due to heat losses

- Big tube, no gravity – what causes limits?
- Radiation heat loss ($t_{rad} \approx t_{chem}$) (Joulin & Clavin, 1976; Buckmaster, 1976)

$$S_{L,\lim} = \frac{1}{\rho_\infty C_p}\sqrt{\frac{1.2\beta\Lambda k_{ad}}{T_{ad}}}$$

- What if not at limit? Heat loss still decreases S_L, actually 2 possible speeds for any value of heat loss, but lower one generally unstable

$$S^2 \ln S^2 = -Q;$$

$$S \equiv \frac{S_{L(non-ad)}}{S_{L(ad)}},$$

$$Q \equiv \frac{\beta\Lambda\alpha^2}{k(T_{ad}-T_\infty)S_{L(ad)}^2}$$

Flammability limits due to heat losses

- Doesn't radiative loss decrease for weaker mixtures, since temperature is lower? NO!

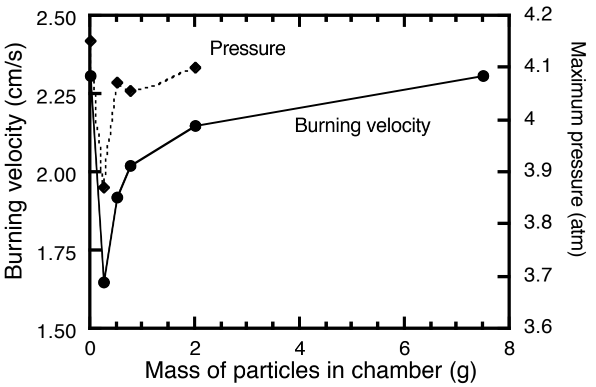
$$\text{Impact of heat loss} \sim \frac{\text{Heat loss rate}}{\text{Heat release rate}} \sim \frac{T^2}{e^{-E/\Re T}} \Uparrow \text{ as } T \Downarrow$$

- Predicted $S_{L,lim}$ (typically 2 cm/s) consistent with µg experiments (Ronney, 1988 [below]; Abbud-Madrid & Ronney, 1990)

Fuel	Pressure, Torr	Composition (see legend)	Estimated E_a kcal/mole	$S_{u,lim}$, calculated, cm/sec	$S_{u,lim}$, measured cm/sec
CH_4	1500	0.532	47.4	1.30	1.04
	760	0.513	43.6	1.73	1.47
	250	0.474	31.6	2.46	2.02
	100	0.441	27.8	3.48	2.80
	50	0.418	26.2	4.68	3.67
CH_4	760	0.25,54.7%	43.6	1.71	1.44
	760	0.75,81.5%	43.6	1.73	1.61
	760	0.88,83.2%	43.6	1.75	1.47
	760	1.00,83.6%	43.6	1.82	1.94
	760	1.20,79.6%	55.7	2.33	2.61
	760	1.50,73.4%	55.7	2.48	2.15
	760	2.00,62.5%	55.7	2.72	2.70

Radiation absorption effects

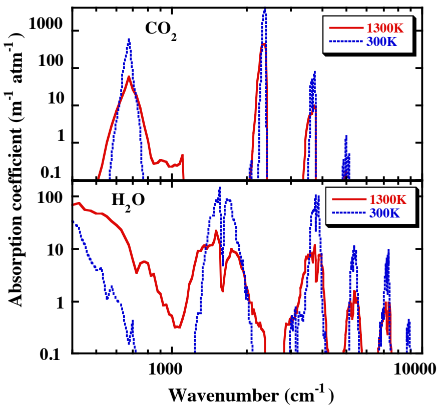
- Is radiation always a loss mechanism?
  - Reabsorption may be important when $a_P^{-1} < d$
  - Small concentration of blackbody particles - decreases S_L (more radiative loss)
  - More particles - reabsorption extend limits, increases S_L



Abbud-Madrid & Ronney (1993)

Radiation absorption effects

- Why do limits exist even when reabsorption effects are considered and the ambient mixture includes absorbers?
  - Spectra of product H_2O different from CO_2 (Mechanism I)
  - Spectra broader at high T than low T (Mechanism II)
  - Some radiation reaches upstream boundary due to "gaps" in spectra - product radiation that cannot be absorbed upstream
  - As a result, dramatic difference in S_L & limits compared to optically thin (Ju et al., 1998)



