Syngas & Hydrogen Combustion Reduced Order Reactions: Flame speeds, Mechanism validation, Reduced Mechanism

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Research goal:
Gas Turbine Engines Using Hydrogen Syngas

•Validated, reduced order kinetic mechanism
•Understanding kinetics at engine conditions

- High pressure (~10-30 atm)
- Lean/rich conditions for low NOx (3ppm)
- Preheated temperature (~ 650K)
- High levels of diluents (N_2, H_2O,or CO_2)

Research Tasks

- Methodology to measure flame speeds at high pressure
- Flame speed measurements and mechanism validation
- Dynamic multiscale (AMS) kinetic reduction algorithm

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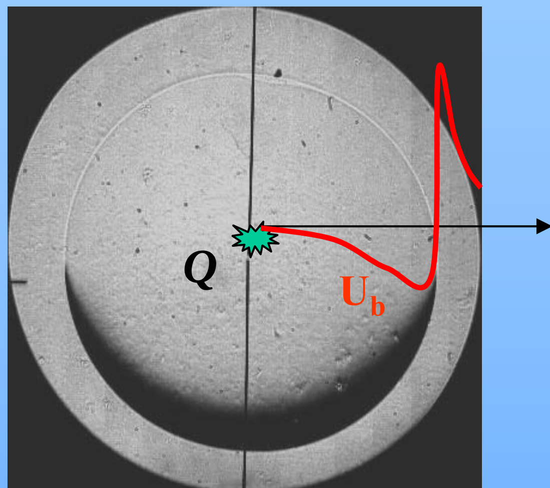
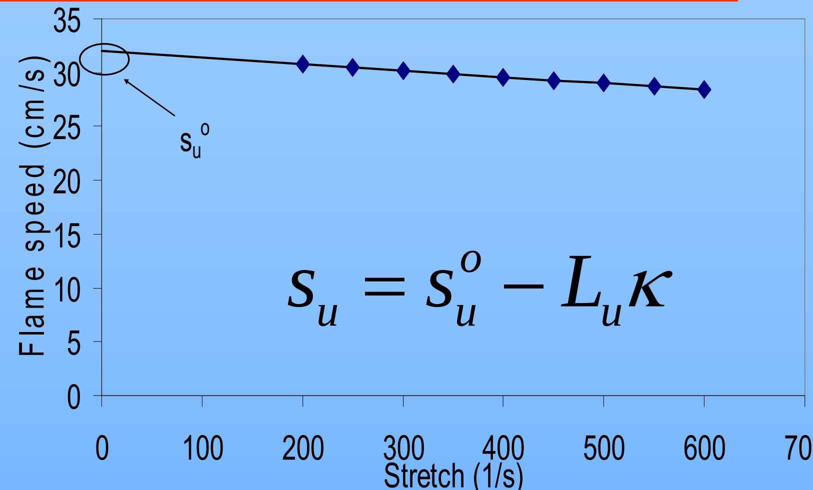
1. Methodology to measure rigorous flame speeds

Flame speed measurement using spherical flames





Assumption: Burned gas velocity is zero!

$$S_u = V_f \rho_b / \rho_u \qquad (1)$$

What if the burned gas velocity is not zero?

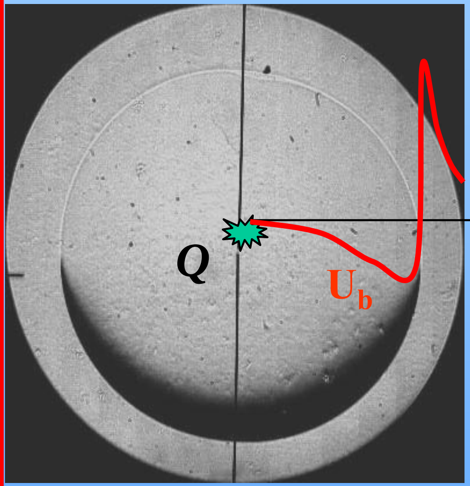
Flame speed error will be:

$$\varepsilon \equiv \frac{S_u - \hat{S}_u}{\hat{S}_u} = \frac{u_b}{V_f} \qquad (2)$$

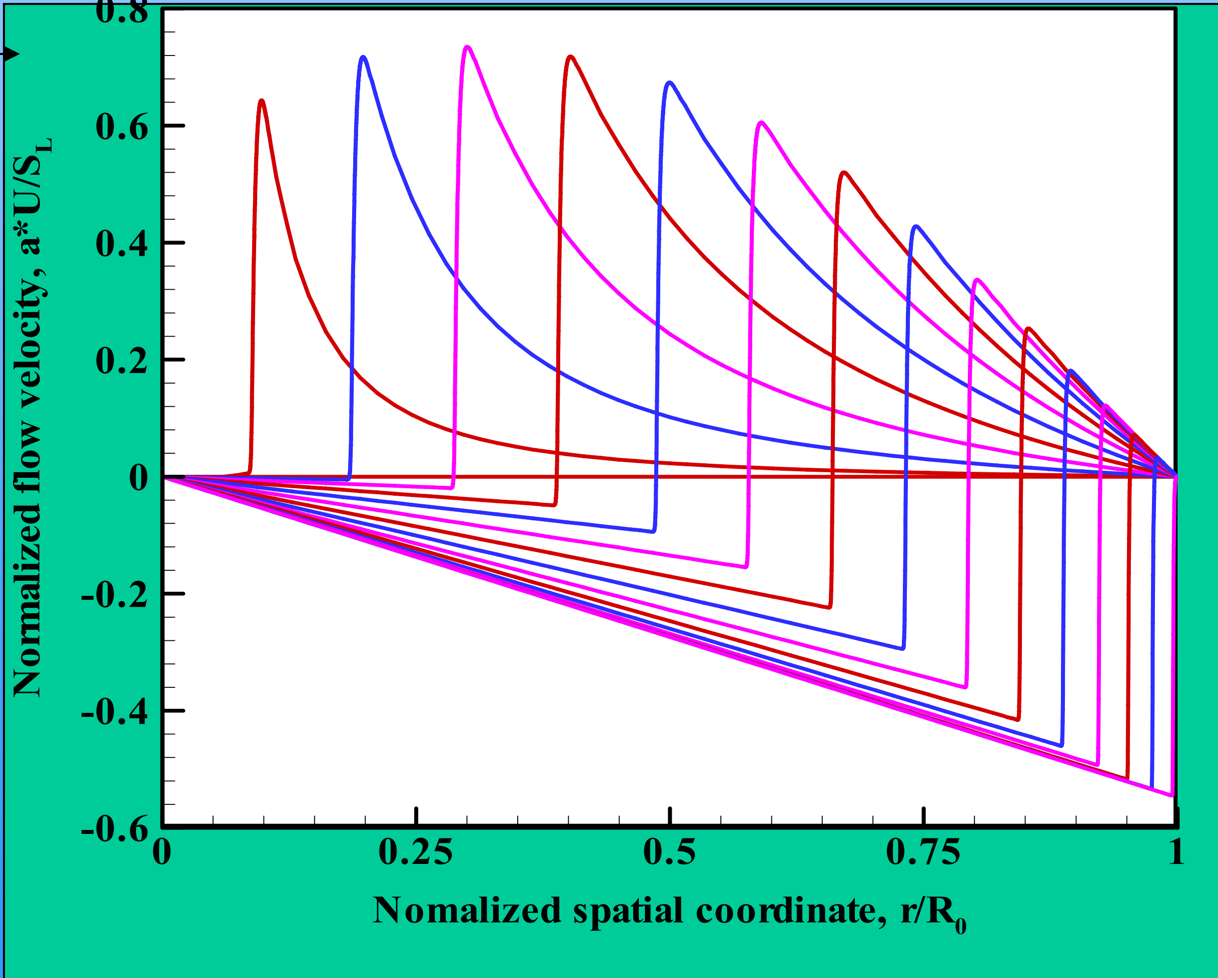
What causes non-zero burned gas velocity?

Compression, non-symmetric flow, radiation…

Flame speed measurement using a spherical bomb: *Compression induced burned gas velocity*



Velocity distribution V(r,t)



Chen & Ju 2007

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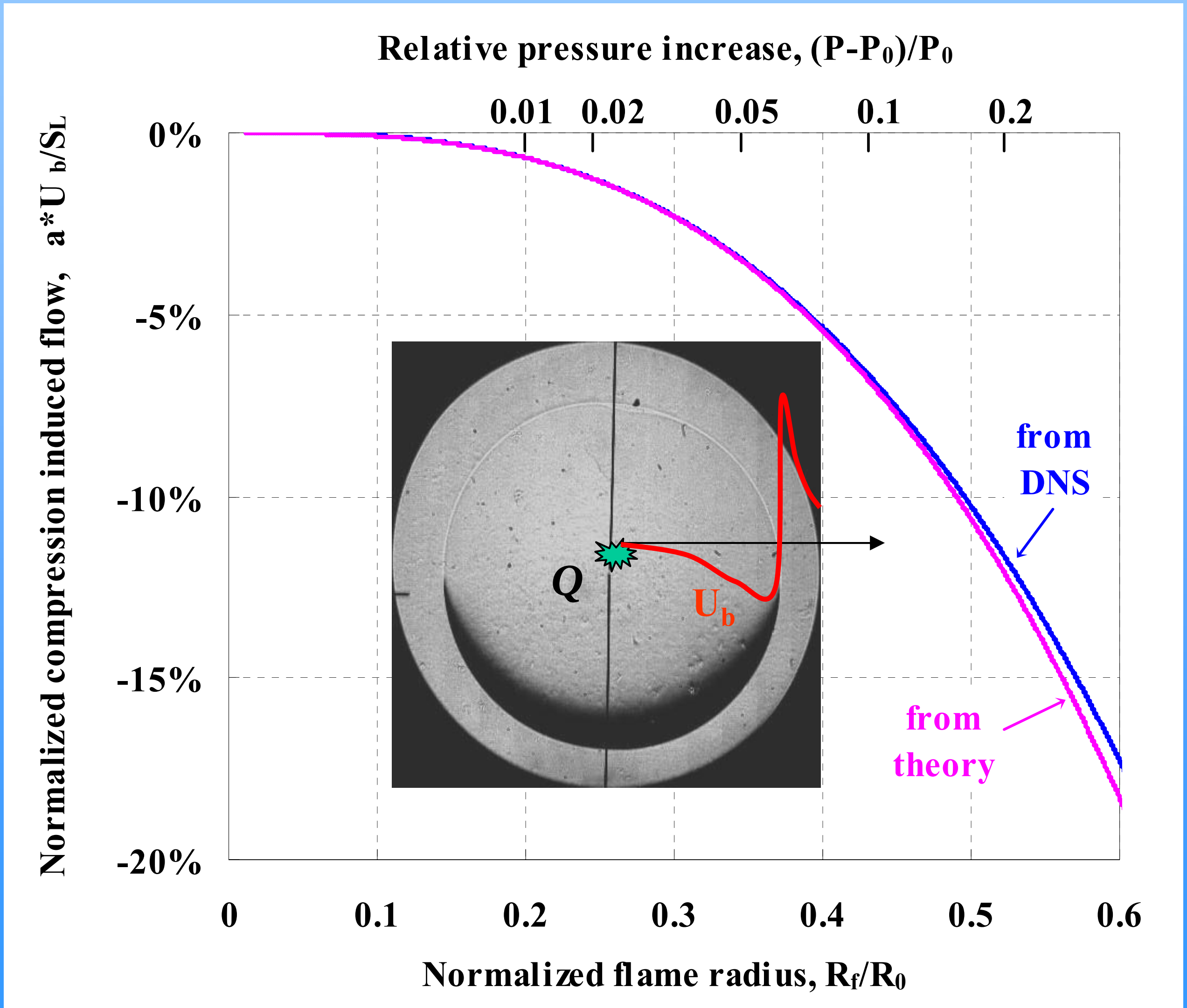
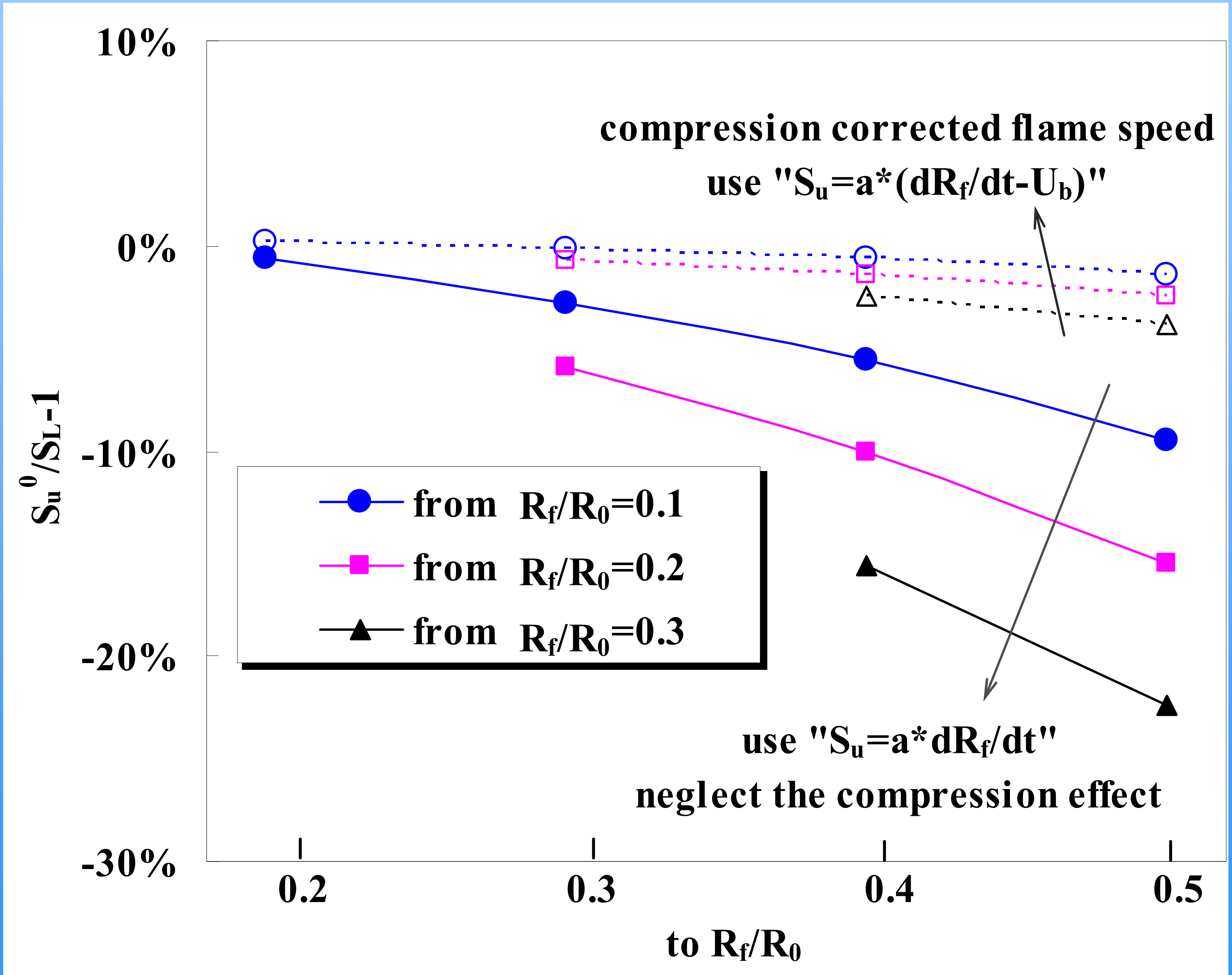
Constant pressure method: *Compression induced burned gas velocity*

