Flame Chemistry, Mechanism Reduction, and Computational Flame Diagnostics

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1. Nonlinear extrapolation for laminar flame speeds
2. Laminar flame speeds of cyclo-alkanes at elevated pressures
3. NTC-affected ignition in nonpremixed counterflow

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1. Explosive mode analysis of NTC-affected chemical stabilities
2. Computational flame diagnostics

1. Nonlinear Extrapolation for Laminar Flame Speeds

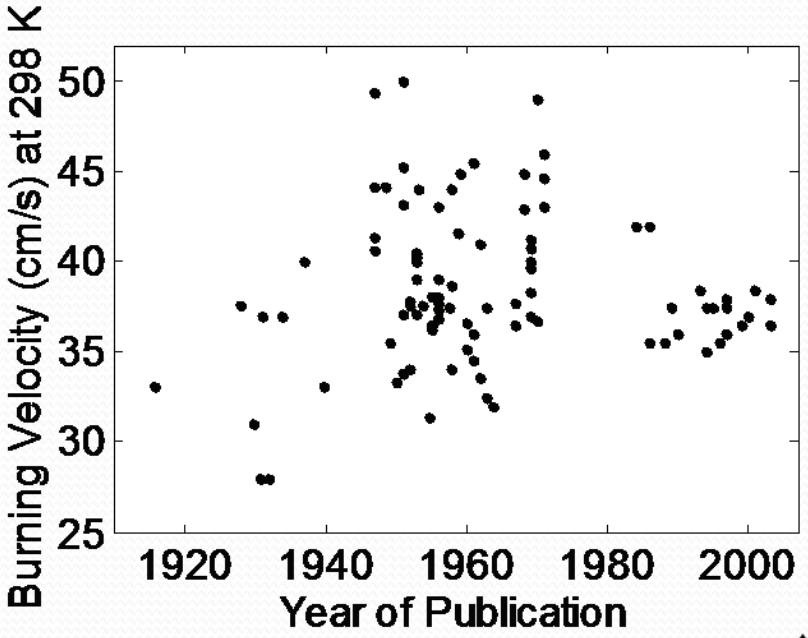
Need to Eliminate Stretch Effects

- Earlier experimental flame speeds "contaminated" by stretch effects
- Extensive data on s_L have been acquired through linear extrapolation of

$$s \approx s_L + L\kappa$$

- Recent studies on large fuel molecules (hence Large L) show strong nonlinear variation

Reported Experimental Measurements of the Maximum Burning Velocity for Methane/Air Flames at 1 atm, 298K

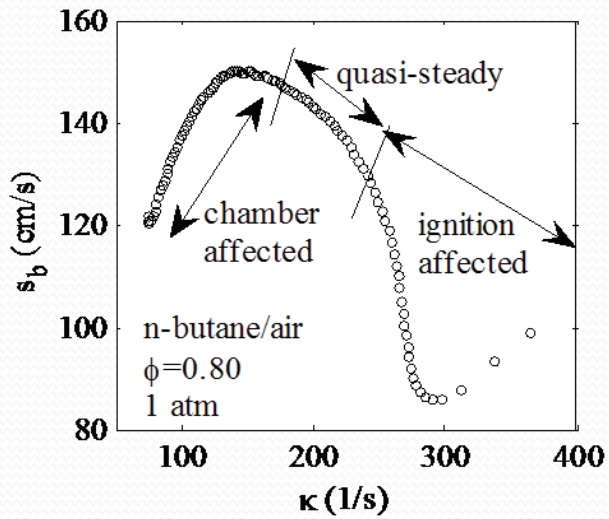


Strong Nonlinearity in Expanding Flames

- Care needed in selecting freely-propagating data
- Useable data exhibit substantial nonlinearity
- Use (quasi-steady) nonlinear flame speed expression in fitting

$$\left(\frac{s}{s^0}\right)^2 \ln\left(\frac{s}{s^0}\right)^2 = -2\frac{L\,\kappa}{s^0}$$

- Analysis still may not be sufficiently rigorous



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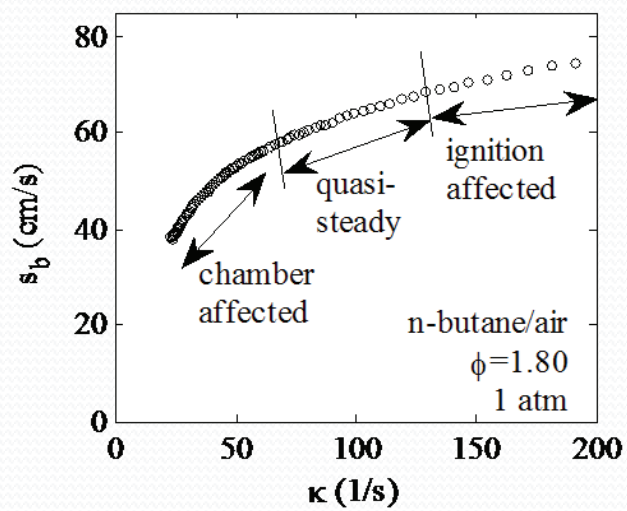