Absorption spectra of CO_2 & H_2O at 300K & 1300K

Modeling of reabsorption effects (Ju et al., 1998)

- CHEMKIN, steady planar 1D energy & species cons. equations
- 18-species, 58-step CH_4 oxidation mechanism (Kee *et al.*)
- Boundary conditions
  - Upstream - T = 300K, inflow velocity S_L at $x = L_1 = -30$ cm
  - Downstream - zero gradients of T & composition at $x = L_2 = 400$ cm
- Radiation model
  - CO_2, H_2O and CO; Wavenumbers (ω) 150 - 9300 cm^{-1}
  - Statistical Narrow-Band model for overlapping absorption lines (see Excel spreadsheet)
  - 300K black walls at upstream & downstream boundaries
- Mixtures $CH_4 + \{0.21O_2+(0.79-\gamma)N_2+ \gamma\ CO_2\}$ - substitute CO_2 for N_2 in "air" to assess effect of absorbing ambient
- Practical applications
  - Combustion at high pressures and in large furnaces
    - IC engines: 40 atm - Planck mean absorption length ≈ 4 cm for combustion products ≈ cylinder size
    - Furnaces - $L_P \approx 1.6$ m - comparable to boiler dimensions
  - Exhaust-gas recirculation - absorbing CO_2 & H_2O in unburned mixture

Radiation absorption effects - flame structure

USC Viterbi School of Engineering

- Adiabatic flame (no radiation)
  - The usual behavior
- Optically-thin
  - Volumetric loss always positive
  - Maximum T < adiabatic
  - T decreases "rapidly" in burned gases
  - "Small" preheat convection-diffusion zone - similar to adiabatic flame
- With reabsorption
  - Volumetric loss negative in reactants - indicates net heat transfer from products to reactants via reabsorption
  - Maximum T > adiabatic due to radiative preheating
  - T decreases "slowly" in burned gases - heat loss reduced
  - "Small" preheat convection-diffusion zone PLUS ”huge" convection-radiation preheat zone

Radiation absorption effects - flame structures

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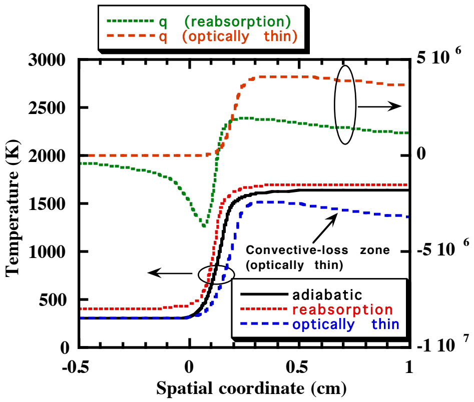
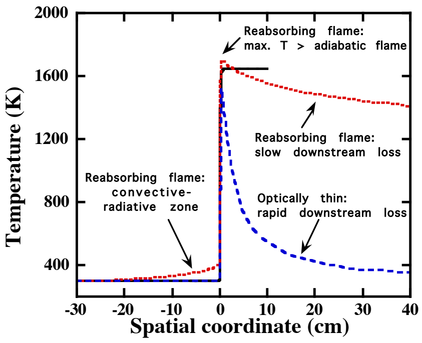


Flame zone detail

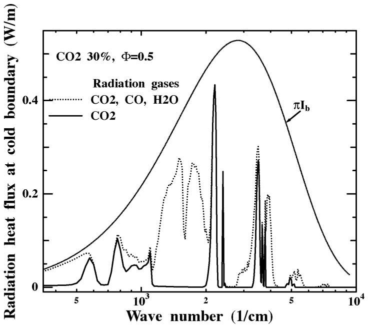


Radiation zones (large scale)

Mixture: CH_4 in "air", 1 atm, equivalence ratio (ϕ) = 0.70; γ = 0.30 ("air" = 0.21 O_2 + .49 N_2 + .30 CO_2)

Radiation absorption effects - spectra

- Flux at upstream boundary shows spectral regions where radiation can escape - "gaps" due to mismatch between radiation emitted at the flame front and that which can be absorbed by the reactants
- Depends on "discontinuity" (as seen by radiation) in T and composition at flame front - doesn't apply to downstream radiation because T gradient is small
- Behavior cannot be predicted via simple mean absorption coefficients - critically dependent on compositional & temperature dependence of spectra