Theoretical estimation:



Constant Pressure Method: *Compression Corrected Flame Speed (CCFS)*



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What if a cylindrical chamber is used?

Princeton 8.3 cm dia by 13 cm length (Law),
10 cm dia by 15 cm length (Ju)

Other groups too!

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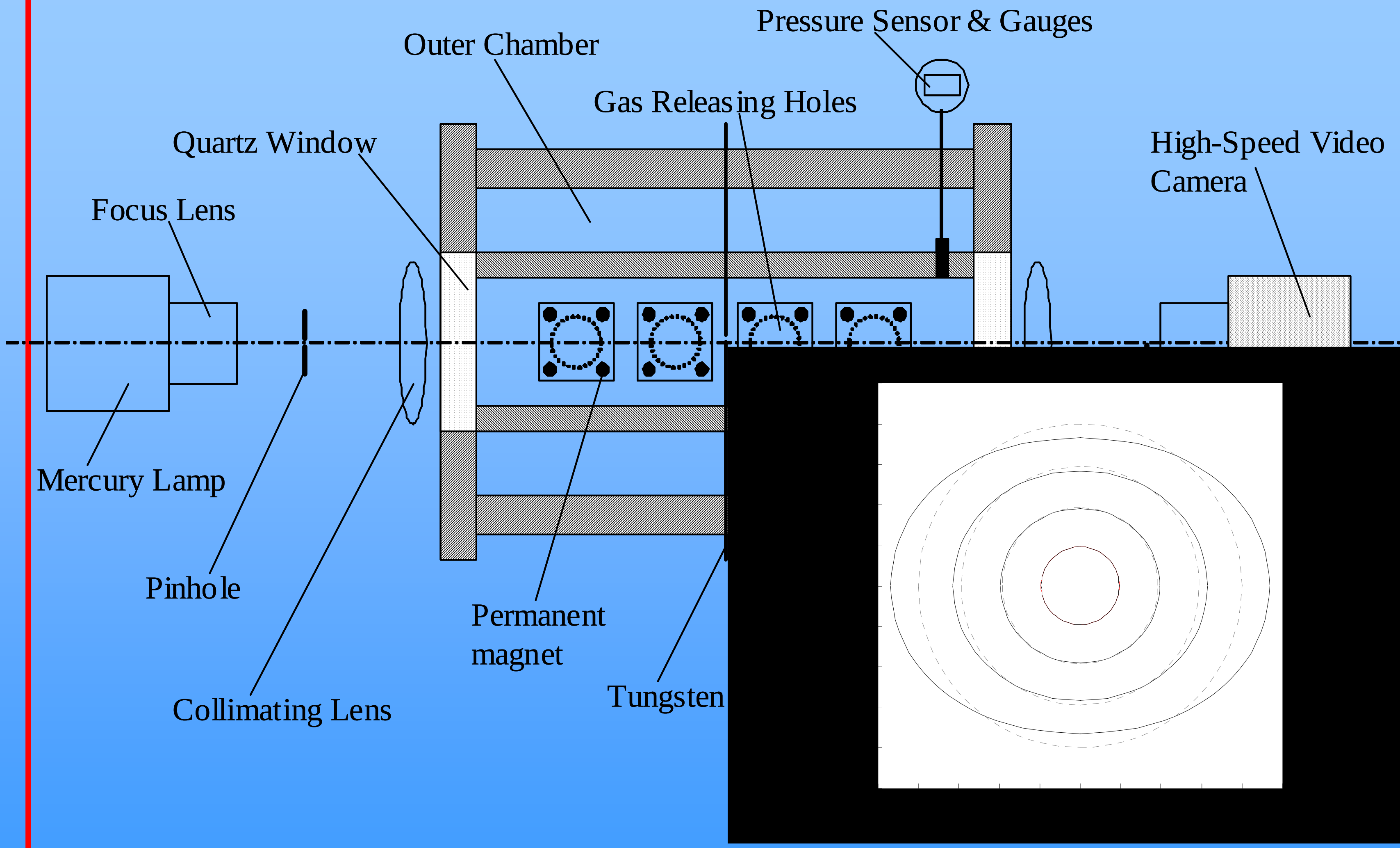
What if a cylindrical chamber is used?



Non-symmetrical flow induced burned gas velocity!