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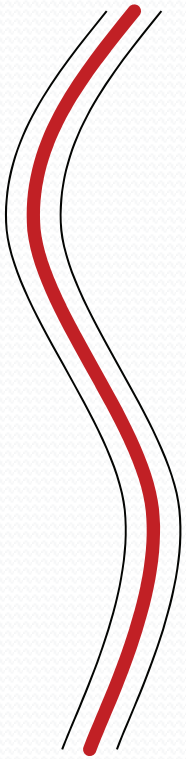
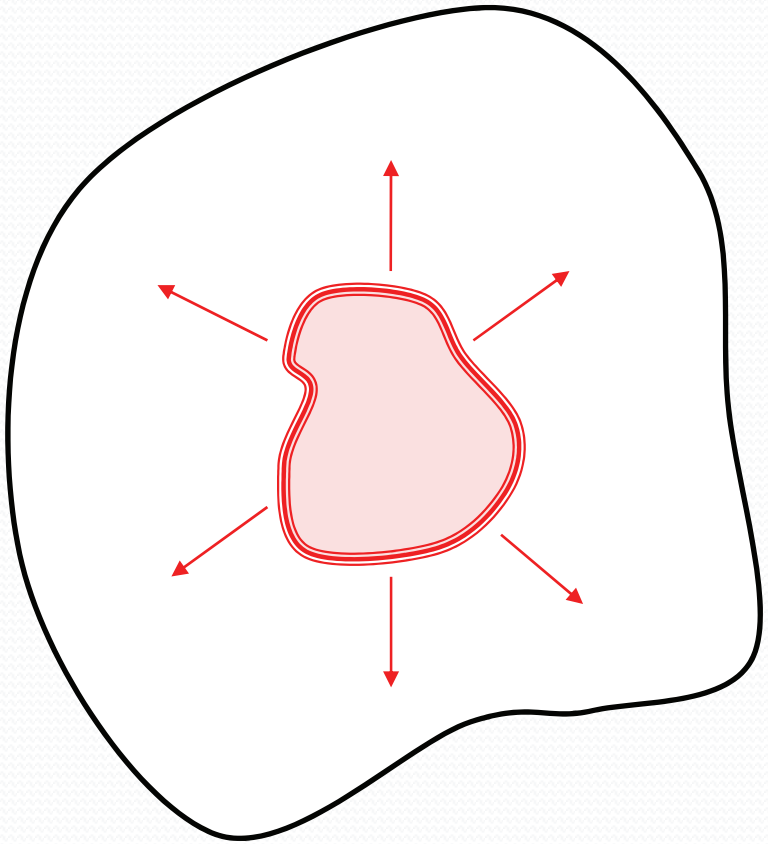
Key Elements of Analysis



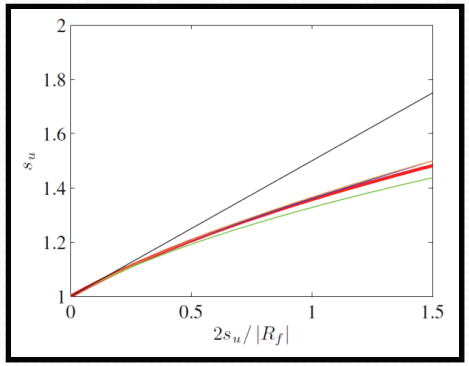
Thin Flame



Arbitrary Geometry

Extrapolation Relations for Unconfined Spherical Flames

Description	Equation	Expansion
Present Result	$s_u^2 \ln s_u^2 = \frac{ds_u}{dR_f} - \frac{2}{R_f} s_u$	$s_u \left(1 + \frac{1}{R_f} + \frac{1}{R_f^2} + \frac{2}{3}\frac{1}{R_f^3}\right) = 1$
Quasi-Steady	$s_u^2 \ln s_u^2 = -\frac{2}{R_f} s_u$	$s_u \left(1 + \frac{1}{R_f} + \frac{3}{2}\frac{1}{R_f^2} + \frac{8}{3}\frac{1}{R_f^3}\right) = 1$
Markstein	$s_u = 1 - \frac{1}{R_f}$	$s_u \left(1 + \frac{1}{R_f} + \frac{1}{R_f^2} + \frac{1}{R_f^3}\right) = 1$
Linear	$s_u = 1 - \frac{s_u}{R_f}$	$s_u \left(1 + \frac{1}{R_f}\right) = 1$



