LEAN HYDROGEN FLAME DEFLAGRATION VELOCITIES AND FLAMMABILITY LIMITS

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General Basic Equations
Measured and Calculated Deflagration Velocities
Observations of Cellular Flames
Flame Balls
Influences of Soret Diffusion
The Lean Flammability Limit
Flame-Ball Deflagration Model
Approaches to Turbulent Combustion

REACTING NAVIER-STOKES CONSERVATION EQUATIONS

$$\partial\rho/\partial t + \nabla\cdot(\rho\nu) = 0.$$

$$\partial\nu/\partial t + \nu\cdot\nabla\nu = -(\nabla\cdot \mathrm{P})/\rho + \sum_{i=1}^{N} Y_i f_i$$

$$\rho\frac{\partial}{\partial t}\left(h + \frac{1}{2}v^2\right) + \rho v\cdot\nabla\left(h + \frac{1}{2}v^2\right) = \frac{\partial p}{\partial t} + \nabla\cdot\left[(pU - \mathrm{P})\cdot v\right] + \rho\sum_{i=1}^{N} Y_i f_i\cdot(v + V_i) - \nabla\cdot q.$$

$$\partial Y_i/\partial t + v\cdot\nabla Y_i = w_i/\rho - \left[\nabla\cdot(\rho Y_i V_i)\right]/\rho$$

$$P = \left[p + \left(\frac{2}{3}\mu - \kappa \right)(\nabla \cdot v) \right] U - \mu \left[(\nabla v) + (\nabla v)^T \right]$$

$$q = -\lambda \nabla T + \rho \sum_{i=1}^{N} h_i Y_i V_i + R^0 T \sum_{i=1}^{N} \sum_{j=1}^{N} \left(\frac{X_j D_{Ti}}{W_i D_{ij}} \right) (V_i - V_j) + q_R$$

$$\nabla X_i = \sum_{j=1}^{N} \left(\frac{X_i X_j}{D_{ij}} \right) (V_j - V_i) + (Y_i - X_i) \left(\frac{\nabla p}{p} \right) + \frac{\rho}{p} \sum_{j=1}^{N} Y_i Y_i (f_i - f_j)$$

$$+ \sum_{j=1}^{N} \left[\left(\frac{X_i X_j}{\rho D_{ij}} \right) \left(\frac{D_{Tj}}{Y_j} - \frac{D_{Ti}}{Y_i} \right) \right] \frac{\nabla T}{T}, \quad i = 1, \ldots, N.$$

$$w_i = W_i \sum_{k=1}^{M} \left(\nu''_{ik} - \nu'_{ik} \right) B_k T^{\alpha_k} e^{-\left(E_k / R^0 T \right)} \sum_{j=1}^{N} \left(\frac{X_j p}{R^0 T} \right)^{\nu'_{j,k}}, \quad i = 1, \ldots, N$$

$$p = \rho R^0 T \sum_{i=1}^{N} \left(Y_i / W_i \right)$$

$$h = \sum_{i=1}^{N} h_i Y_i$$

$$h_i = h_i^o + \int_{T^0}^{T} c_{p,i} dT, \quad i = 1, \ldots, N$$

$$X_i = \frac{\left(Y_i / W_i \right)}{\sum_{j=1}^{N} \left(Y_j / W_j \right)}, \quad i = 1, \ldots, N.$$

Symbols	Meaning
B_k	A constant in the frequency factor for the kth reaction*
c_{pi}	Specific heat at constant pressure for species i*
D_{ij}	Binary diffusion coefficient for species i and j*
D_{Ti}	Thermal diffusion coefficient for species i*
E_k	Activation energy for the kth reaction*
f_i	External force per unit mass on species i*
h	Enthalpy per unit mass for the gas mixture†
h_i	Specific enthalpy of species i†
h_i^o	Standard heat of formation per unit mass for species i at temperature T^o*
M	Total number of chemical reactions occurring*
N	Total number of chemical species present*
p	Hydrostatic pressure†
P	Stress tensor†
q	Heat-flux vector†
q_R	Radiant heat-flux vector*
R^0	Universal gas constant*
T	Temperature†
T^0	A fixed, standard reference temperature*
Vi	Diffusion velocity of species i†
v	Mass-average velocity of the gas mixture†
W_i	Molecular weight of species i*
w_i	Rate of production of species i by chemical reactions (mass per unit volume per unit time)†
X_i	Mole fraction of species i†
Y_i	Mass fraction of species i†
α_k	Exponent determining the temperature dependence of the frequency factor for the kth reaction*
κ	Bulk viscosity coefficient*
λ	Thermal conductivity*
μ	Coefficient for (shear) viscosity*
ν'_{ik}	Stoichiometric coefficient for species i appearing as a reactant in reaction k*
ν''_{ik}	Stoichiometric coefficient for species i appearing as a product in reaction k*
ρ	Density†

* Parameters that must be given in order to solve the conservation equations.
† Quantities determined by the equations given here.