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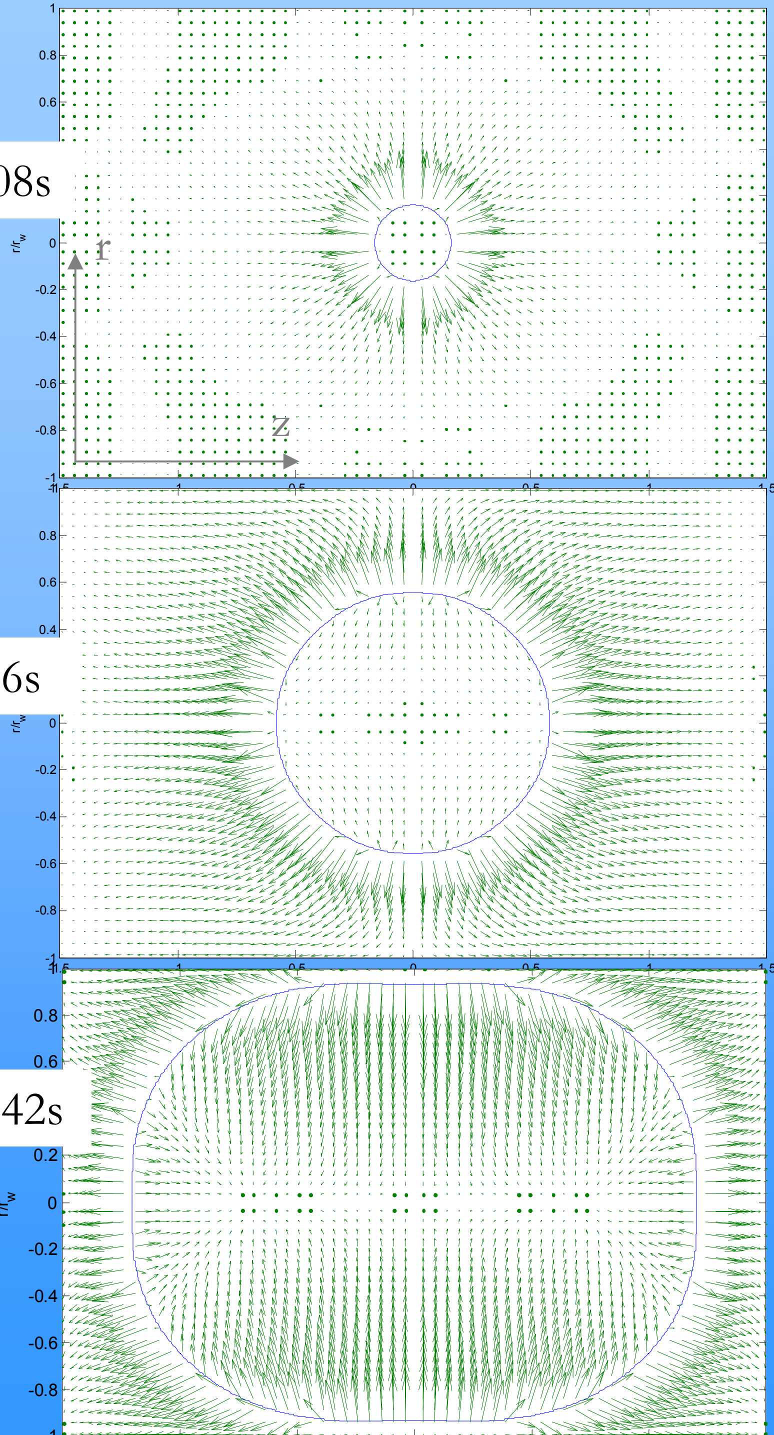
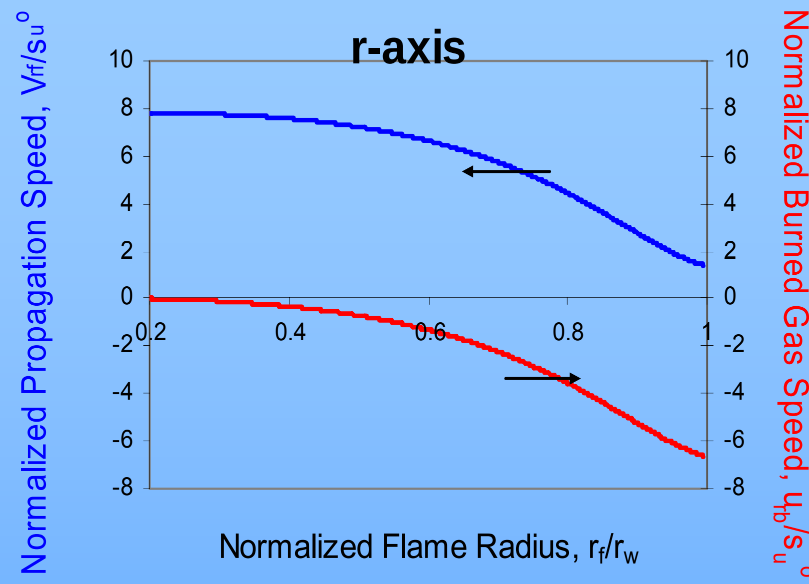
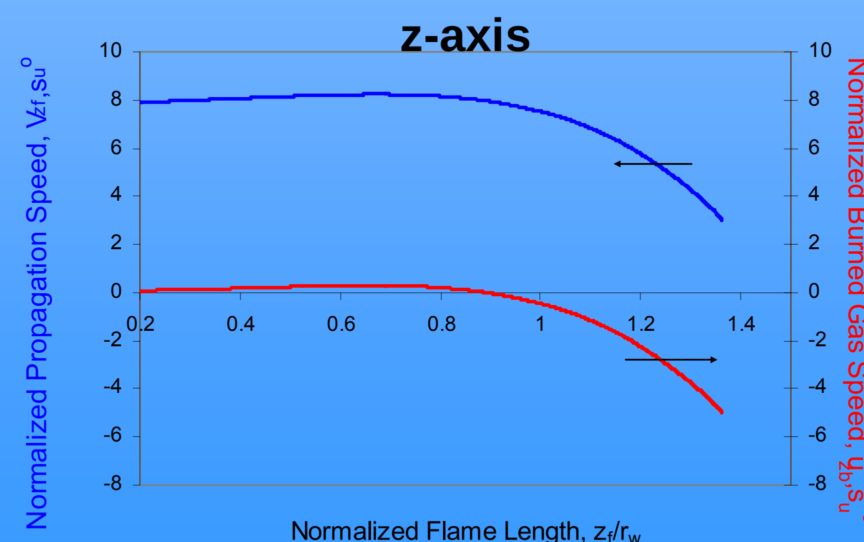
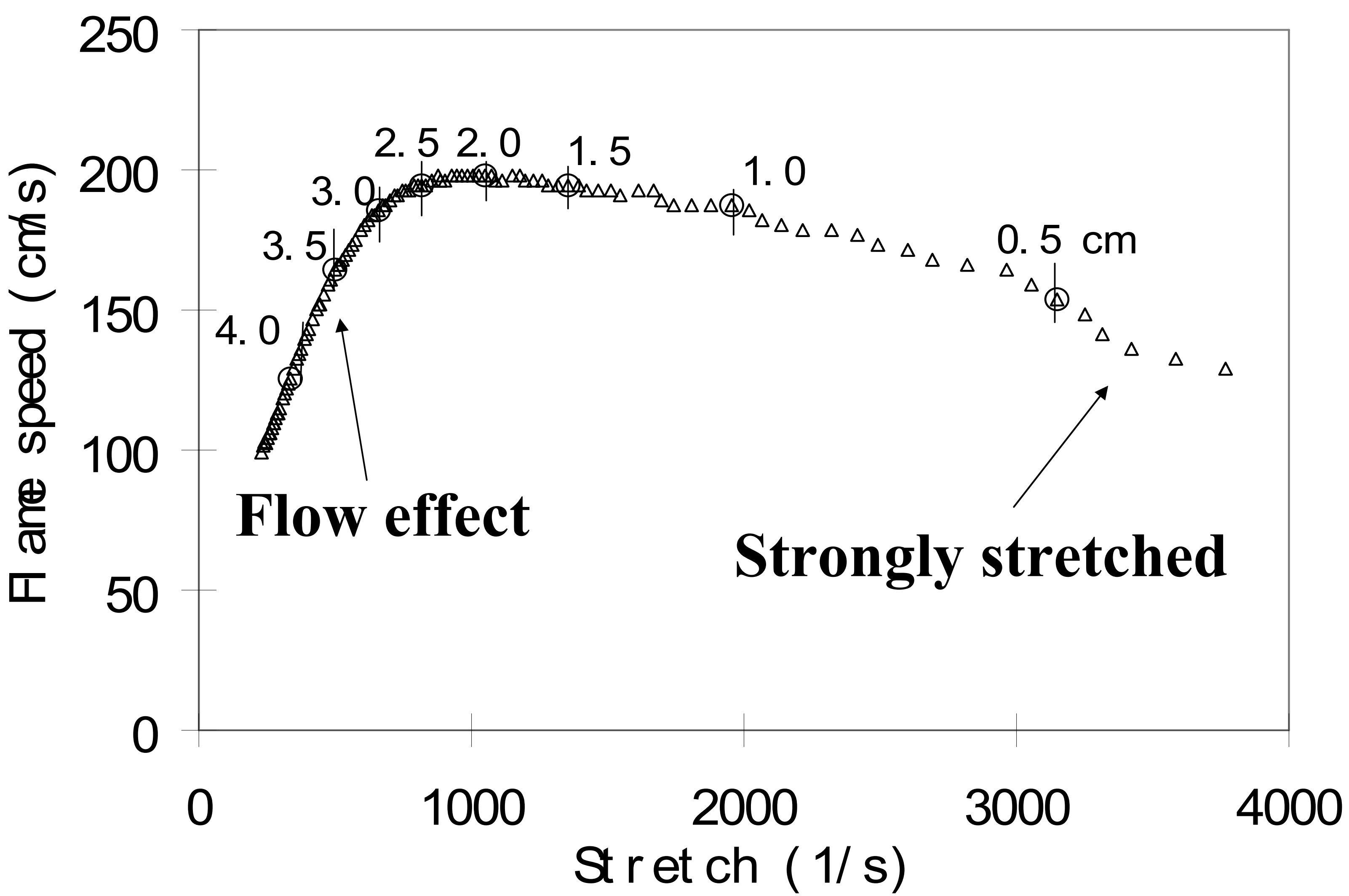






Burke, Chen, Ju, Dryer, submitted for publication

Flame speed history in a cylindrical chamber



Difficulty in linear extrapolation!

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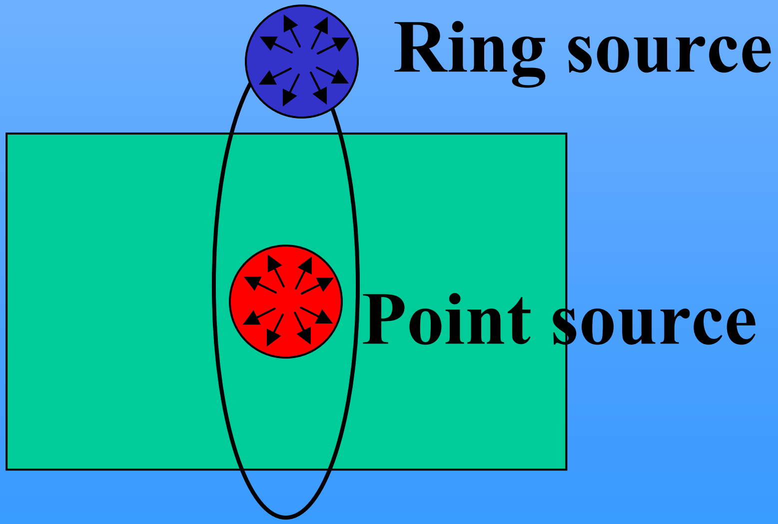
How to improve the measurements

Potential flow:

$$\phi = \frac{m(t)}{4\pi \cdot r} + \int_0^{2\pi} \frac{q(t)}{\left\{[x-\eta\cdot\cos(\gamma)]^2 + [y-\eta\cdot\sin(\gamma)]^2 + z^2\right\}^{1/2}} \cdot d\gamma$$



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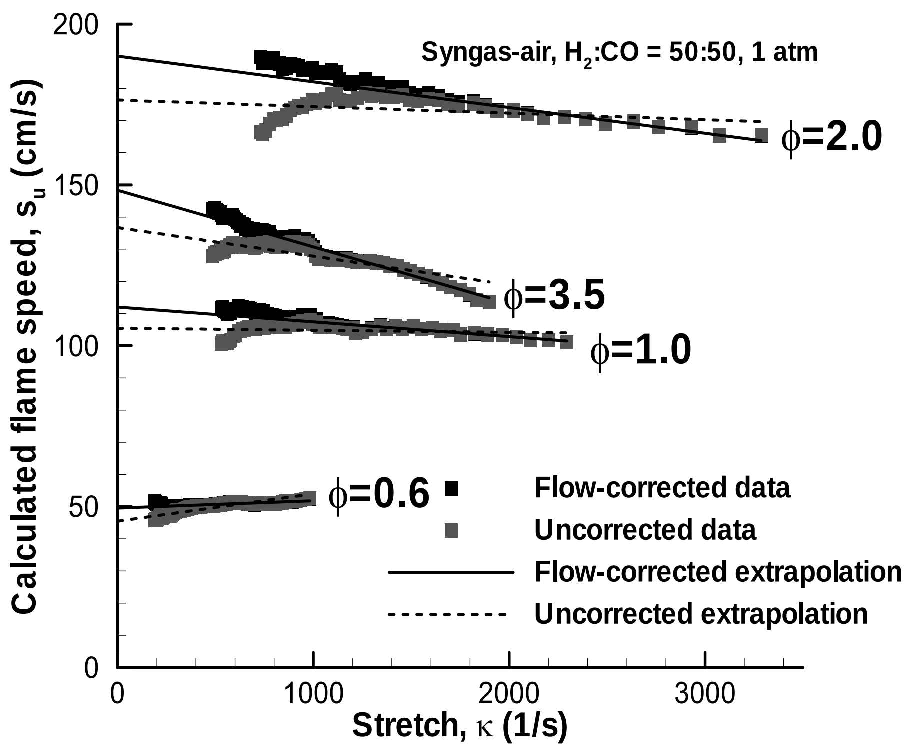
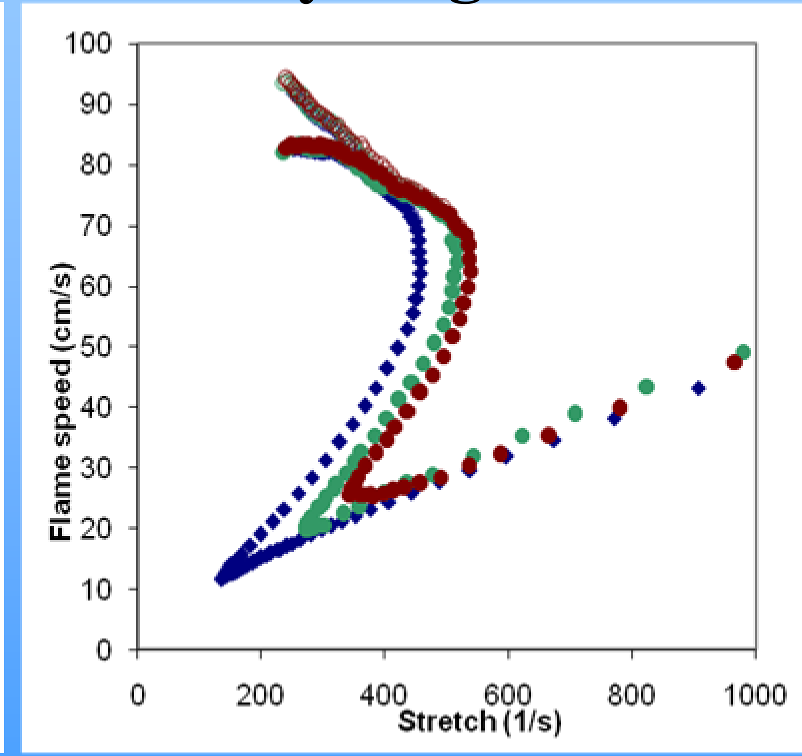
Flow correction of flame speeds