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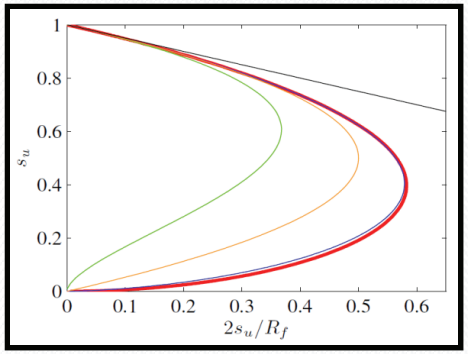

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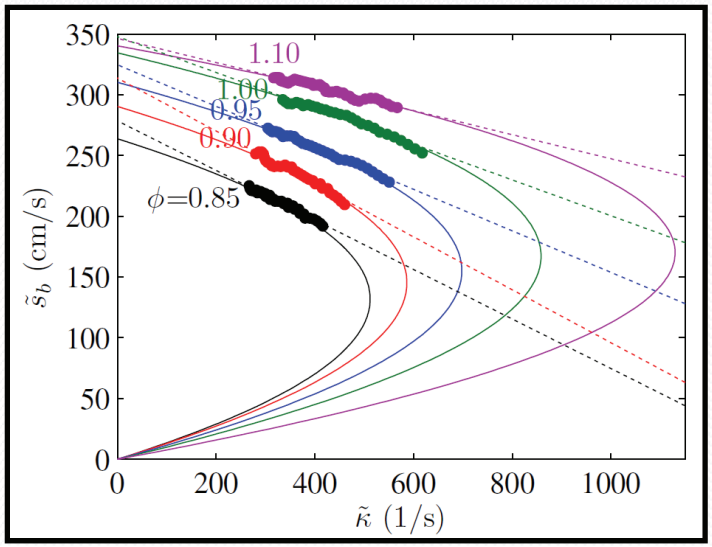
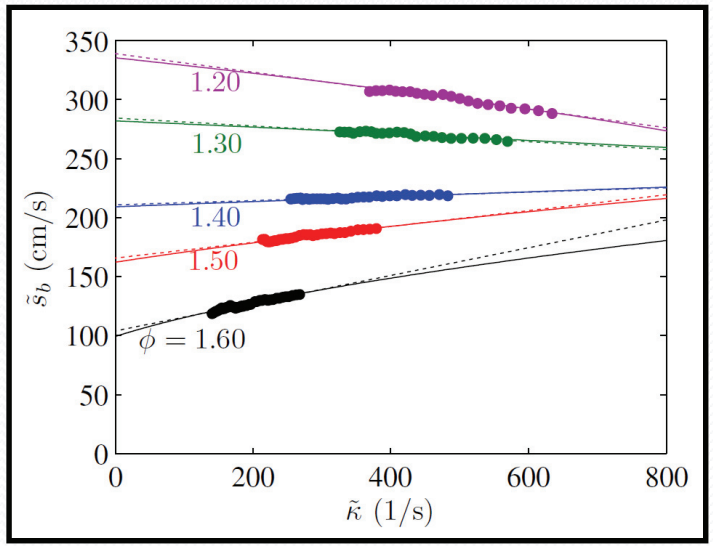
Representative Extrapolations





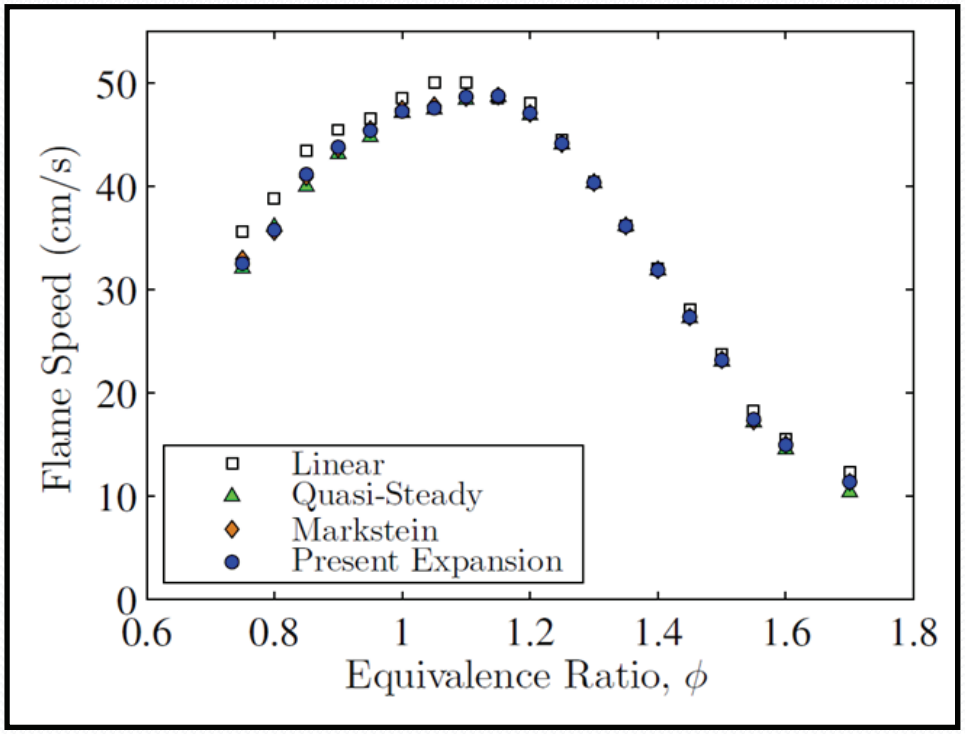
- Present Expansion Integrated (Dimensional Form):