BURNING-VELOCITY EXPERIMENTS

(Most accurate results are most recent, 1990 or later.)

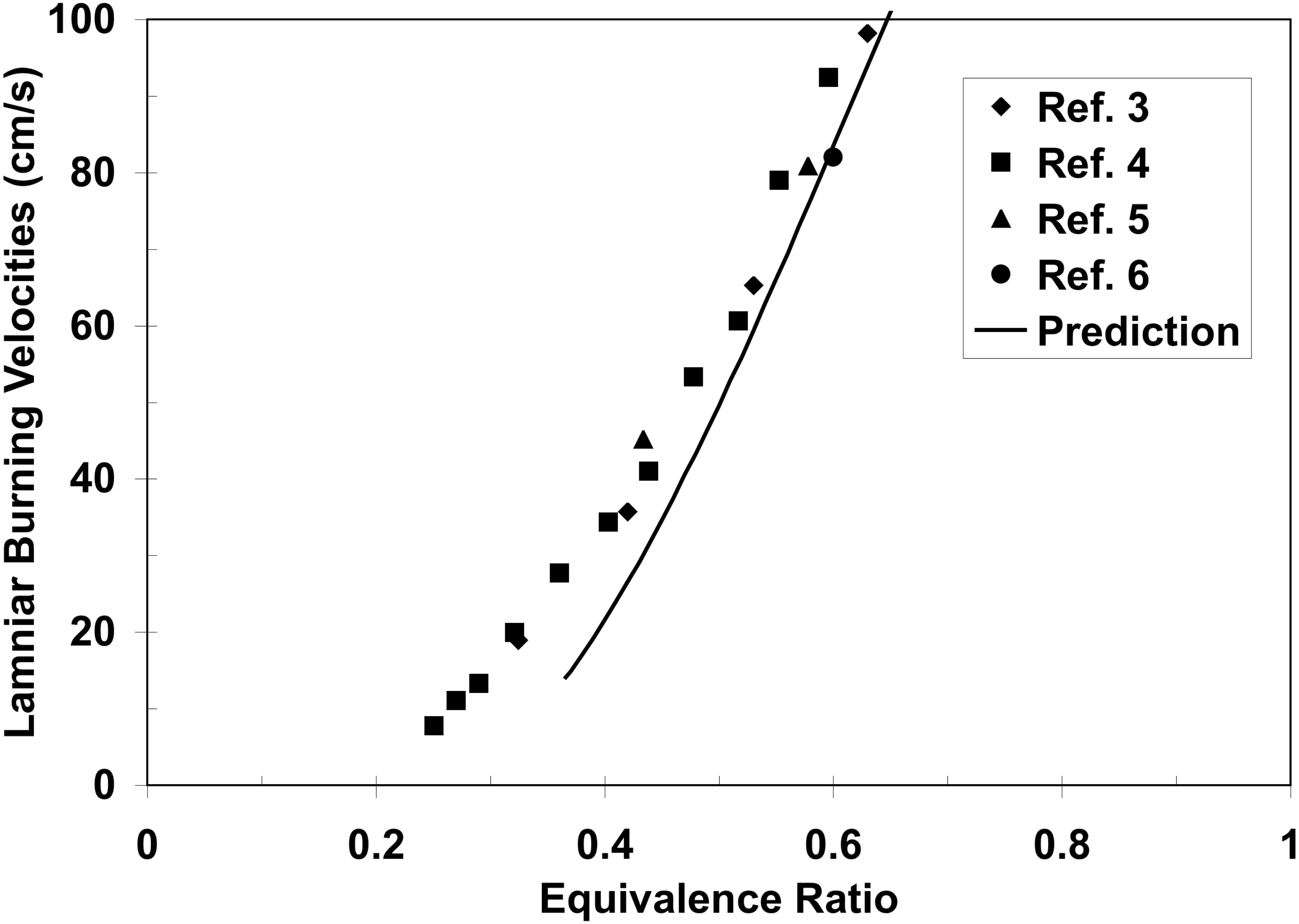
Spherically Expanding Flames:
- British Gas with Alan Williams, 1990 (#3).
- Steve Tse et al, 2000 (#5).
- Gerry Faeth's group, 2001 (#6).