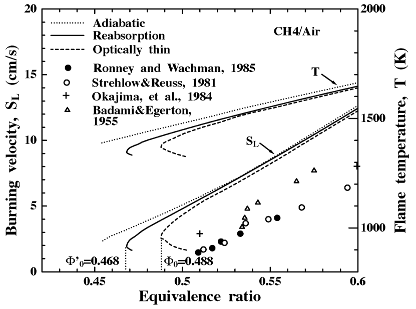
Spectrally-resolved radiative flux at upstream boundary for a reabsorbing flame

(πI_b = maximum possible flux)

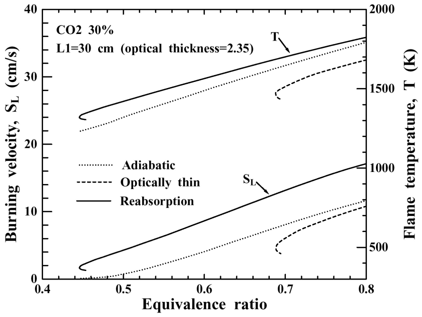
Reabsorption effects - burning velocities

- CH_4-air (γ = 0)
  - Minor differences between reabsorption & optically-thin
  - ... but $S_{L,lim}$ 25% lower with reabsorption; since $S_{L,lim}$ ~ (radiative loss)$^{1/2}$, if net loss halved, then $S_{L,lim}$ should be $1 - 1/\sqrt{2}$ = 29% lower with reabsorption
  - $S_{L,lim}/S_{L,ad} \approx 0.6$ for both optically-thin and reabsorption models - close to theoretical prediction ($e^{-1/2}$)
  - Interpretation: reabsorption eliminates downstream heat loss, no effect on upstream loss (no absorbers upstream); classical quenching mechanism still applies
  - All experiments lie below predictions - are published chemical mechanisms accurate for very lean mixtures?
- γ = 0.30 (38% of N_2 replaced by CO_2)
  - Massive effect of reabsorption
  - S_L much higher with reabsorption than with no radiation!
  - Lean limit much leaner (ϕ = 0.44) than with optically-thin radiation (ϕ = 0.68)

Reabsorption effects - burning velocities



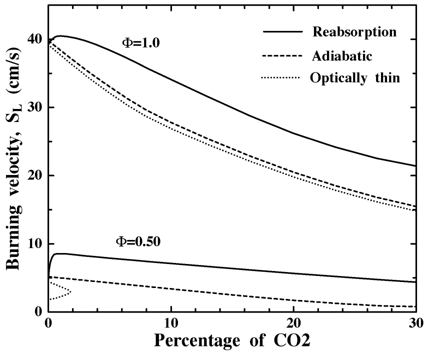
γ = 0 (no CO_2 in ambient)



γ = 0.30

Reabsorption effects of γ (CO_2 substitution)

- ϕ = 1.0: little effect of radiation;
- ϕ = 0.5: dominant effect - why?
  - (1) ϕ = 0.5: close to radiative extinction limit - large benefit of decreased heat loss due to reabsorption by CO_2
  - (2) ϕ = 0.5: much larger Boltzman number (defined below) (B) (≈127) than ϕ = 1.0 (≈11.3); B ~ potential for radiative preheating to increase S_L
- Note with reabsorption, only 1% CO_2 addition nearly doubles S_L due to much lower net heat loss!



Effect of CO_2 substitution for N_2 on S_L

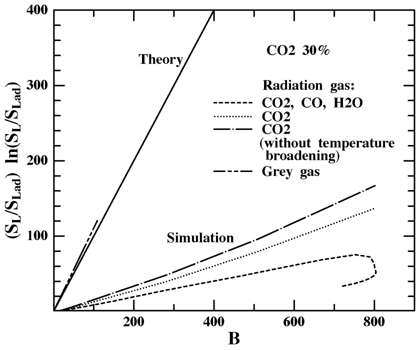
$$B = \frac{\text{Blackbody radiative heat flux at } T_{ad}}{\text{Convective enthalpy flux through flame front}}\frac{\partial \ln(S_L)}{\partial \ln(T_{ad})} = \frac{\sigma\left(T_{ad}^4 - T_\infty^4\right)}{\rho_\infty S_{L,ad} C_P T_{ad}}\frac{\beta}{2}$$