$$\tilde{s}_b^0\tilde{t} + \tilde{c} = \tilde{r}_f + 2\tilde{L}_b \ln \tilde{r}_f - 4\frac{\tilde{L}_b^2}{\tilde{r}_f} - \frac{8}{3}\frac{\tilde{L}_b^3}{\tilde{r}_f^2}$$

- Linear Expression Integrated (Dimensional Form):

$$\tilde{s}_b^0\tilde{t} + \tilde{c} = \tilde{r}_f + 2\tilde{L}_b \ln \tilde{r}_f$$

Representative Extrapolated Data



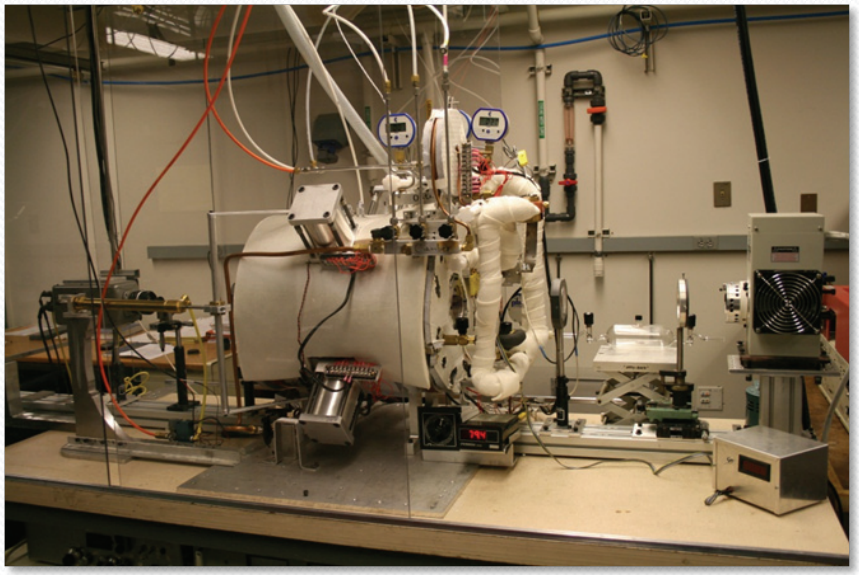
N-Heptane/air, 1atm., 353 K

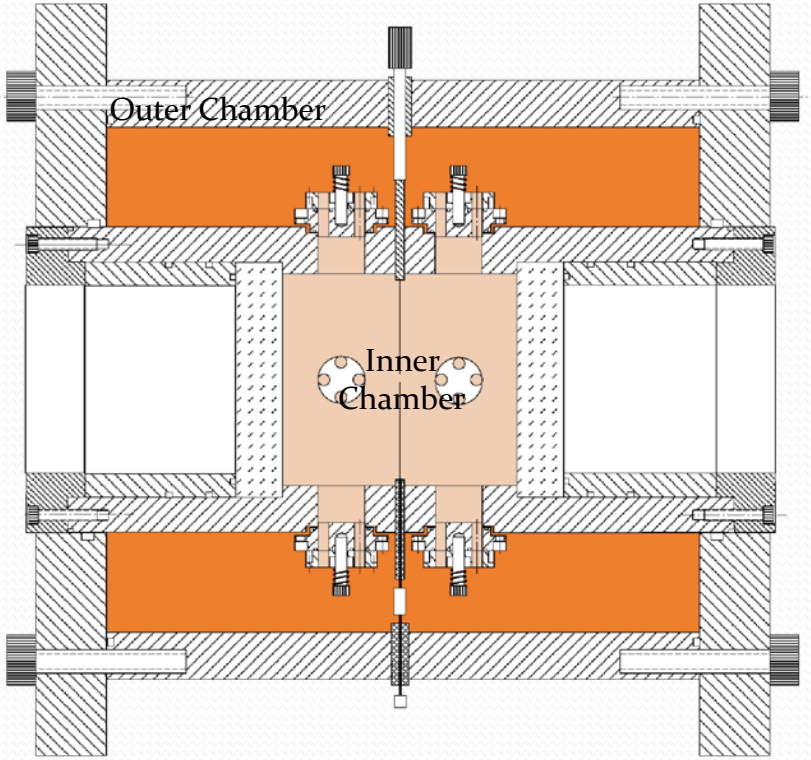
2. Laminar Flame Speeds of Cyclo-alkanes