Syngas-air



Rich hydrogen-air



Improves accuracy and extends range of flame radii that can be used for extrapolation

Allows for flame speed determination for flames with long ignition transients

Burke, Chen, Ju, Dryer, submitted for publication

Chen, Burke, Ju, Proc. Comb. Inst. (2009)

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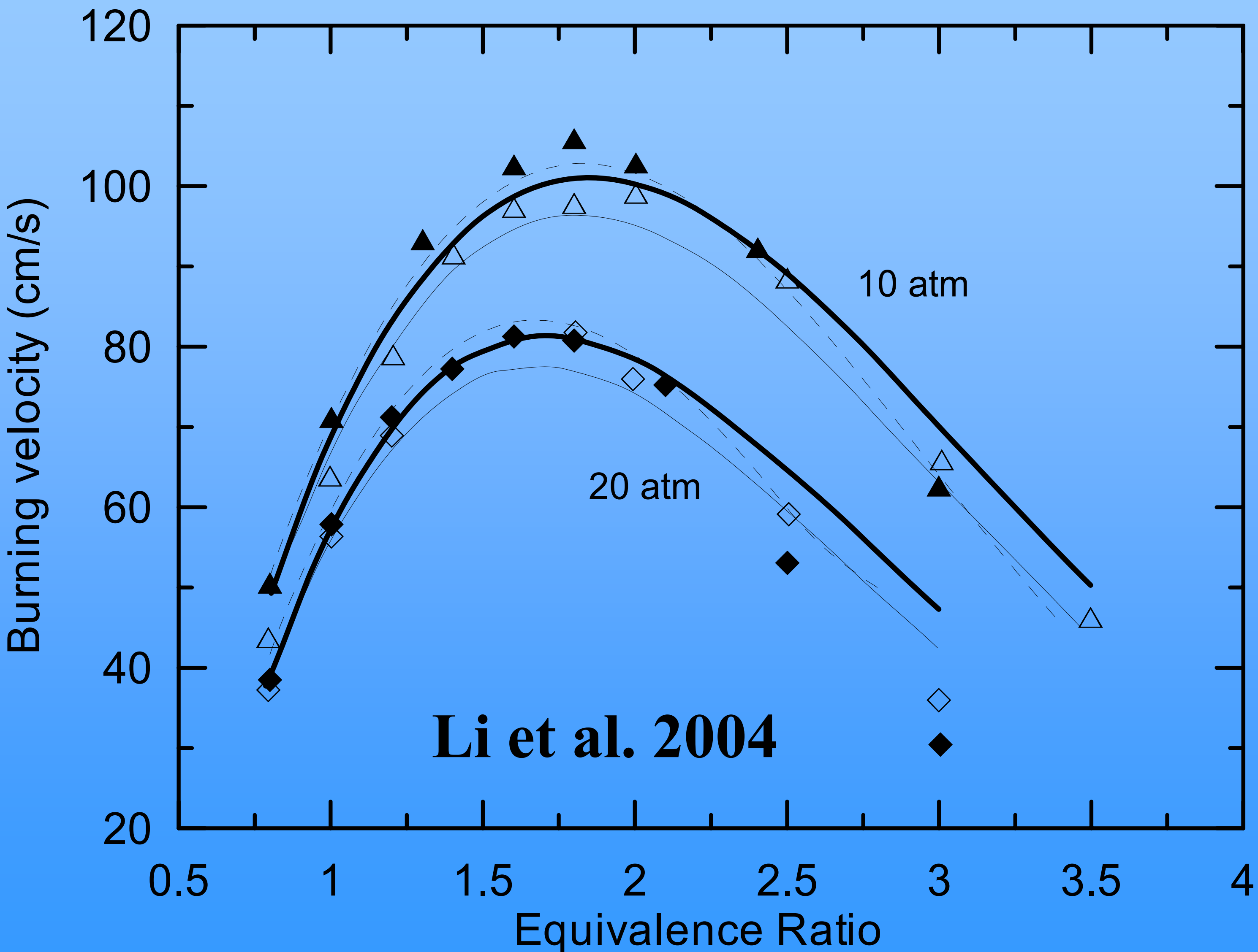
2. Flame speed measurements & kinetic mechanism

Experimental data: Syngas flame speed at 10, 20 atm

H_2-CO = 25:75 syngas mixture in O_2+7He



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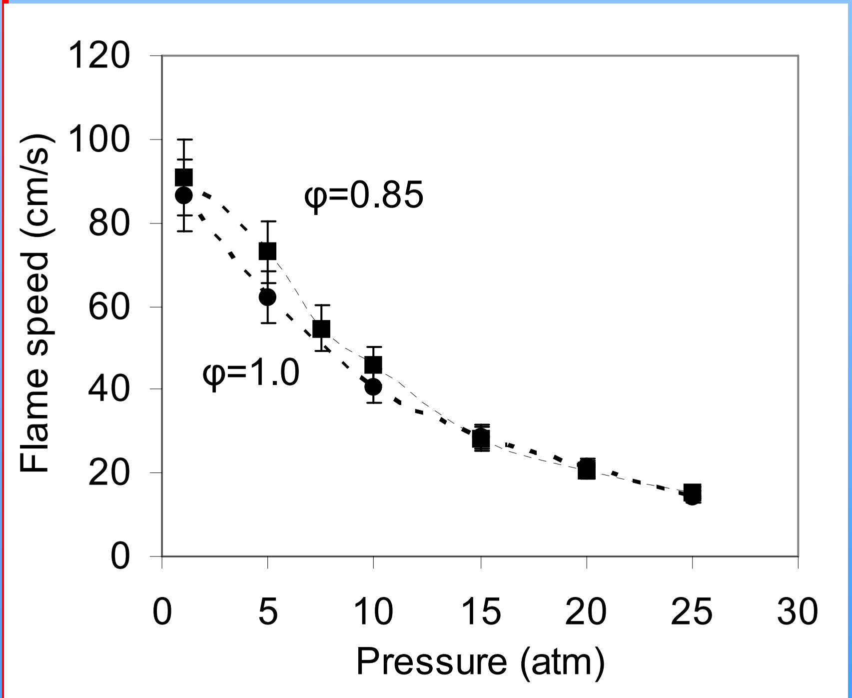
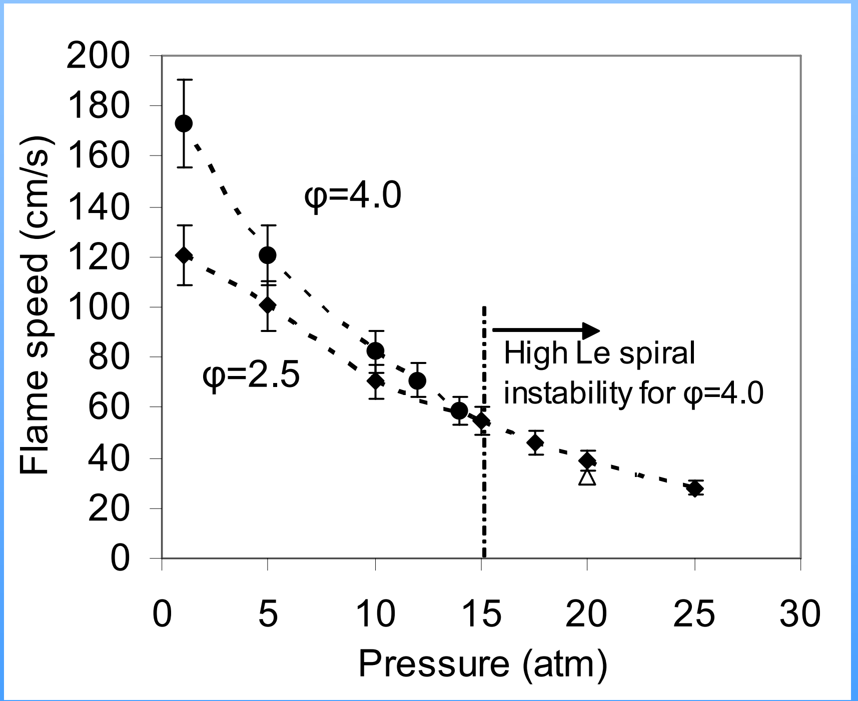
Discrepancy in rich conditions

Experimental Data (hydrgon-air): Flame speed vs. Pressure

Lean $H_2/O_2/He$ of Flame Temp=1600K



Rich $H_2/O_2/Ar$ or N_2 of Flame Temp=1600K



- **Flame speed decreases uniformly with pressure**
- **Flame speed becomes insensitive to mixture composition, but only to temperature at high Pressure**

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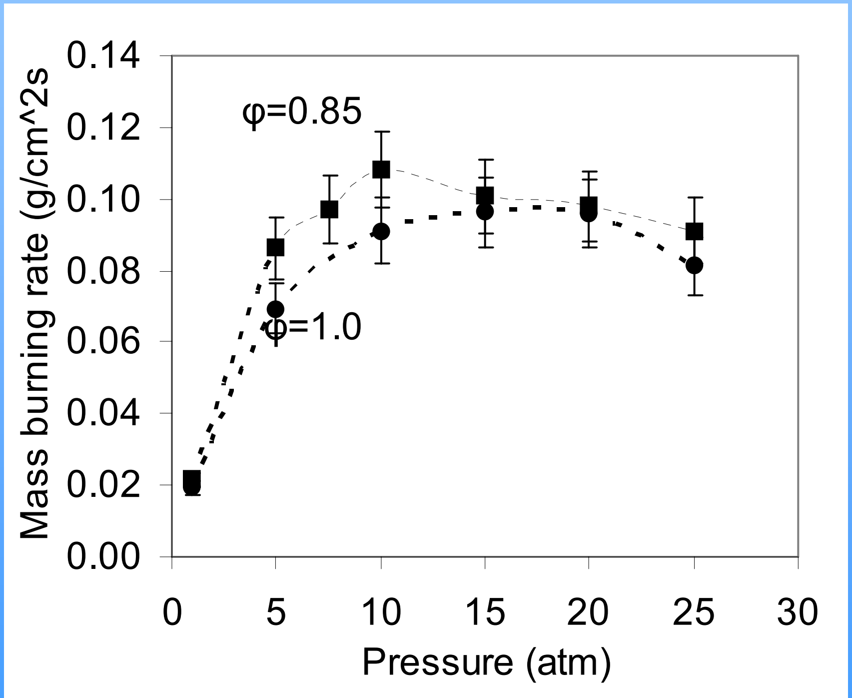
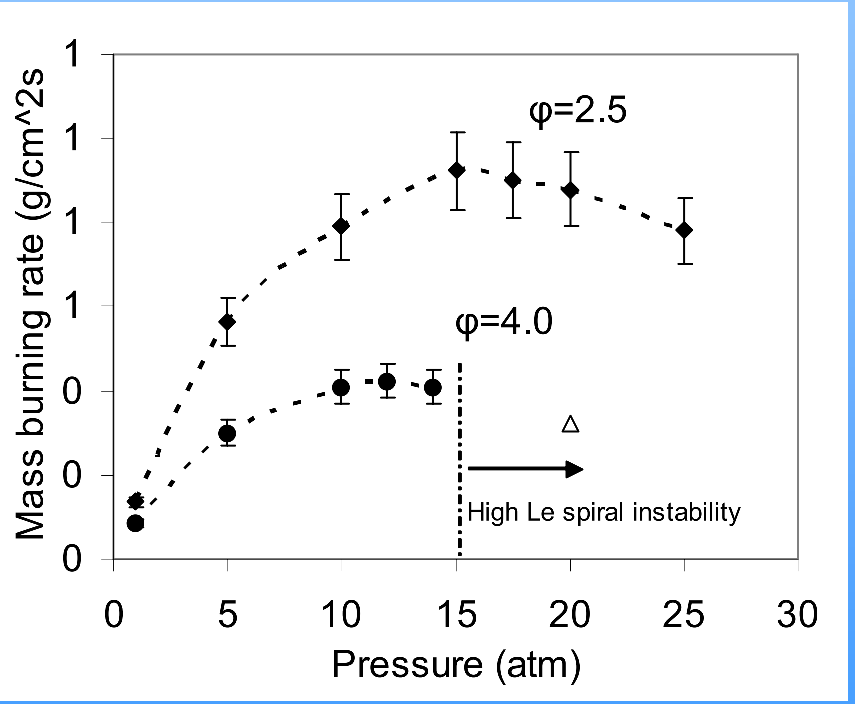
Experimental data:
Mass Burning Rate vs. Pressure