Back-to-Back Counterflow Flames:
- Fokion and Ed, 1990 (#4).

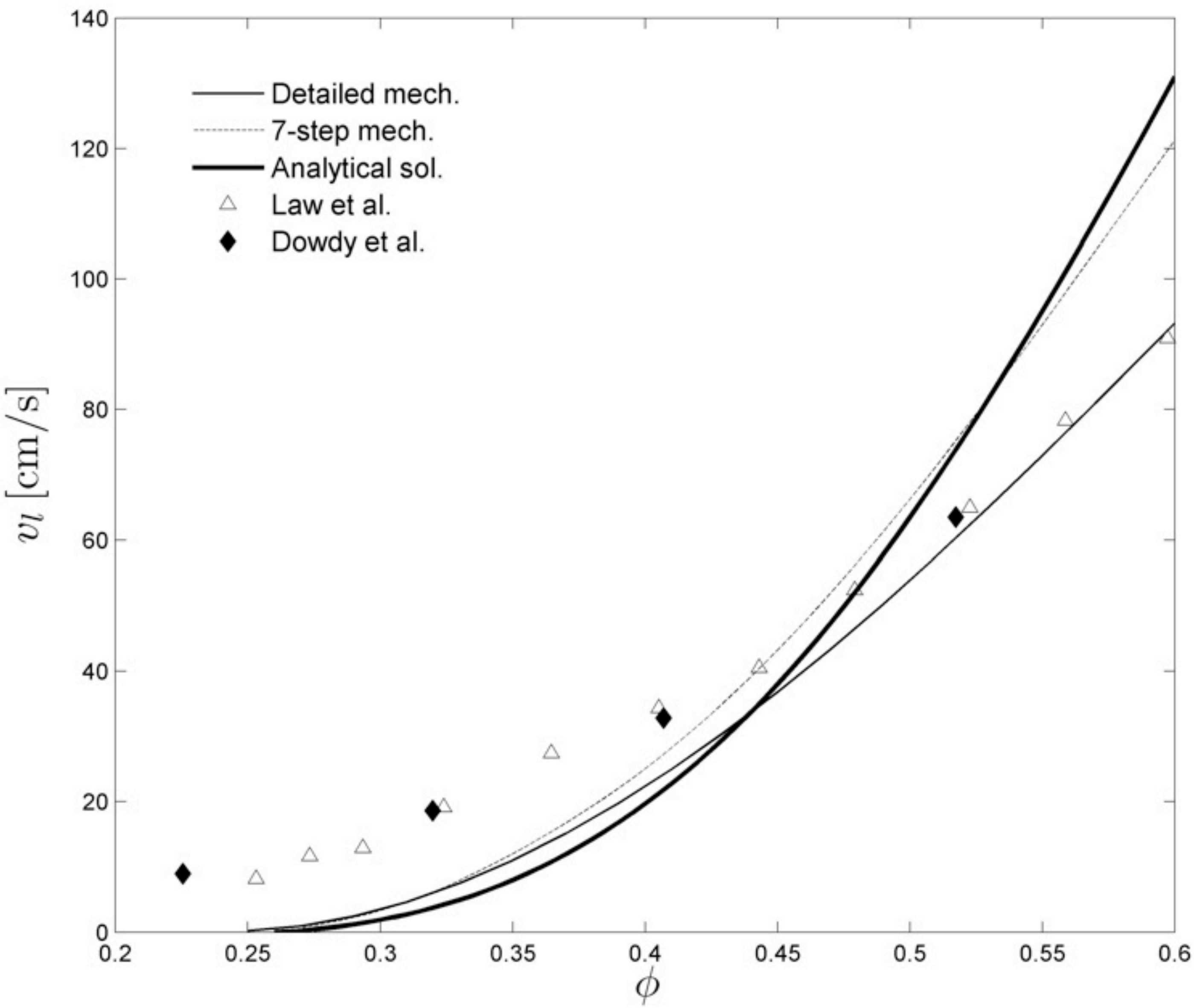
Spherical-flame authors comment on cell formation. Counterflow said to be perfectly flat, but preferential transverse diffusion may still occur.

Experiments all agree.