Motivation

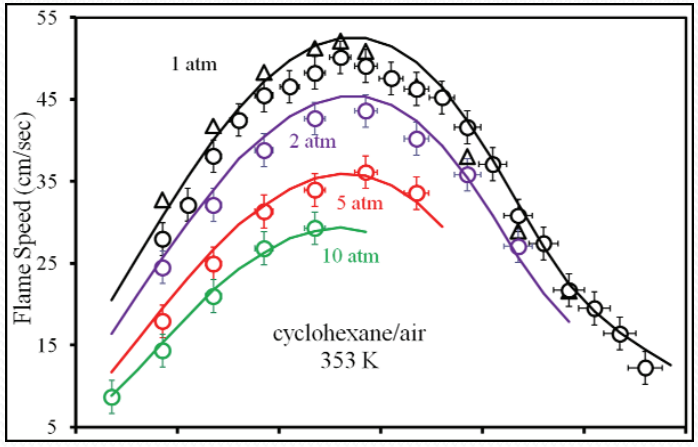
- Cyclohexane and its mono-alkylated cyclohexanes are components of surrogate fuels
- Only atmospheric laminar flame speeds have been experimentally determined for these fuels (Ji & Egolfopoulos, 2011), showing reduced flame speeds for the mono-alkylated cyclohexanes.
- Lower flame speeds attributed to the 3-body termination reactions
- Need high-pressure flame speed data:
  - For mechanism development
  - Accentuate the pressure-dependent 3-body termination reaction