Reabsorption - comparison to analytic theory

- Joulin & Deshaies (1986) - analytical theory

$$\left(\frac{S_L}{S_{L,ad}}\right)\ln\left(\frac{S_L}{S_{L,ad}}\right) = B$$

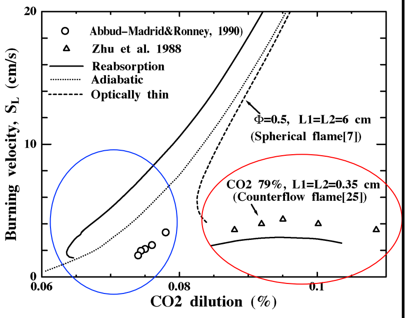
- Comparison to computation - poor
- Better without H_2O radiation (mechanism (I) suppressed)
- Slightly better still without T broadening (mechanism (II) suppressed, nearly adiabatic)
- Good agreement when $L(\omega) = L_P$ = constant - emission & absorption across entire spectrum rather than just certain narrow bands.
- Drastic differences between last two cases, even though both have no net heat loss and have same Planck mean absorption lengths!



Effect of different radiation models on S_L and comparison to theory

Reabsorption - comparison with experiment

- No directly comparable expts., BUT…
- Zhu, Egolfopoulos, Law (1988)
  - $CH_4 + (0.21O_2 + 0.79\ CO_2)$ ($\gamma = 0.79$)
  - Counterflow twin flames, extrapolated to zero strain
  - $L_1 = L_2 \approx 0.35$ cm chosen since 0.7 cm from nozzle to stagnation plane
  - No solutions for adiabatic flame or optically-thin radiation (!)
  - Moderate agreement with reabsorption
- Abbud-Madrid & Ronney (1990)
  - $(CH_4 + 4O_2) + CO_2$
  - Expanding spherical flame at μg
  - $L_1 = L_2 \approx 6$ cm chosen (≈ flame radius)
  - Optically-thin model over-predicts limit fuel conc. & $S_{L,lim}$
  - Reabsorption model underpredicts limit fuel conc. but $S_{L,lim}$ well predicted - net loss correctly calculated



Comparison of computed results to experiments where reabsorption effects may have been important

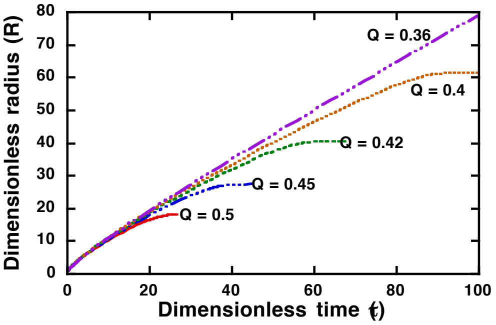
Combined stretched & heat loss

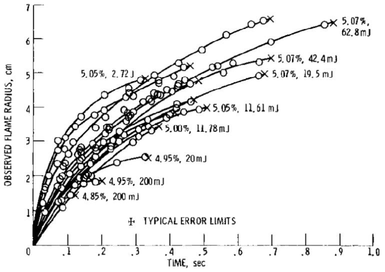
- Spherical expanding flames, Le < 1: stretch allows flames to exist in mixtures below radiative limit until flame radius r_f is too large & curvature benefit too weak (Ronney & Sivashinsky, 1989)

$$\Sigma \equiv \frac{1}{A}\frac{dA}{dt} = \frac{1}{4\pi r_f^2}\frac{d}{dt}\left(4\pi r_f^2\right) = \frac{2}{r_f}\frac{dr_f}{dt} \Rightarrow \frac{dS}{dR} + S^2 \ln S^2 = \frac{2S}{R} - Q$$

- Adds stretch term (2S/R) (R = scaled flame radius; R > 0 for Le < 1; R < 0 for Le > 1) and unsteady term (dS/dR) to planar steady equation
- Dual limit: radiation at large r_f, curvature-induced stretch at small r_f (ignition limit)

Combined stretched & heat loss





Theory (Ronney & Sivashinsky, 1989)

Experiment
(Ronney, 1985)

More on flammability limits in tubes

- Experiments show that the flammability limits are wider for upward than downward propagation, corresponding to $S_{L,lim,down} > S_{L,lim,up}$ since S_L is lower for more dilute mixtures
- …but note according to the models, $S_{L,lim,down} > S_{L,lim,up}$ when
  $Ra < 10{,}000\ f^{12}$
- but also need Pe > 40 (not in heat-loss limit)
  $Ra > 18{,}000$
  $\Rightarrow$ at high Le (high f) & $18{,}000 < Ra < 10{,}000\ f^{12}$, upward limits may be narrower than downward limits (?!?)
- Never observed, but appropriate conditions never tested - high Le, moderate Ra

Turbulent limit behavior?