LEAN-FLAME DISAGREEMENTS



COMPARISONS WITH EXPERIMENT



COMPUTATIONAL AND EXPERIMENTAL EVIDENCE FOR CELLS

Computations performed without cross transport (Soret, Dufour) or radiation, using GRI 2.11, expected under these conditions to give results qualitatively similar to San Diego Mech, employed for previous comparison with experiment.

Cell-like flame caps seen for upward propagation already in 1914 by Coward & Brinsley, in determining standard limits for upward (4%) and downward (11%) propagation, and cell-like fingers observed later for downward propagation.

Lean-cell existence thus is well established.

2D DNS Method

Computationally efficient simulations based on:

1. Adaptive mesh refinement (AMR) resolves flame structures on large computational domains:
   - 4.5 × 9.0 cm and 7.5 × 12.0 cm in the present cases
   - 117 micron resolution for flame zone ≈1 mm thick

2. Low Mach-number equations allow large time steps:
   - simulation periods sufficient to see long-term behavior
   - 10 to 12 seconds of fluid-chemistry interaction

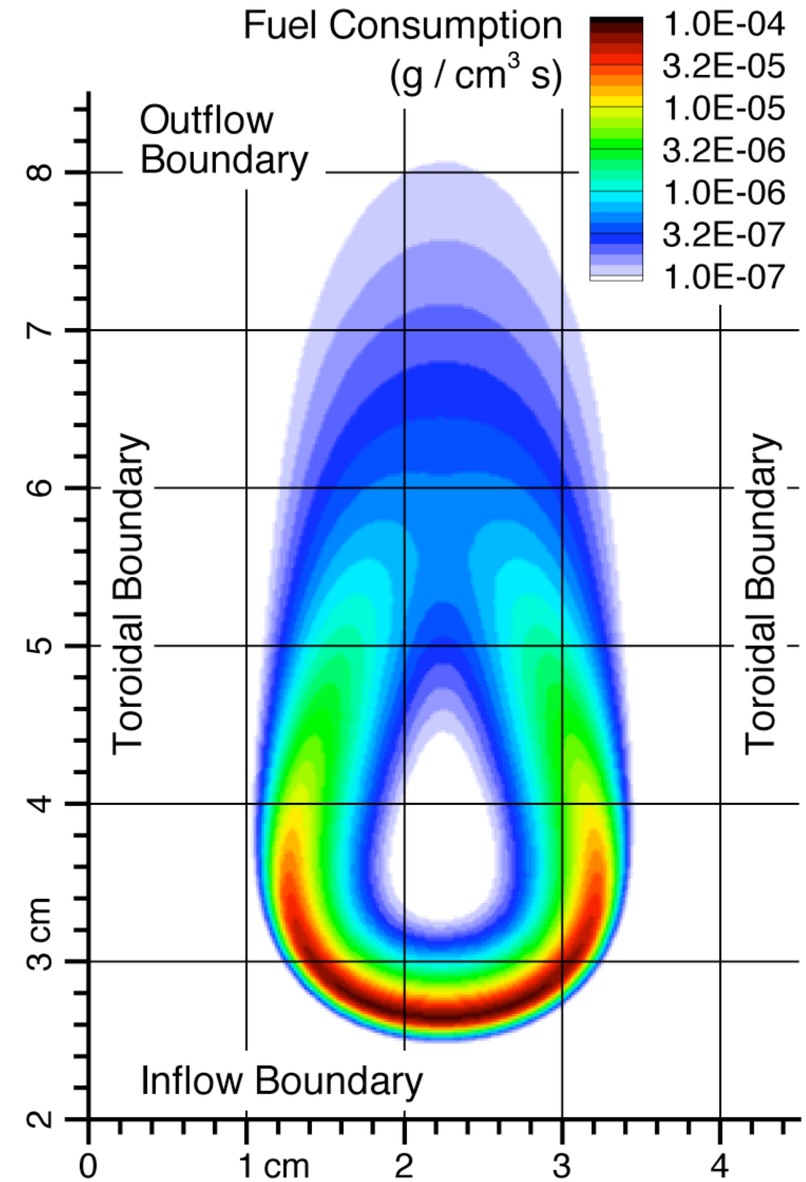
Fundamental studies (two dimensional) can be made on small parallel computers (16 processors).

Mathematical algorithms described by: Day and Bell, *CTM*, **4**:535–556 (2000).