Lean $H_2/O_2/He$ of Flame Temp=1600K



Rich $H_2/O_2/Ar$ or N_2 of Flame Temp=1600K



- Mass burning rate increases with pressure until 15 atm, then decreases with pressure

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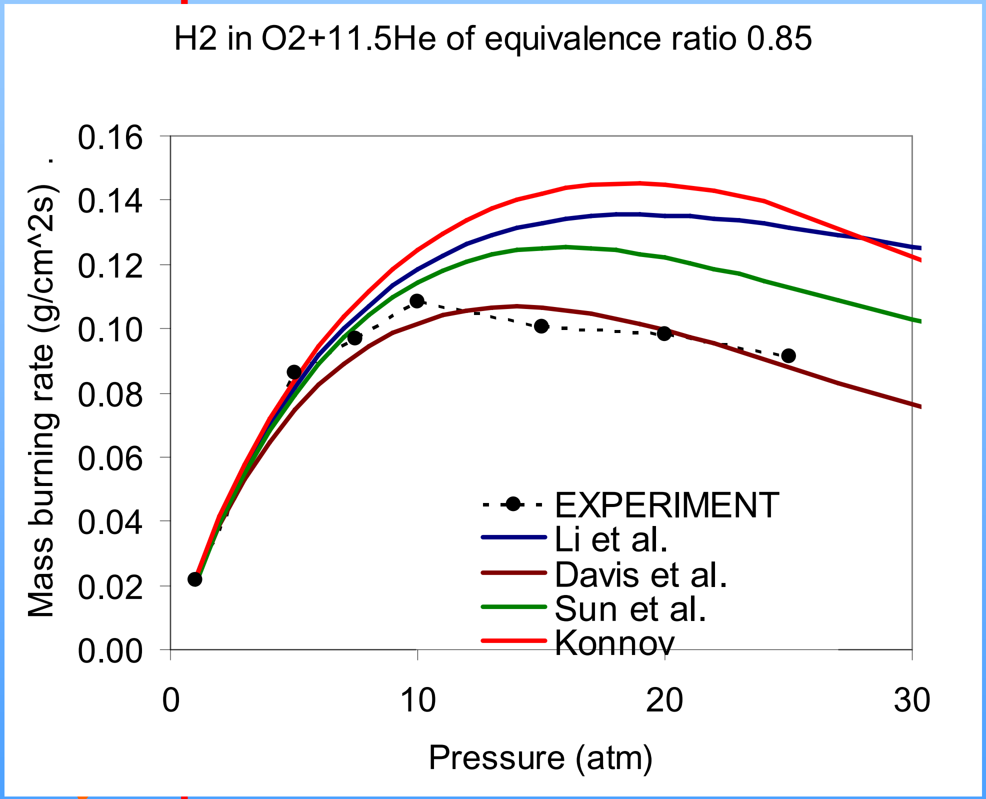
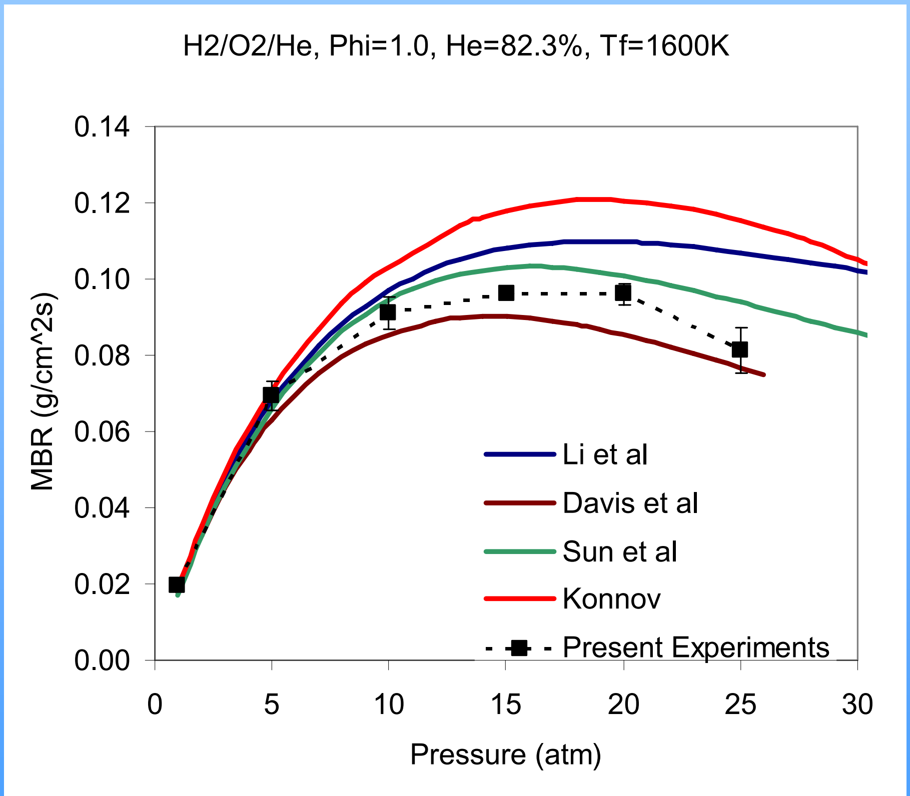
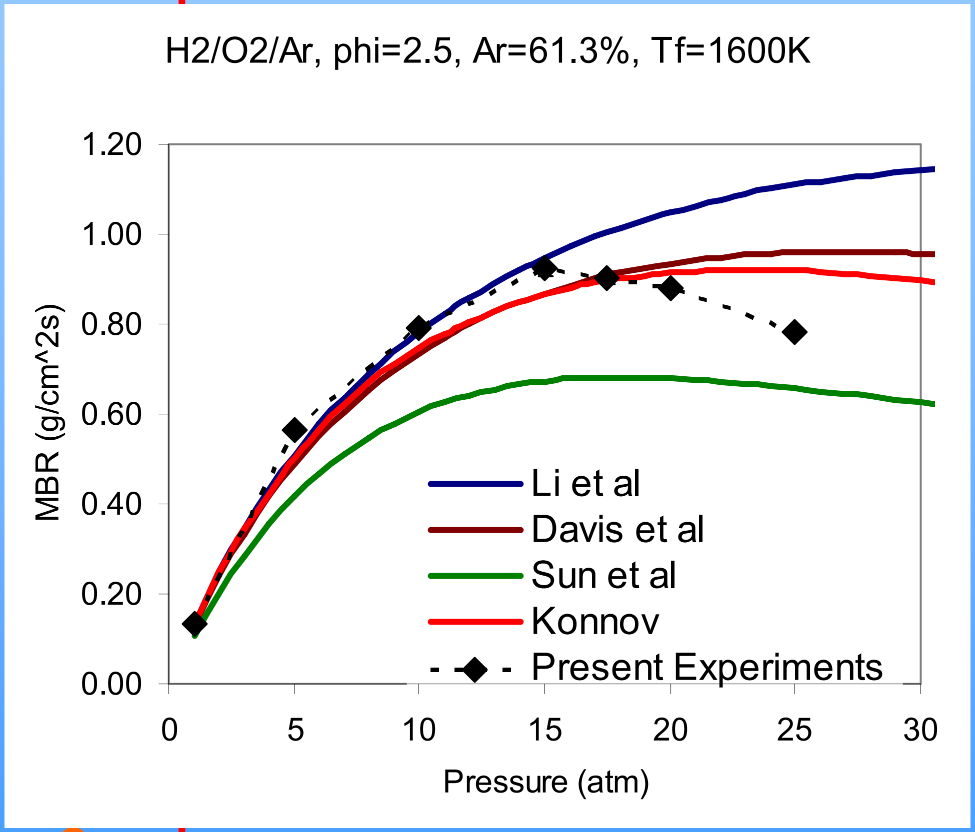
Validation of kinetic mechanism
Lean mixtures at high pressures

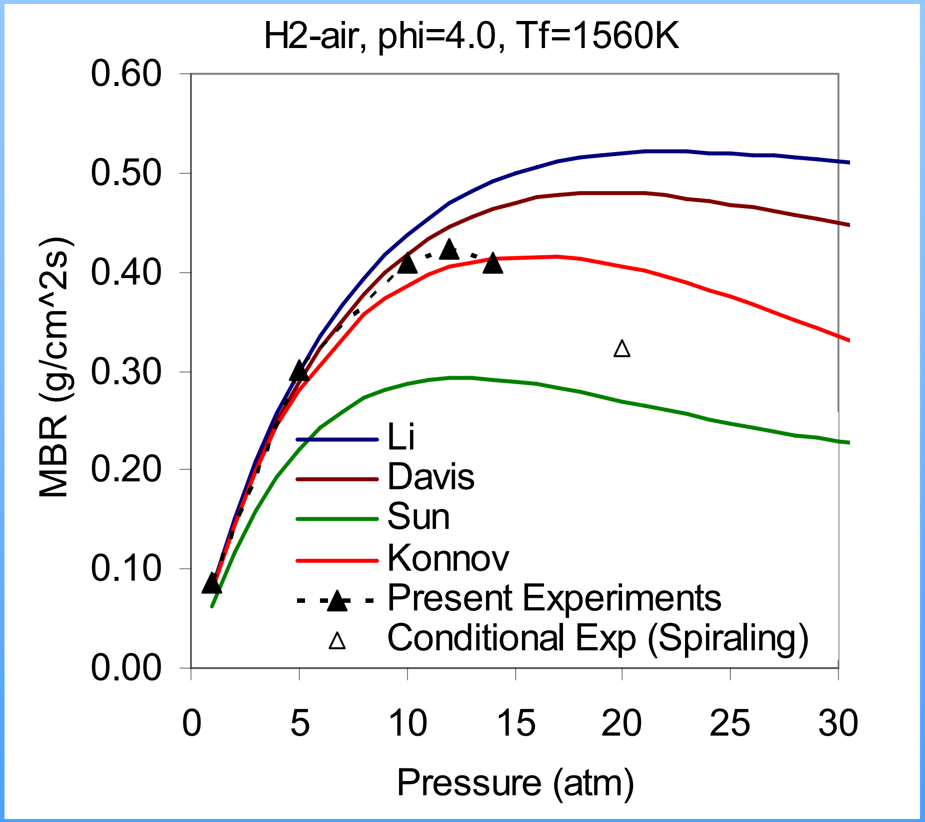




- Mechanism of Davis et al. outperforms the other models
- Other models disagree by up to ~40%

Validation of kinetic mechanism
Rich mixtures at high pressures





- **No model accurately predicts trends above 15 atm**
- **Differences among models reach about a factor of 2**

What happens kinetically when the pressure increases?

- Three-body reactions favored
  - e.g.