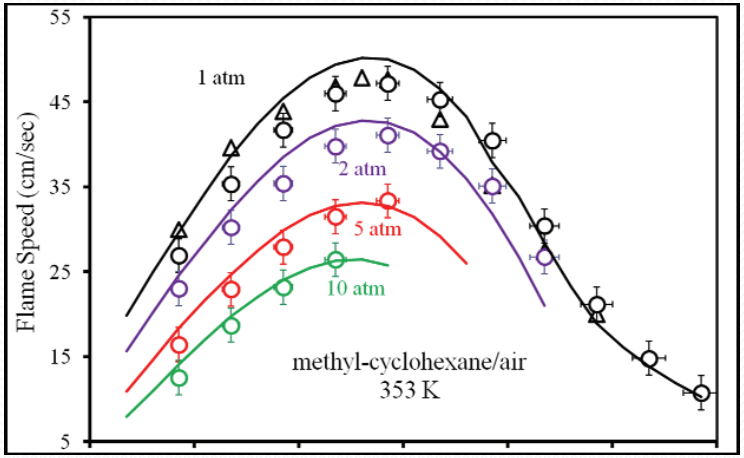
Experimental Configuration

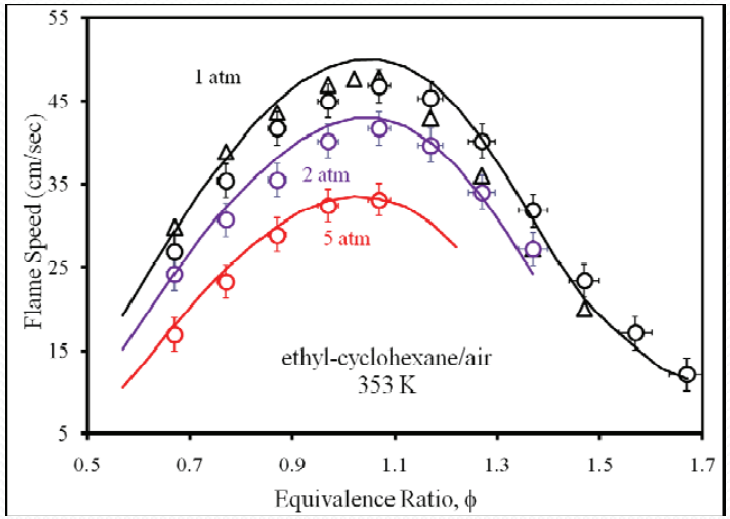




Representative Data and Comparison: Pressure = 1 atm

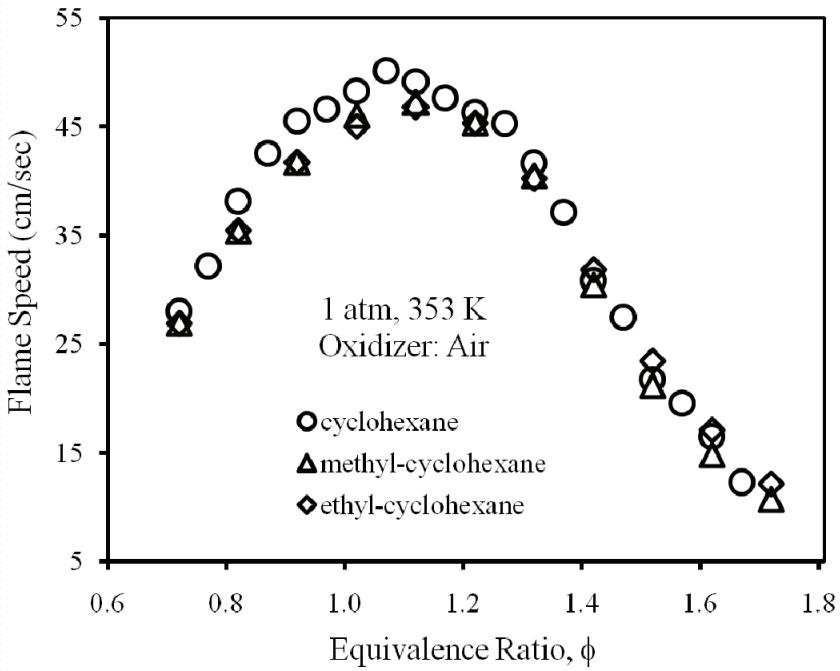


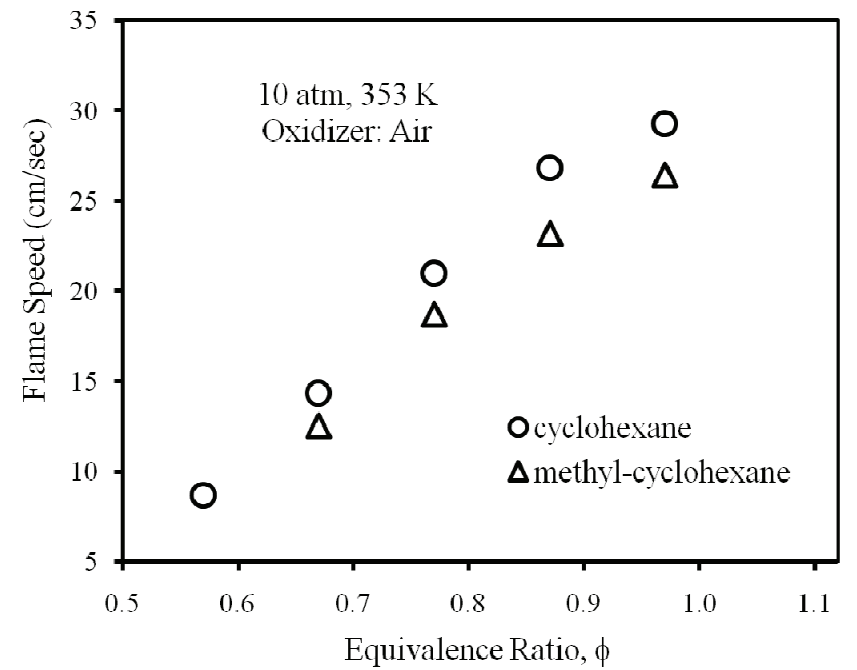




Triangles: Ji & Egolfopoulos
Circles: Present
Lines: JetSurF 2.0

Quantitative High Pressure Effects



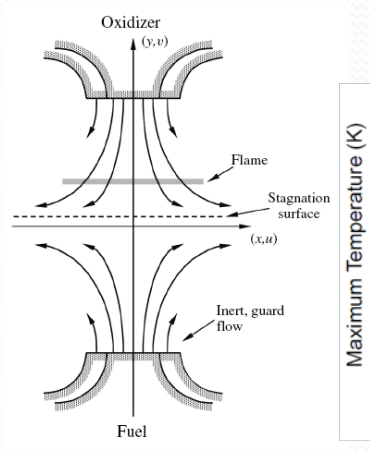


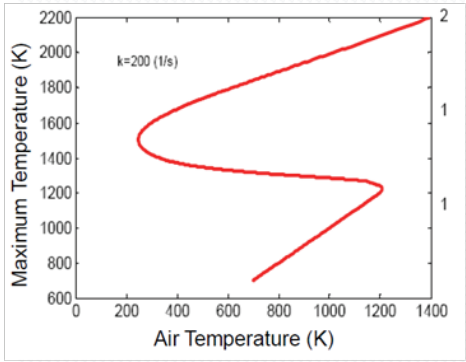
- At both low and high pressures, methyl- and ethyl-cyclohexanes have similar flame speeds which are lower than those of cyclohexane
- The difference increases (5% vs.13%) from 1 atm to 10 atm.
- Additional data up to 20 atm.