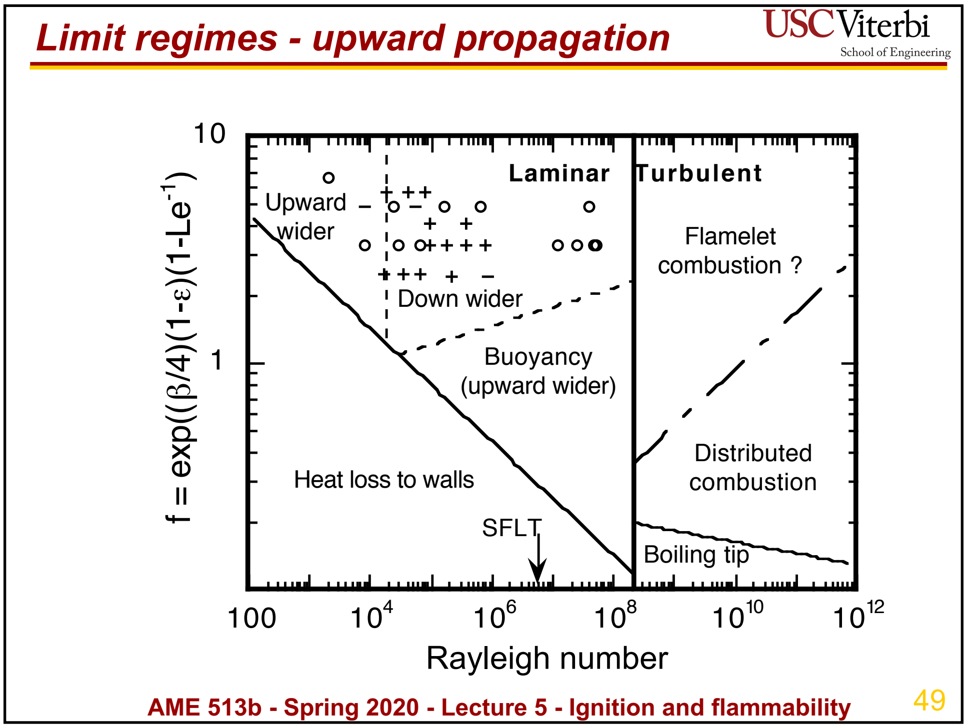
- Burned gases are turbulent if Re > 2000
  - Upward limit: $Re \approx S(\rho_\infty/\rho_{ad}-1)d/\nu \Rightarrow Ra > 300 \times 10^6$
  - Downward limit: $Re \approx S_L(\rho_\infty/\rho_{ad}-1)d/\nu \Rightarrow Ra > 40 \times 10^9$ - not accessible with current apparatus
- "Standard" condition (5 cm tube, air, 1 atm):
  - $Ra \approx 3.0 \times 10^6$: always laminar!