$H+O_2 \rightarrow OH+O$ (R1)

*chain-branching

*favored at lower pressures and higher temperatures

$H+O_2 \xrightarrow{+M} HO_2$ (R9)

*chain-termination

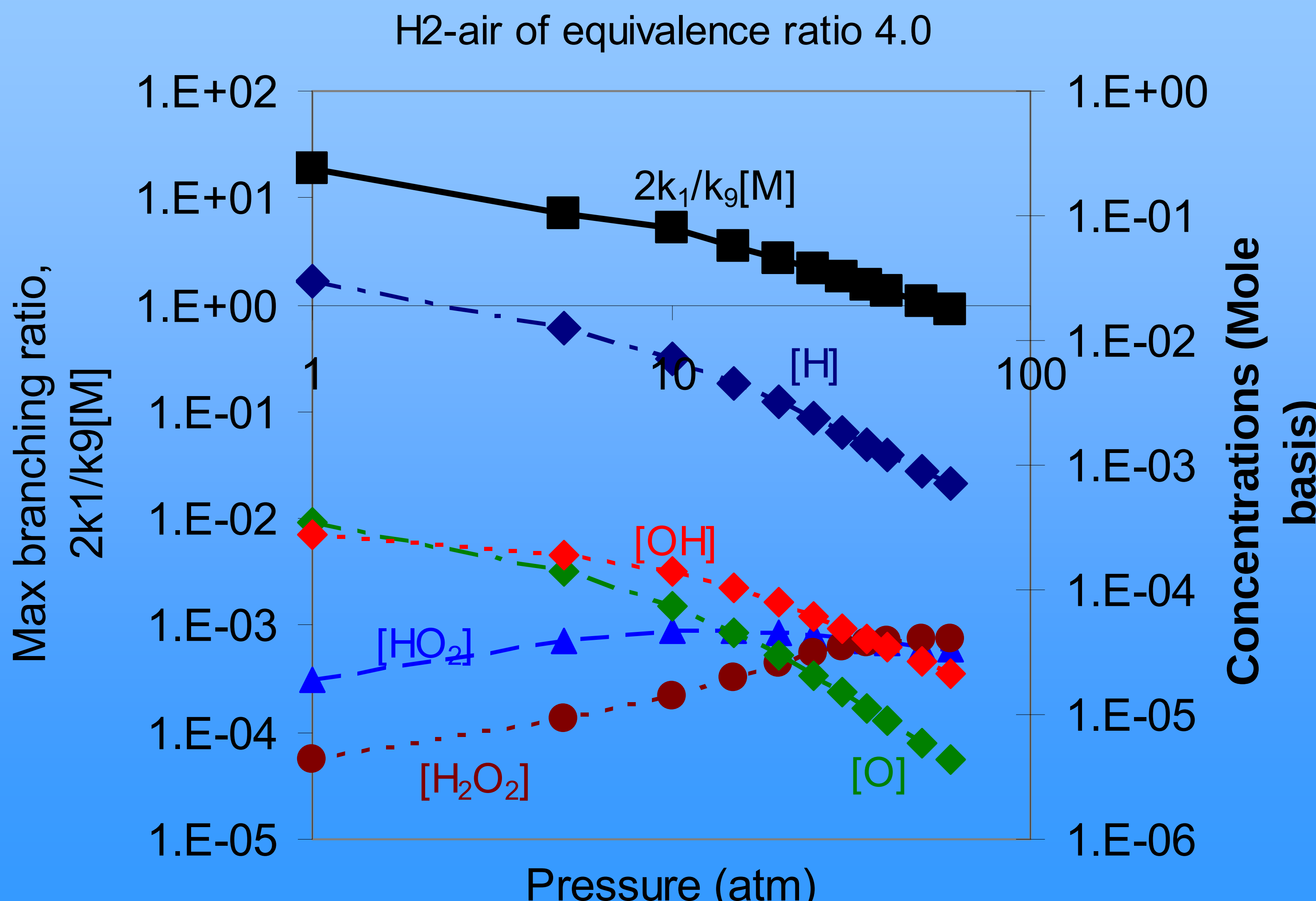
*favored at higher pressures and lower temperatures

- Extended Second Limit – demarcates 2 distinct kinetic regimes

$$\frac{2k_1}{k_9[M]} < 1 \longrightarrow$$ Overall mechanism is chain branching

$$\frac{2k_1}{k_9[M]} > 1 \longrightarrow$$ Overall mechanism is chain terminating

Branching Ratios and Radical Pool Decrease with Pressure



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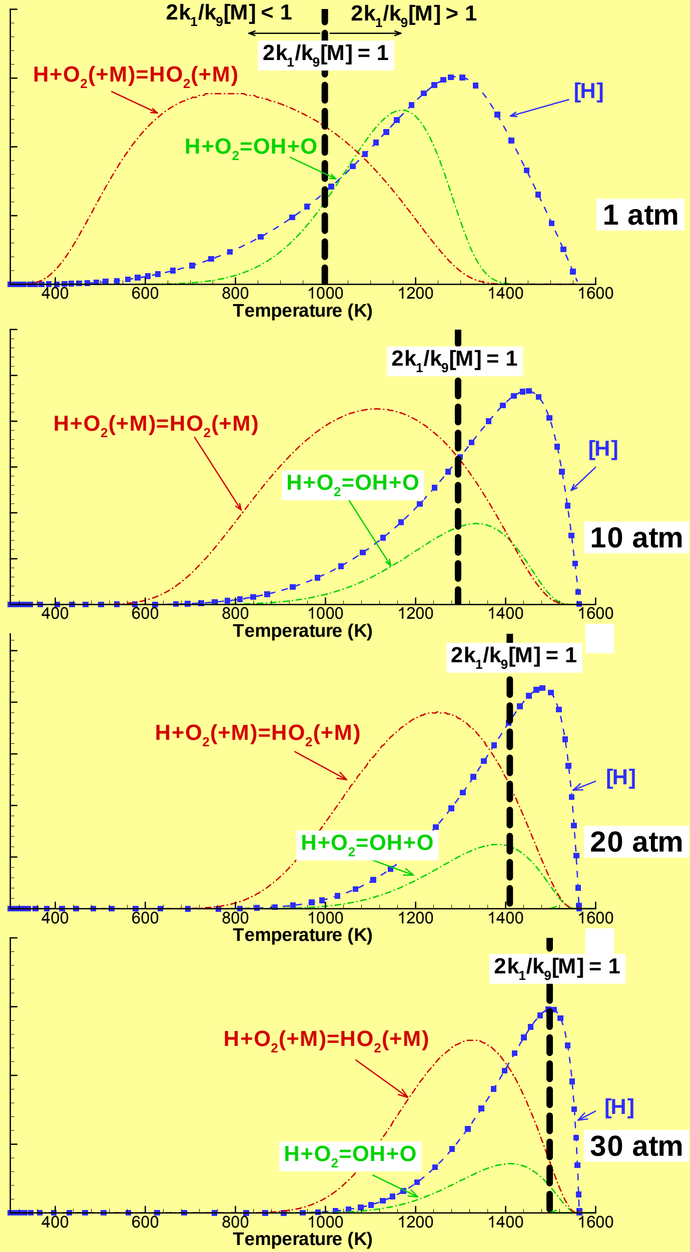
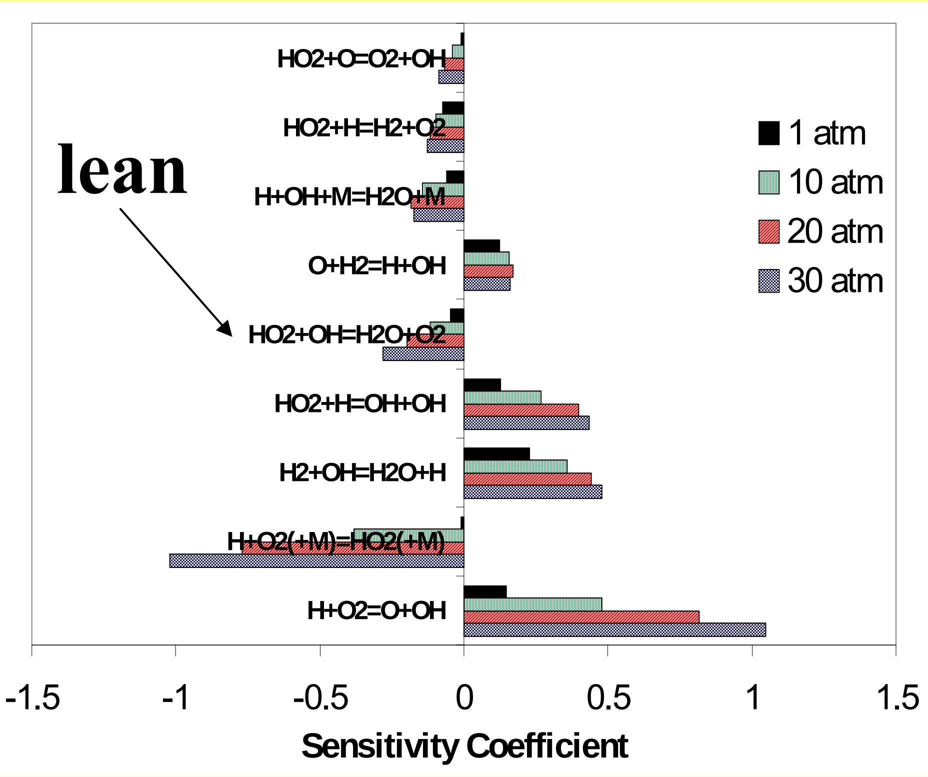
Flux Analysis

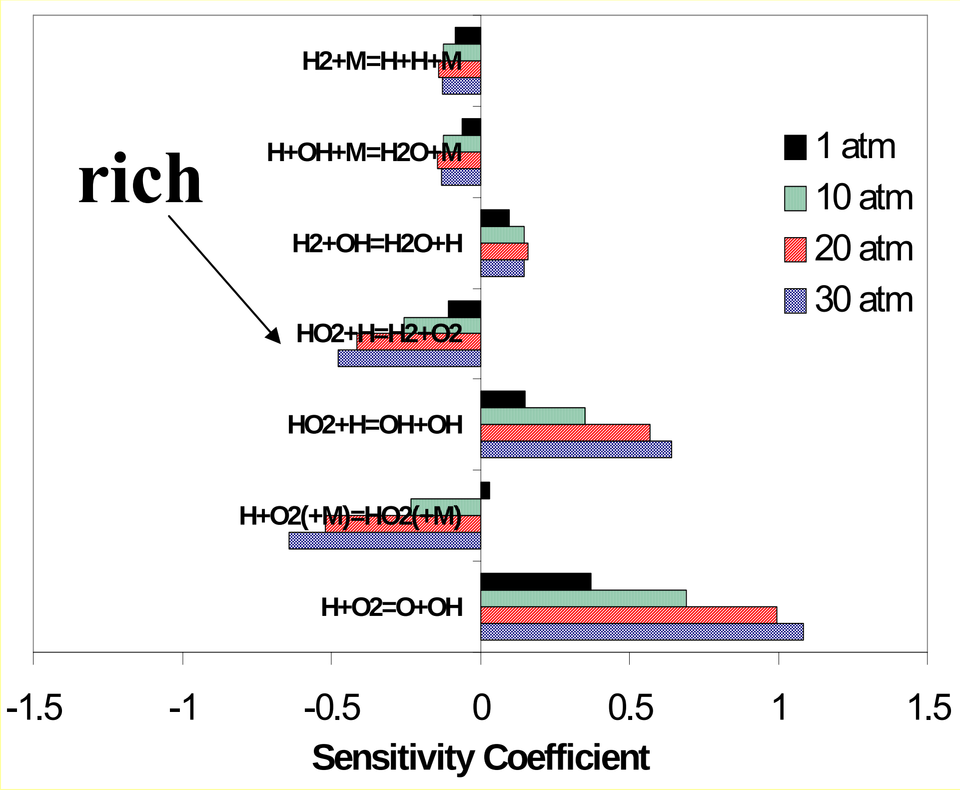


Extended Second Limit →

- Extended Second Limit, $2k_1/k_9[M]=1$, pushed to higher temperatures with increasing pressure
  - Raises the overall activation energy
    →Pushes the radical concentration and reaction flux profiles toward the back end of the flame
  - Extended second limit as a chemical singularity
    →Sensitivity to rate parameters blows up
    →Small differences in parameters make a big difference