3. NTC-Affected Ignition in Nonpremixed Counterflow

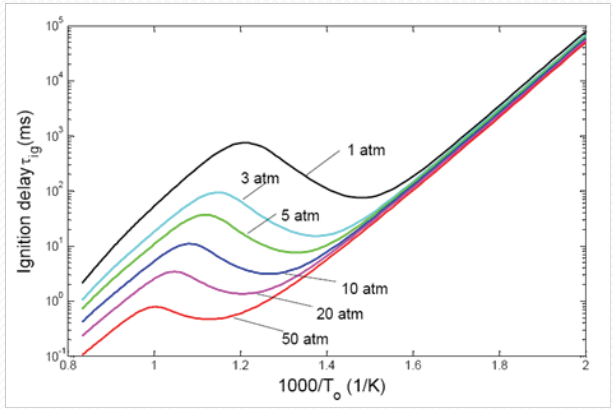
Motivation

- NTC behavior:
  - Extensively observed in homogeneous systems
  - Inferred from detailed computational studies of transport-affected inhomogeneous flows (J.H. Chen; K. Seshadri)
  - Need clear theoretical/experimental manifestation of global ignition & combustion behavior in response to flow straining and nonuniformity (e.g. counterflow)
- Search and identify NTC behavior for nonpremixed counterflow at:
  - low strain rates
  - high pressures