2D DNS Results

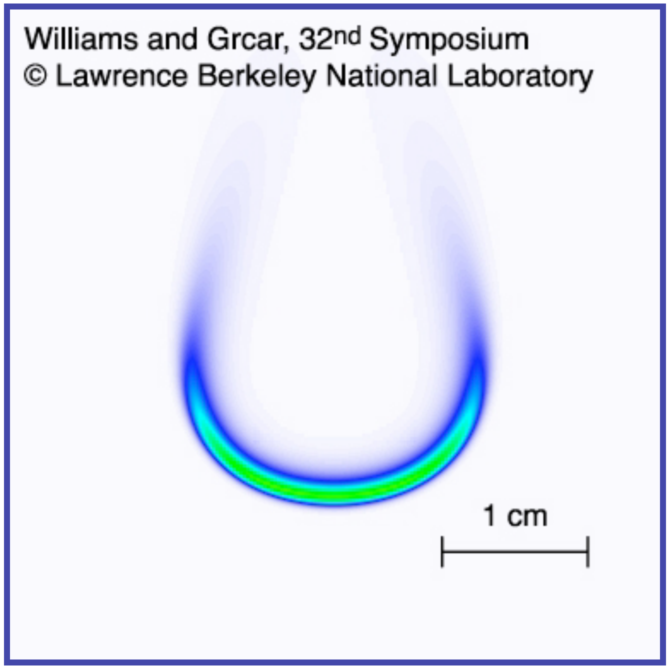
Begin with a planar, adiabatic flame at an equivalence ratio 0.3 and allow it to propagate at 1 atm, initial T 298K, into a mixture with an equivalence ratio 0.2.

The elongated flame ball develops and propagates steadily at a burning velocity of about 1.3 cm/s.



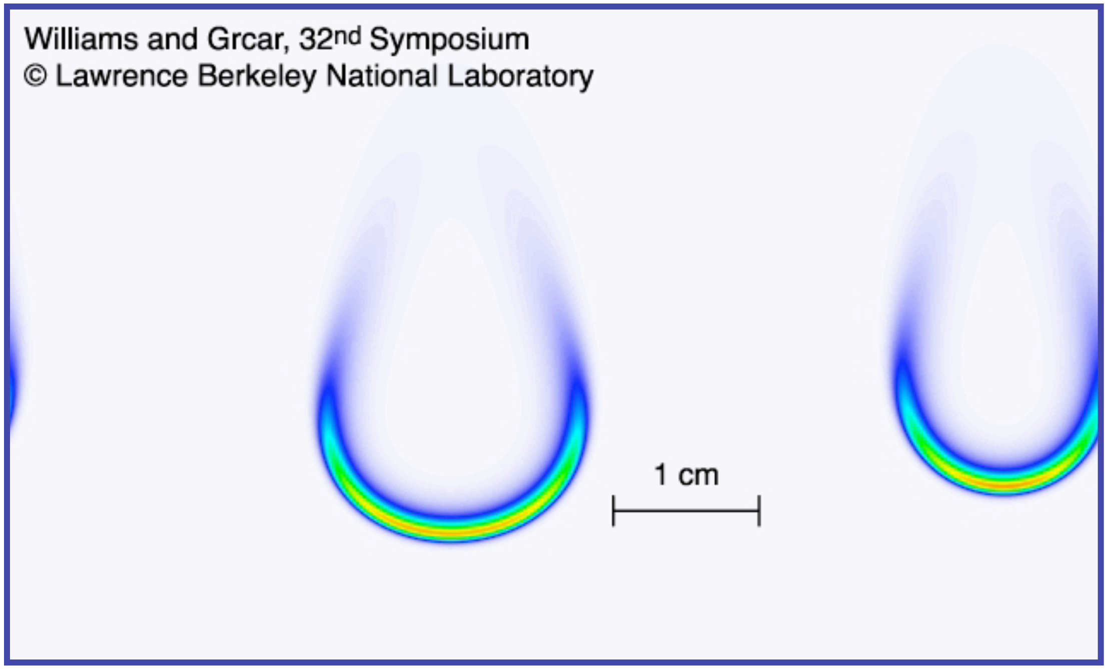
2D DNS Results

4.5 cm domain



10 seconds

7.5 cm wide domain



12 seconds

Downward Propagation in a 2-inch Tube

(Equivalence Ratio of 0.5)

Experiments at UCSD in 1978.

(Flame-fingers made photographable by addition of small (0.25%) amounts of CF_3Br.)