Sensitivity Analysis

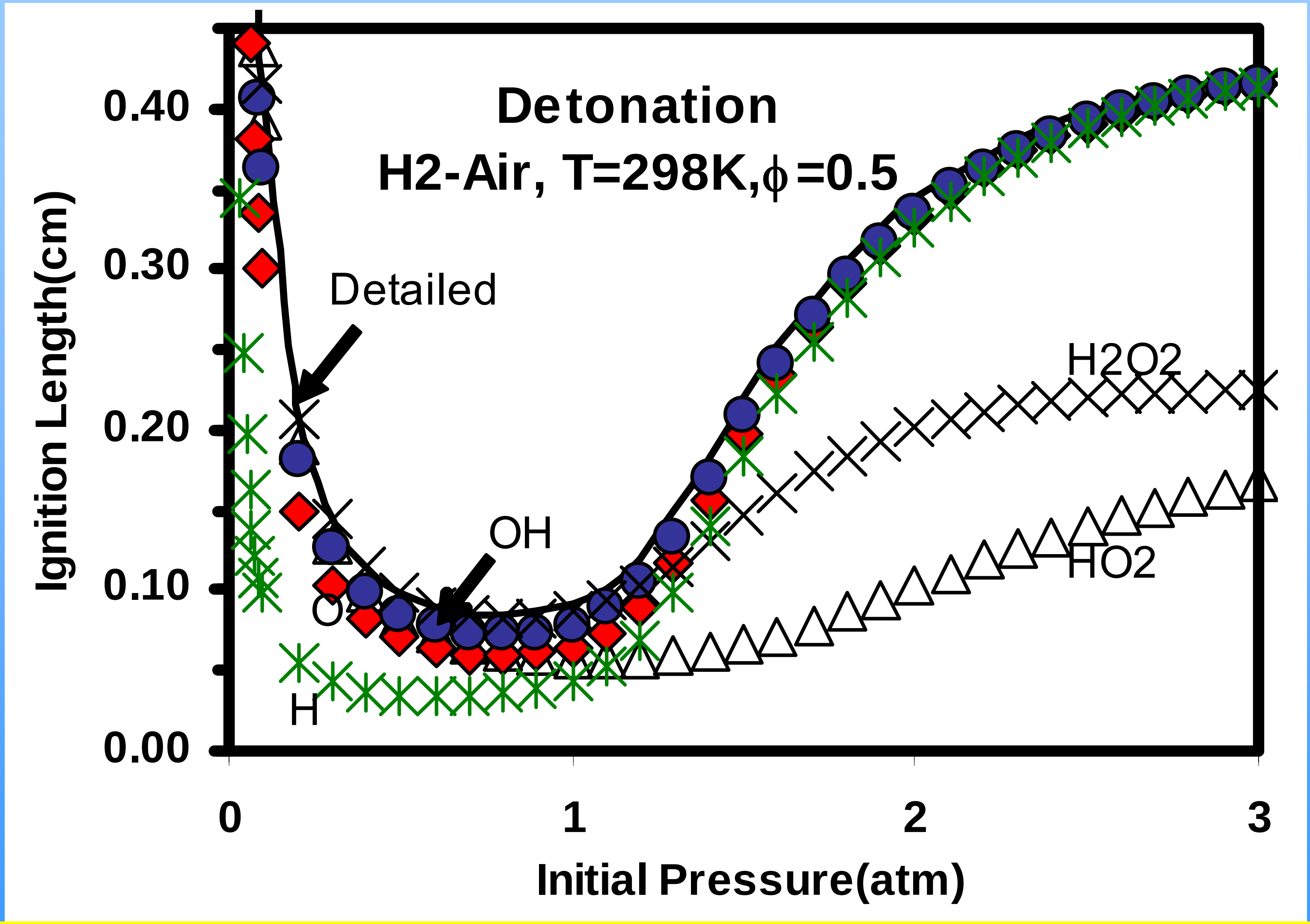




- Competition of $H+O_2$ and $H+O_2+M$ becomes more sensitive with increasing pressure
- HO_2 + H/OH reactions become more sensitive with increasing pressure at rich and lean conditions.
- No simple fix of the kinetic mechanism.

3. Development of reduced order kinetic mechanism

A close look at hydrogen mechanism vs. QSS species



No species can be reduced! How to reduce chemistry size?

Time scales are very different!

A dynamic multi-scale (DMS) kinetic reduction model

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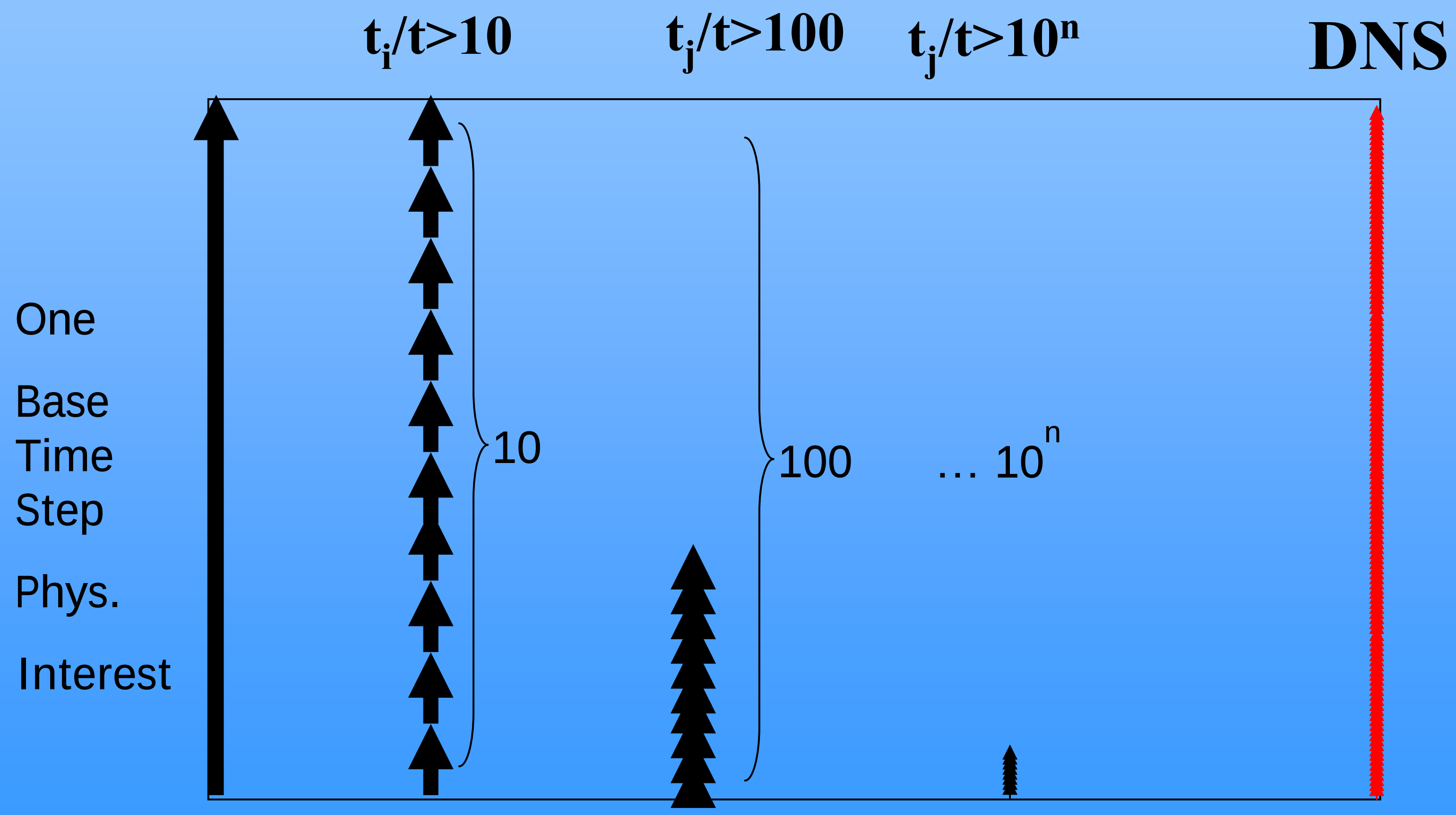
Time scales in reactive flow

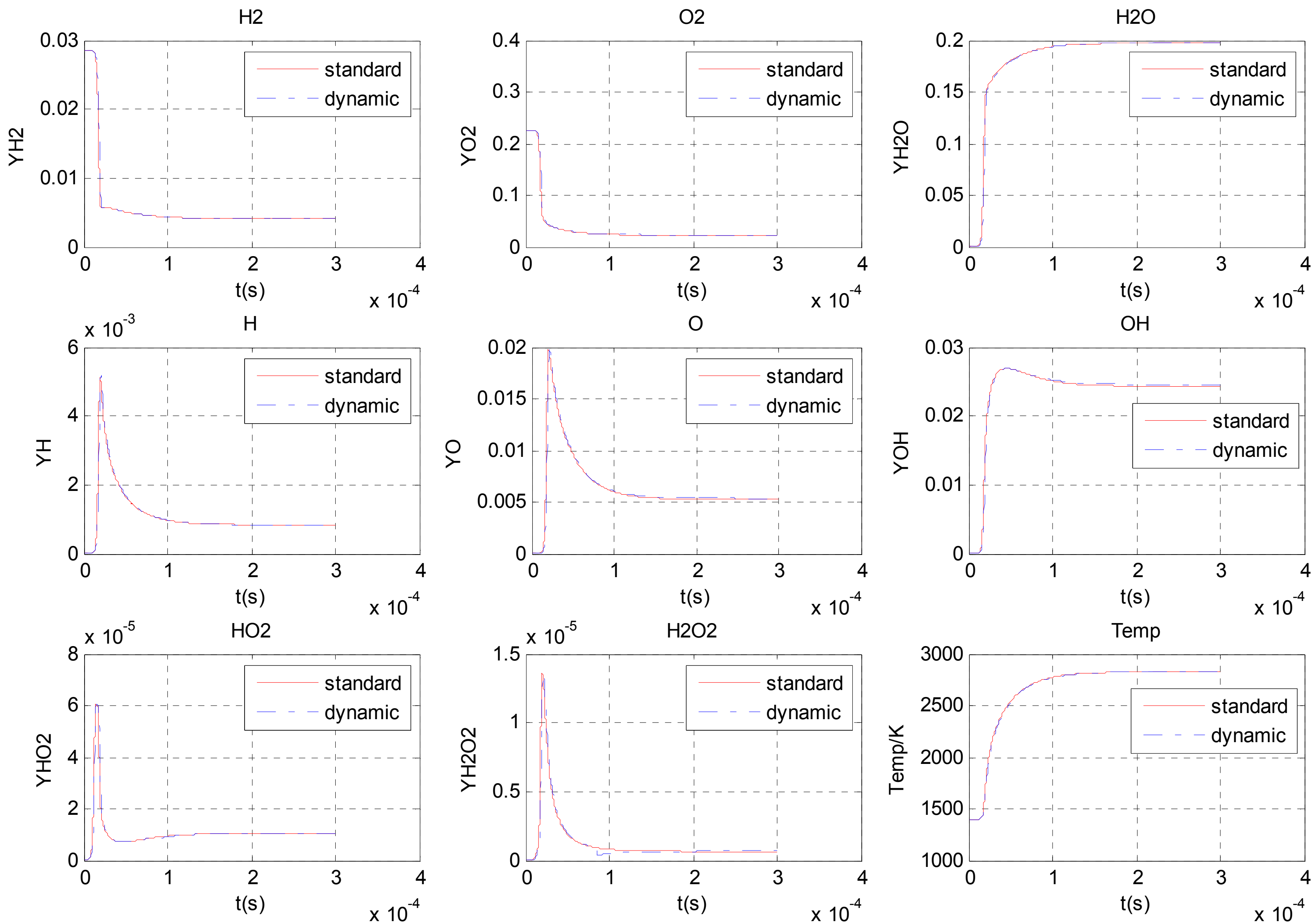
Chemical time		Physical time	
	1 s		
NO formation	10^{-2}s	Flow time	
Product formation H2O	10^{-4}s	Transport time Molecular Turbulent	Physical interest
Radical formation H	10^{-6}s		
Radical formation H2O2 quasi-steady state?	10^{-8}s	Radiation transfer quasi-steady state	?
	10^{-10}s		DNS Time-step