Approach

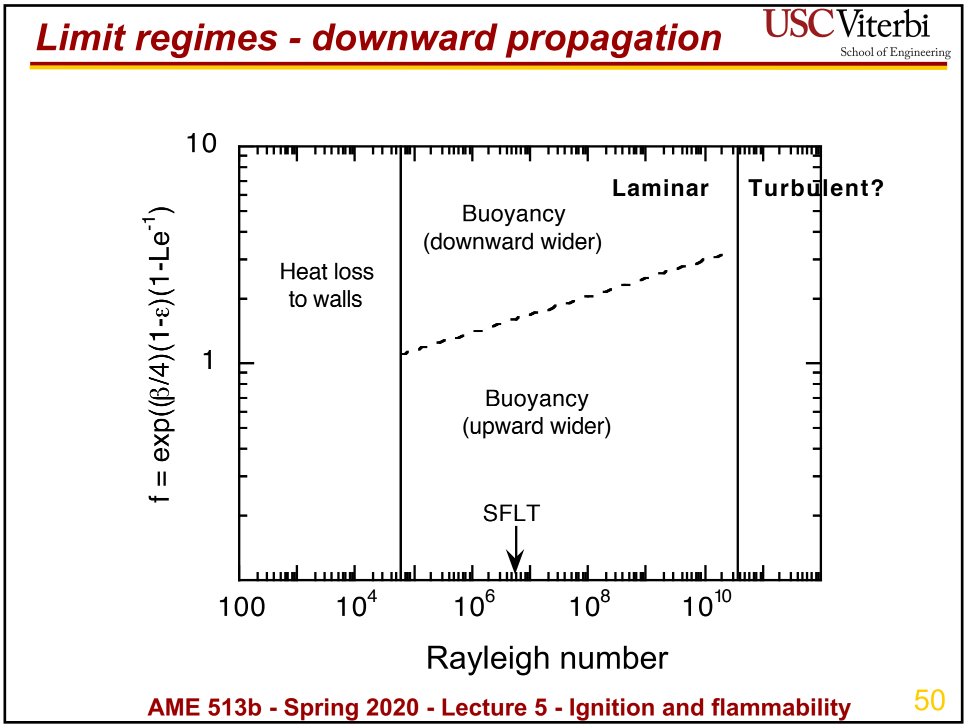
- Study limit mechanisms by measuring $S_{b,lim}$ for varying
  - Tube diameter
  - $\alpha = \alpha$(diluent, pressure)
  - Le ≡ = Le(diluent, fuel)
  - and determine scaling relations (Pe_{lim} vs. Ra & Le)
- Apparatus
  - Tubes with $0.5 \text{ cm} < D < 20 \text{ cm}$; open at ignition end
  - He, Ne, N_2, CO_2, SF_6 diluents
  - $0.1 \text{ atm} < P < 10 \text{ atm}$
  - $2 \times 10^2 < Ra < 2 \times 10^9$
  - Absorption tank to maintain constant P during test
  - Thermocouples
- Procedure
  - Fixed fuel:O_2 ratio
  - Vary diluent conc. until limit determined
  - Measure $S_{b,lim}$ & temperature characteristics at limit

Results - laminar flames

- Upward limit
  - Low Ra
    - » $Pe_{lim} \approx 40 \pm 10$ at low Ra
    - » Highest T near centerline of tube
  - High Ra
    - » $Pe_{lim} \approx 0.3 Ra^{1/2}$ at high Ra
    - » Highest T near centerline (low Le)
    - » Highest T near wall (high Le)
    - » Indicates strain effects at limit
- Downward
  - $Pe_{lim} \approx 40 \pm 10$ at low Ra
  - $Pe_{lim} \approx 1.5 Ra^{1/3}$ at high Ra
- Upward limits narrower than downward limits at high Le & moderate Ra, e.g. lean C_3H_8-O_2-Ne, P = 1 atm, D = 2.5 cm, Le ≈ 2.6, Ra ≈ 19,000: fuel up / fuel down ≈ 0.83