Flame Balls

- Hydrogen flame balls are well known; in Ronney's Space-Shuttle experiments they lasted longer than an Earth orbit.
- Steady and spherically symmetrical.
- Convection and accumulation terms zero; balance between reaction and diffusion.
- Corresponding balance between heat release and heat conduction.
- Thin reaction zone; solution for structure especially simple.

IDEAL FLAME-BALL ANALYSIS

α= Thermal Diffusivity. D= Hydrogen Diffusion Coefficient.

Lewis Number $L=\alpha/D$. Y= Fuel Mass Fraction.

Q= Heat Released per Unit Mass of Fuel Consumed.

For the variable $X=T+QY/(c_pL)$, the reaction term vanishes, so that $\rho Dr^2 dX/dr=\text{constant}$, which =0 at r=0, so X=constant. Evaluate the constant at infinity and inside the flame ball (Y=0).

Flame-Ball Temperature: $T_f=T_u+QY_u/(c_pL)$

By Contrast, Adiabatic Flame Temperature: $T_{af} = T_u + QY_u/c_p$

Solutions outside the flame ball: $Y=Y_u(1-R/r)$ and $T=T_f-(T_f-T_u)(1-R/r)$

Mass Rate of Consumption of Fuel per unit volume: $\rho YA\exp(-T_a/T)$

balances $d(\rho Dr^2 dY/dr)/dr/r^2$; integrate across thin flame; get $(dY/dr)^2$.

Flame-Ball Radius:

$$R = [(T_a QY_u)/(c_p L^2 T_f^{\,2})]\sqrt{D/(2Ae^{-T_a/T_f})}$$

Total Mass Rate of Consumption of Fuel by Flame Ball:

$$m = 4\pi R\rho DY_u$$

INFLUENCES OF THE SORET EFFECT

Diffusion Velocity:

$$V_{H_2} = -D\frac{dY}{dr} + k_T D\left(\frac{1}{T}\right)\frac{dT}{dr},$$

Diffusion Equation:

$$\frac{1}{r^2}\frac{d}{dr}\left[\rho D r^2\left(\frac{dY}{dr} - \frac{k_T}{T}\frac{dT}{dr}\right)\right] = \omega,$$

Energy Conservation:

$$\frac{1}{r^2}\frac{d}{dr}\left(\rho\alpha r^2\frac{dT}{dr}\right) = -\frac{Q}{c_p}\omega.$$

Adding:

$$\left(\frac{Q}{c_p}\right)\rho D r^2\left(\frac{dY}{dr} - \frac{k_T}{T}\frac{dT}{dr}\right) + \rho\alpha r^2\frac{dT}{dr} = 0,$$

Flame-Ball Temperature:

$$T_f = T_u + \frac{\left(Q/c_p\right)DY_u}{\alpha - \left(Q/c_p\right)\left(k_T/T\right)D}.$$

Effective Lewis Number:

$$Le_{eff} = Le\left(1 - \frac{Q}{c_p}\frac{k_T}{T}\right)$$

LEAN FLAMMABILITY LIMIT

- Equate flame-ball temperature to crossover temperature T_c , about 1000K.
- Flame-ball $T_f = T_u + QY_u/(c_p L)/[1 - \text{Soret}]$.
- Resulting equivalence ratio well below 0.1.
- Uncertainty in the Soret coefficient is so large that the lean limit may not exist.
- By contrast, for the planar flame the limit is slightly below an equivalence ratio of 0.3.

Safety Relevance

A hydrogen Lewis number of 0.3 means a flame-ball T increase of 3 times that of planar flames.

If the flammability limit is a crossover flame temperature of 1000K, the limit for planar flames, an equivalence ratio near 0.3, becomes less than 0.1 for the flame-ball arrays, much leaner.

MODEL FOR DEFLAGRATION VELOCITIES

- Deficiency in chemical kinetics unlikely.
- Cellular flames likely cause disagreements with experiments.
- The limit of a flame cell is a flame ball.
- *The opposite limit of the transversely homogeneous deflagration is a planar array of flame balls.*

THEORY

Let the number of flame balls per unit area be $1/a^2$. (Then a is the average transverse spacing of flame balls.)

Let the mass rate of consumption of fuel by a flame ball be m.

Then the mass rate of consumption of fuel per unit area by the array is m/a^2.

If v is the deflagration velocity and the initial mass of fuel per unit volume is $Y_u\rho$,

$$Y_u \rho v = m/a^2$$

QUESTION

What is the value of a?

Answer: I have no idea, but it certainly cannot be less than a flame-ball radius, R.