Algorithm of Dynamic Multi-Scale (DMS) Modeling

Every species decays at its own time scale: $Y_i = Y_{i0} e^{-\frac{t}{\tau_i}}$



Validation of DMS method (hydrogen)



Computational efficiency of DMS method Hydrogen/air ignition

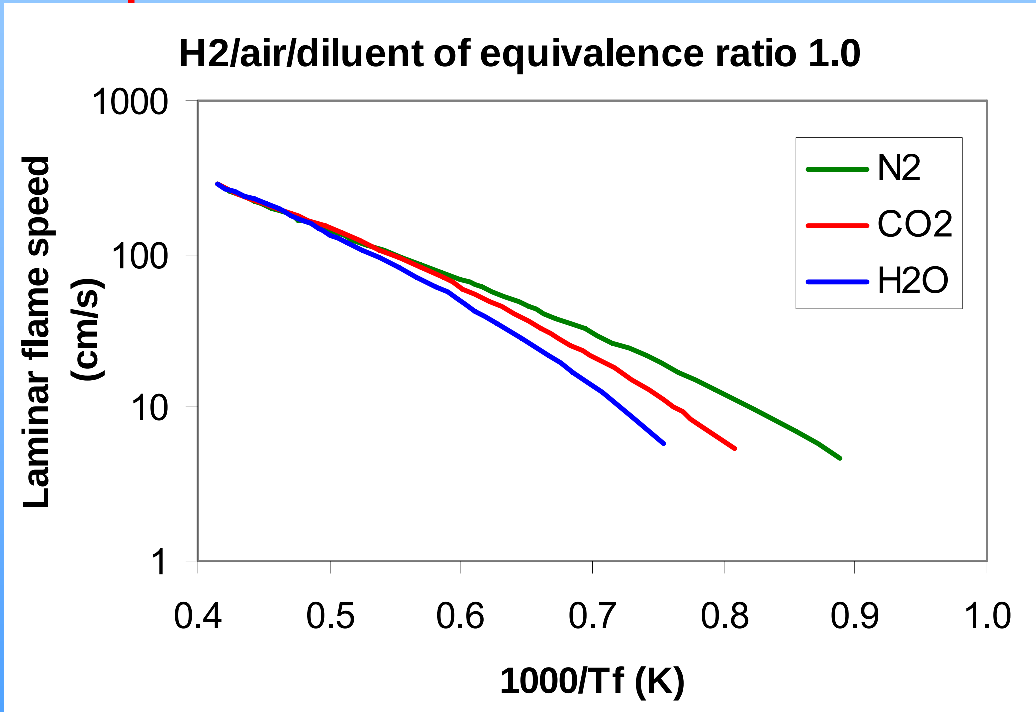
No.	Max. calculation times for groups	Base time step	Average calculation time (s)	Figure No.
For fix time step Euler				
1	1	Step=Min. characteristic time	5.8	
For Static Multi-scale(2 Groups)				
1	9	Min. characteristic time	5.3	
2	5	Min. characteristic time	3.8	
3	3	Min. characteristic time	2.9	
4	2	Min. characteristic time	1.9	
For Dynamic Multi-scale method vs. Direct numerical simulation of Euler equation, 6.66 s needed				
1	50%	Min. characteristic time*100	0.57 s	DMTS-11

With a detailed mechanism and no QSS assumption, Computation is speeded 10 times!

Conclusions

- A rigorous experimental approach for flame speed measurement at high pressures was developed.
- Hydrogen flame speed has a positive dependence on pressure up to 15 atm, but a negative dependence at higher pressures.
- No existing models reproduce measured flame speeds of high pressure rich hydrogen flames.
- A dynamic multiscale model is developed to reduce computational time by one order.

Flame Speeds with Preheat and H_2O dilution (with Counterflow Flames)



Methane diffusion flame with 26% H_2O dilution in fuel stream

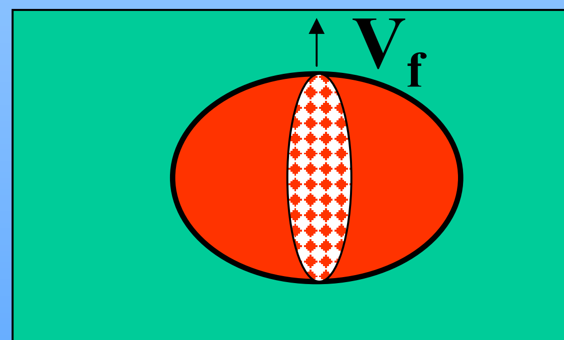
- Performed preliminary tests with PIV system and evaporator
- Currently refining the system to prepare for gathering data

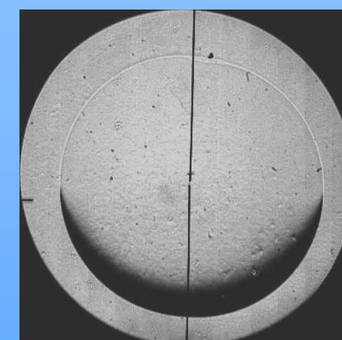
High pressure flame speed (S_u) measurement

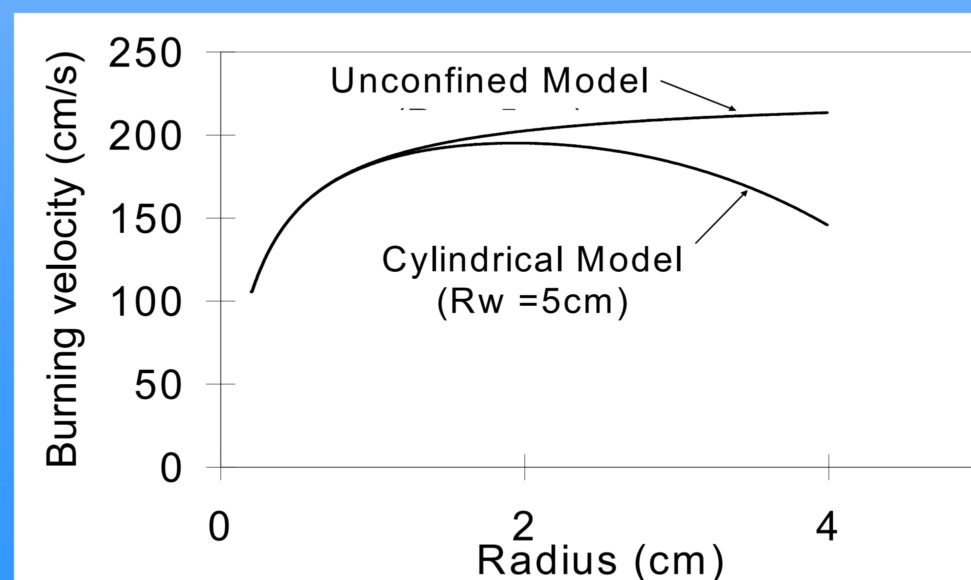
Effect of non-spherical flow, $u_b \neq 0$

$$\frac{s_u^* - s_u}{s_u} = (\sigma - 1) \cdot \frac{\Delta u_f}{u_f} = ?$$

Collimated beam

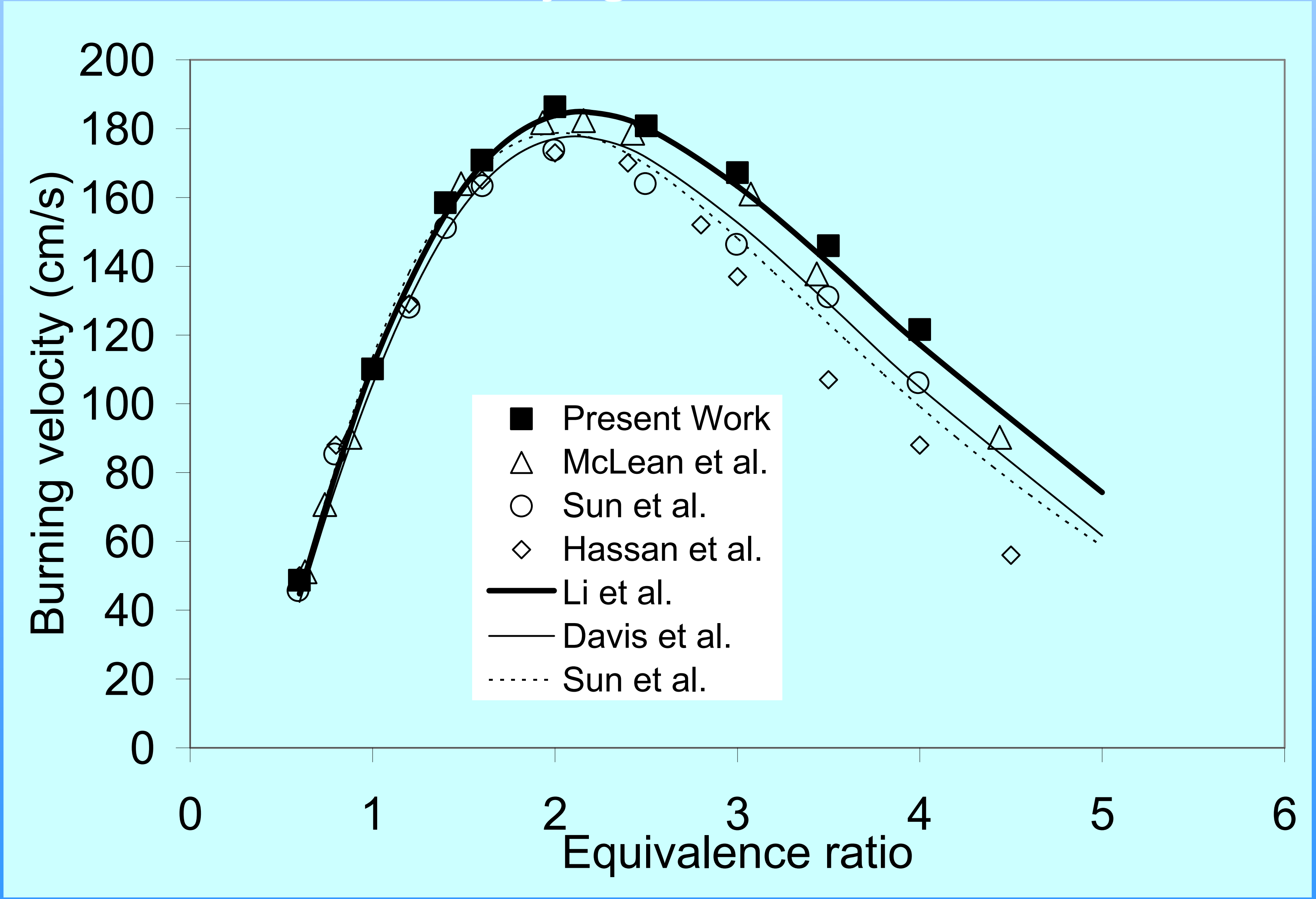






Compression of measured hydrogen syngas flame speed

H2-CO = 50:50 syngas mixture in air at 1 atm



Burke, Qin, Ju & Dryer 2007

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