Limit regimes - upward propagation



Limit regimes - downward propagation

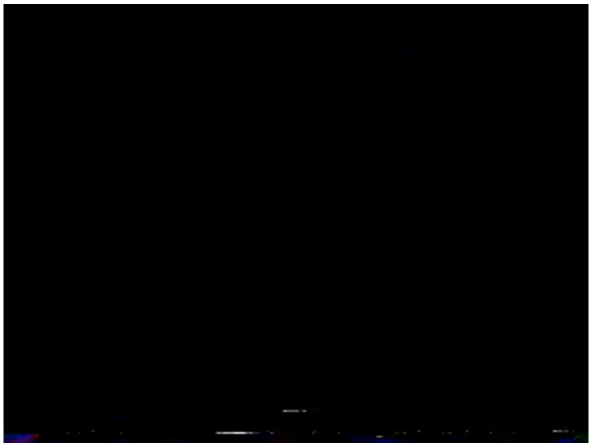
Flamelet vs. distributed combustion

- Abdel-Gayed & Bradley (1989): distributed if $Ka > 0.3$
  $Ka \equiv 0.157\ Re_T^{-1/2} U^2$; $Re_T \equiv u'L_I/\nu$, $U \equiv u'/S_L$
  $L_I \equiv$ integral scale of turbulence
- Estimate for pipe flow
  - $u' \approx 0.05S(\rho_\infty/\rho_{ad}-1)$; $L_I \approx d$
  - $S_{L,lim}$ from Buckmaster & Mikolaitis (1982) model
  - $\Rightarrow Ka \approx 0.0018/f^2\ Ra^{1/4} \approx 0.3/f^2$ at $Ra = 700 \times 10^6$
  - Distributed combustion probable at high Ra, moderate Le
  - Away from limit - wrinkled, unsteady skirt

Limit flame - distributed combustion

C_3H_8-O_2-CO_2, P = 2.5 atm, d = 10 cm, $Le \approx 1.3$, $Ra \approx 6 \times 10^8$

Farther from limit - wrinkled skirt



C_3H_8-O_2-CO_2, P = 2.5 atm, d = 10 cm, Le ≈ 1.3, Ra ≈ 6 x 10^8

Lower Le - boiling tip, no tip opening

C_3H_8-O_2-SF_6, P = 2.5 atm, d = 10 cm, Le ≈ 0.7, Ra ≈ 5 x 10^9

Turbulent flame quenching

- Why does distributed flame exist at $\delta \approx 4d$, whereas laminar flame extinguishes when $\delta \approx 1/40\ d$ (Pe = 40)?
- Analysis
  - $Nu = hd/k \approx 0.023\ Re^{.8}\ Pr^{.3}$ (turbulent heat transfer in pipe)
  - $Q_{loss} \approx hA\Delta T$; $A = \pi d\delta$; let $\delta = n\ D$ (n is unknown)
  - $Q_{gen} \approx \rho_o S_b \pi d^2 C_p \Delta T$; $S_b = 0.3(gd)^{1/2}$
  - $Q_{loss}/Q_{gen} \approx 1/\beta$ at quenching limit
  - $\Rightarrow$ $n \approx 5Gr^{0.1}/\beta$ at quenching limit
- $Gr = 600 \times 10^6$, $\beta = 10 \Rightarrow n = 3.9$ at limit !!!
- But low Le $\Rightarrow S_L$ low at tip opening $\Rightarrow n > 4$ at tip opening $\Rightarrow$ distributed flame not observable

Conclusions

- Probable heat loss & buoyancy-induced limit mechanisms observed
- Limit behavior characterized mainly by Lewis & Rayleigh numbers
- Scaling analyses useful for gaining insight
- Transition to turbulence & distributed-like combustion observed
- High-Ra results may be more applicable to "real" hazards (large systems, turbulent) than classical experiments at low Ra

References

Abbud-Madrid, A., Ronney, P. D., "Effects of Radiative and Diffusive Transport Processes on Premixed Flames Near Flammability Limits," *Twenty Third Symposium (International) on Combustion*, Combustion Institute, 1990, pp. 423-431.

Abbud-Madrid, A., Ronney, P. D., "Premixed Flame Propagation in an Optically-Thick Gas," *AIAA Journal,* Vol. 31, pp. 2179-2181 (1993).

Ballal, D. R., Lefebvre, A. H., "The influence of flow parameters on minimum ignition energy and quenching distance," *15th Symposium (International) on Combustion*, Combustion Institute, 1975, pp. 1473-1481.

Buckmaster, J. D. (1976). The quenching of deflagration waves, *Combust. Flame* 26, 151 -162.

Buckmaster, J. D., Mikolaitis, D. (1982a). The premixed flame in a counterflow, *Combust. Flame* 47, 191-204 .

Buckmaster, J. D., Mikolaitis, D. (1982b). A flammability-limit model upward propagation through lean methan-air mixtures in a standard flammability tube. *Combust. Flame* 45, pp 109-119.

De Soete, G. G., *20th Symposium (International) on Combustion*, Combustion Institute, 1984, p. 161.

Dixon-Lewis, G., Shepard, I. G., *15th Symposium (International) on Combustion*, Combustion Institute, 1974, p. 1483.

Frendi, A., Sibulkin, M., "Dependence of Minimum Ignition Energy on Ignition Parameters," *Combust. Sci. Tech.* 73, 395-413, 1990.