Decision: As an upper bound for the burning velocity, assume a triangular (that is, hexagonal) close-packed planar array of flame balls.

$$a^2=2(3^{1/2})R^2$$

Result: An explicit burning-velocity formula that can be evaluated from flame-ball results.

RESULTS OF THE THEORETICAL FLAME-BALL DEFLAGRATION MODEL

With these flame-ball results, by substitution we see the

Upper Bound $v=2\pi(D/R)/(3^{1/2})$.

Large D and small R favor large v.

Use of the flame-ball formula for R, which in turn involves the flame-ball formula for T_f, then gives a burning-velocity formula that can be compared with the classical planar-flame formula.

Note: The results are based on one-step activation-energy asymptotics, which facilitates comparisons.

COMPARISON WITH CLASSICAL PLANE-FLAME RESULT

Burning Velocity for Flame-Ball Array without Soret:

$$v = \frac{2\pi D}{\sqrt{3}R} = \frac{2\pi Q Y_u}{c_p T_a}\left(1 + \frac{\alpha c_p T_u}{D Q Y_u}\right)^2 \sqrt{\frac{2}{3} D A e^{-T_a / T_f}}$$

where $T_f = T_u + QY_u / (c_p L)$.

Burning Velocity for Planar Flame:

$$v = \frac{Q Y_u}{c_p T_a}\left(1 + \frac{c_p T_u}{Q Y_u}\right)^2 \sqrt{2 \frac{\alpha^2}{D} A e^{-T_a / T_{af}}}$$

where $T_{af} = T_u + QY_u/c_p$.

Note opposite dependences on D and flame-ball T_f (hence v) larger when L<1.

NUMERICAL COMPARISON

Employ the asymptotics result $T_a = 4T_f^2 / \left(T_f - T^0\right)$

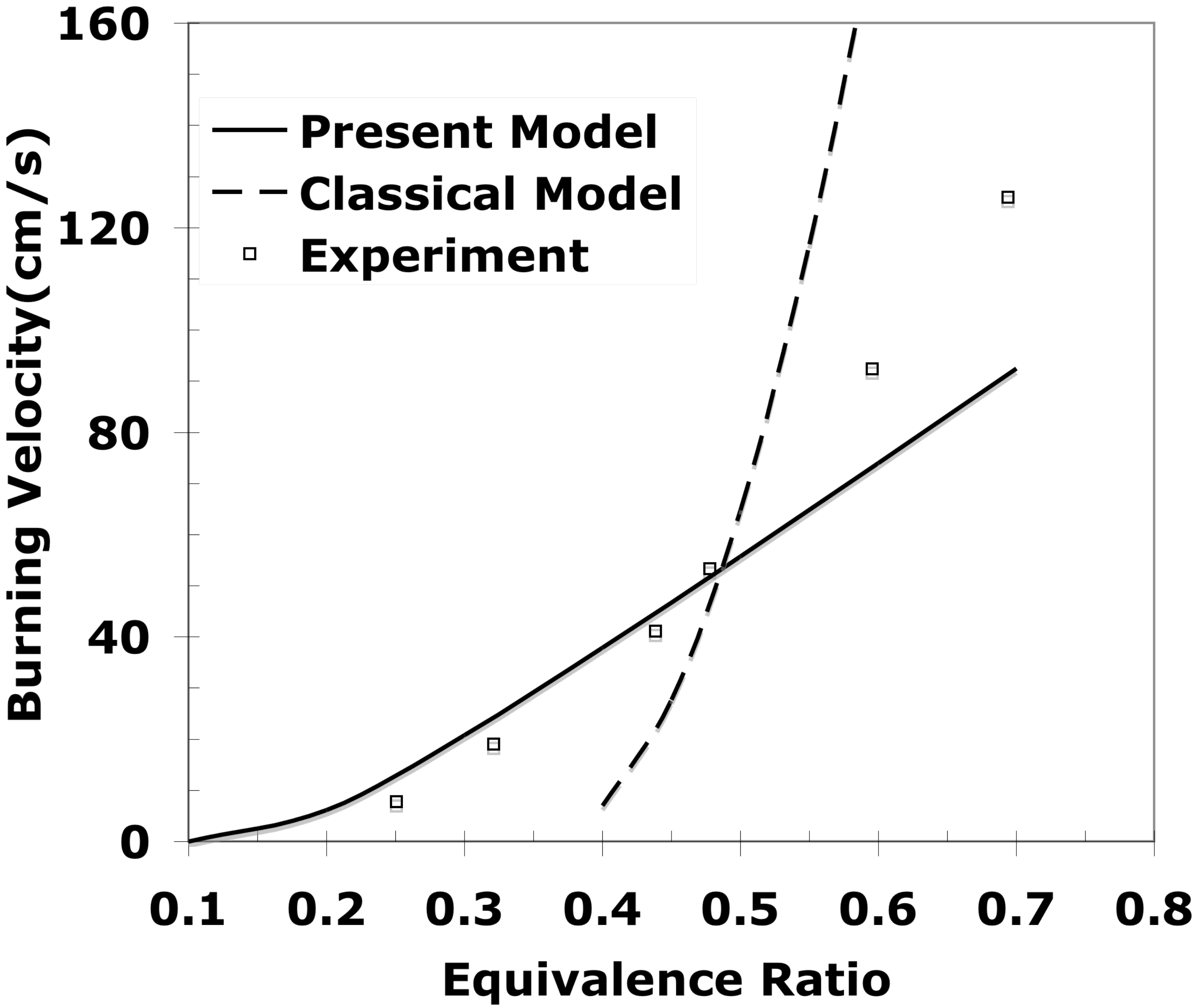
to obtain

$$-\frac{T_a}{2T_f} = -\frac{2}{1-\left(T^0/T_f\right)}$$

in the Arrhenius factor, where T^0 denotes the crossover temperature.

Choose A to fit the experimental v at an equivalence ratio of 0.5 (similar results for a fit at 0.7).

The flame-ball formula agrees better with experiment for equivalence ratios less than 0.5.



Present Model
Classical Model
Experiment
Burning Velocity(cm/s)
0
40
80
120
160
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
Equivalence Ratio

NEEDED IMPROVEMENTS IN THE MODEL

Radiation transport is needed in the flame-ball analysis (to stabilize it).

Soret needs to be included in the flame-ball analysis.

Study of the stability and dynamics of arrays of stable flame-balls are needed because they repel each other, so the planar array seems unstable.

Flame-ball analyses with improved flame chemistry near crossover are needed especially the systematically reduced one-step chemistry of the previous chapter.

More 3-D lean-flame computations with detailed and reduced chemistry are needed to investigate cellular structures and dynamics of flame-ball-like elements.

More microgravity lean-system experiments also would be helpful.

CATEGORIES OF APPROACHES TO ANALYSIS OF TURBULENT COMBUSTION

1. Phenomenological
    a. Quasidimensional
    b. Age Theories
    c. Linear Eddy/One-Dimensional Turbulence
2. Fluids-Based
    a. Direct Numerical Simulation (DNS)
    b. Large-Eddy Simulation (LES)
    c. Moment Methods (RANS)
        i. Algebraic Closures
        ii. k-ε Modeling
        iii Reynolds-Stress Closure
3. Turbulent Burning Velocity (S_T)
    a. Perturbations for Low Intensities and Large Scales
    b. Moment-Method Modeling of the G Equation
    c. Modeling Flame-Surface Evolution, such as Coherent-Flamelet Models (CFM)
    d. Fractals
    e. G-Equations Renormalization
    f. Pseudosolitons
4. Probability-Density Function (PDF)
    a. Flamelets
    b. Presumed PDF
        i. P(Z) for Diffusion Flames
        ii P(c) for Premixed Flames
        iii P(G) for Premixed Flames
    c. Conditional Moment Closure (CMC)
    d. PDF Transport
        i. Linear Mean-Square Estimation (LMSE), also called Interaction by Exchange with the Mean (IEM)
        ii Coalescence-Dispersion (CD)
        iii. Mapping Closure (MC)
        iv. Euclidean Minimum Spanning Tree (EMST)

CONCLUSIONS

- Hydrogen combustion obeys the reacting Navier-Stokes equations.

- Lean hydrogen deflagrations involve cellular flames.

- Measured lean hydrogen deflagration velocities exceed calculated values for planar steady flames because of diffusive-thermal instabilities.

- Flame balls are robust in lean hydrogen mixtures and can form the basis of improved models of lean deflagrations.

- Soret diffusion is important in lean hydrogen flame balls and must affect the lean flammability limit.

- Ultimate lean hydrogen flammability limits may be much leaner than currently believed but are uncertain because of the uncertainty in the value of the Soret coefficient.

- There are four broad categories of approaches to the description of turbulent hydrogen combustion, which involves fluid-mechanical turbulence.