Joulin, G., *Combust. Sci. Tech.* 43, 99 (1985).

References

Giovangigli, V. and Smooke, M. (1992). Application of Continuation Methods to Plane Premixed Laminar Flames, *Combust. Sci. Tech.* 87, 241-256.

Guo, H., Ju, Y., Maruta, K., Niioka, T., Liu, F., *Combust. Flame* 109:639-646 (1997).

Jarosinsky, J. (1983). Flame quenching by a cold wall, *Combust. Flame* 50, 167.

Jarosinsky, J., Strehlow, R. A., Azarbarzin, A. (1982). The mechanisms of lean limit extinguishment of an upward and downward propagating flame in a standard flammability tube, *Proc. Combust. Inst.* 19, 1549-1557.

Joulin, G., Clavin, P. (1976). Analyse asymptotique des conditions d'extinction des flammes laminaries, *Acta Astronautica* 3, 223.

Ju, Y., Masuya, G. and Ronney, P. D., "Effects of Radiative Emission and Absorption on the Propagation and Extinction of Premixed Gas Flames" *Twenty-Seventh International Symposium on Combustion*, Combustion Institute, 1998, pp. 2619-2626.

Ju, Y., Guo, H., Liu, F., Maruta, K. (1999). Effects of the Lewis number and radiative heat loss on the bifurcation of extinction of CH_4-O_2-N_2-He flames, *J. Fluid Mech.* 379, 165-190.

Krivulin, V. N., Kudryavtsev, E. A., Baratov, A. N., Badalyan, A. M., Babkin, V. S. (1981). Effect of acceleration on the limits of propagation of homogeneous gas mixtures, *Combust. Expl. Shock Waves (Engl. Transl.)* 17, 37-41.

Lakshmisha, K. N., Paul, P. J., Mukunda, H. S. (1990). On the flammability limit and heat loss in flames with detailed chemistry, *Proc. Combust. Inst.* 23, 433-440.

Levy, A. (1965). An optical study of flammability limits, *Proc. Roy. Soc. (London)* A283, 134.

References

Kingdon, R. G., Weinberg, F. J., *16th Symposium (International) on Combustion*, Combustion Institute, 1976, p. 747-756.

Kono, M., Kumagai, S., Sakai, T., *16th Symposium (International) on Combustion*, Combustion Institute, 1976, p. 757.

Kono, M., Hatori, K., Iinuma, K., *20th Symposium (International) on Combustion*, Combustion Institute, 1984, p. 133.

Lewis, B., von Elbe, G., *Combustion, Flames, and Explosions of Gases, 3rd ed.*, Academic Press, 1987.

Lim, E. H., McIlroy, A., Ronney, P. D., Syage, J. A., in: Transport Phenomena in Combustion (S. H. Chan, Ed.), Taylor and Francis, 1996, pp. 176-184.

Ronney, P.D., "Effect of Gravity on Laminar Premixed Gas Combustion II: Ignition and Extinction Phenomena," *Combustion and Flame*, Vol. 62, pp. 120-132 (1985).

Ronney, P.D., "On the Mechanisms of Flame Propagation Limits and Extinction Processes at Microgravity," *Twenty Second Symposium (International) on Combustion*, Combustion Institute, 1988, pp. 1615-1623.

Ronney, P. D., "Understanding Combustion Processes Through Microgravity Research," *Twenty-Seventh International Symposium on Combustion*, Combustion Institute, Pittsburgh, 1998, pp. 2485-2506

Ronney, P.D., Sivashinsky, G.I., "A Theoretical Study of Propagation and Extinction of Nonsteady Spherical Flame Fronts," *SIAM Journal on Applied Mathematics*, Vol. 49, pp. 1029-1046 (1989).

Sloane, T. M., Ronney, P. D., "A Comparison of Ignition Phenomena Modeled with Detailed and Simplified Kinetics," *Combustion Science and Technology*, Vol. 88, pp. 1-13 (1993).

References

Tromans, P. S., Furzeland, R. M., *21st Symposium (International) on Combustion*, Combustion Institute, 1986, p. 1891.

Wang, Q., Ronney, P. D. (1993). Mechanisms of flame propagation limits in vertical tubes, Paper no. 45, Spring Technical Meeting, Combustion Institute, Eastern/Central States Section, March 15-17, 1993, New Orleans, LA.