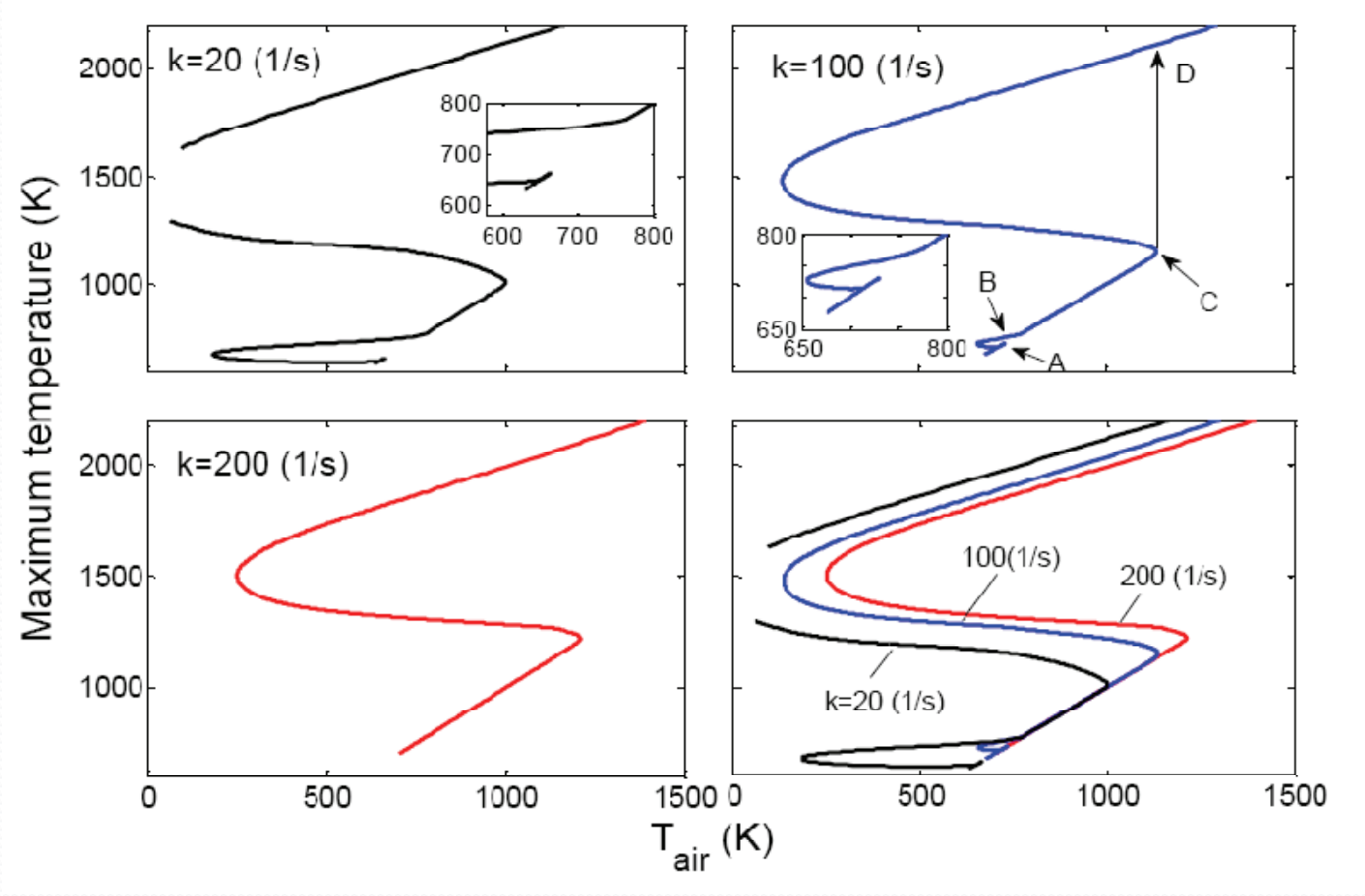
Heptane/air



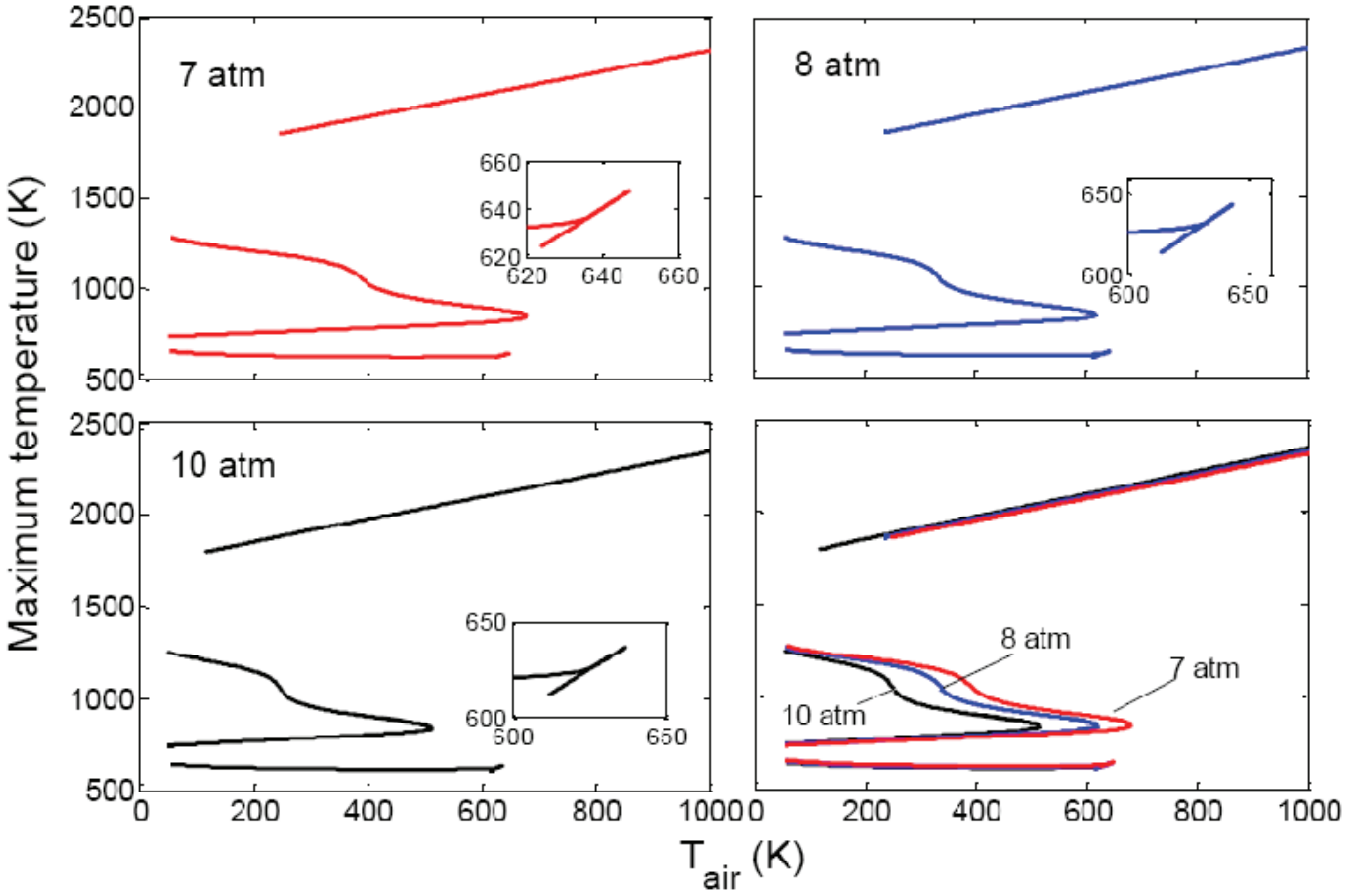
System Specifications

- Nonpremixed diluted n-heptane against heated air
- LLNL detailed mechanism of 561 species and 2539 reactions reduced to a skeletal mechanism of 88 species and 387 reactions using directed relation graph (DRG).
- Qualitatively similar results also obtained for DME

NTC Response Observed at Low Strain Rates (1 atm)

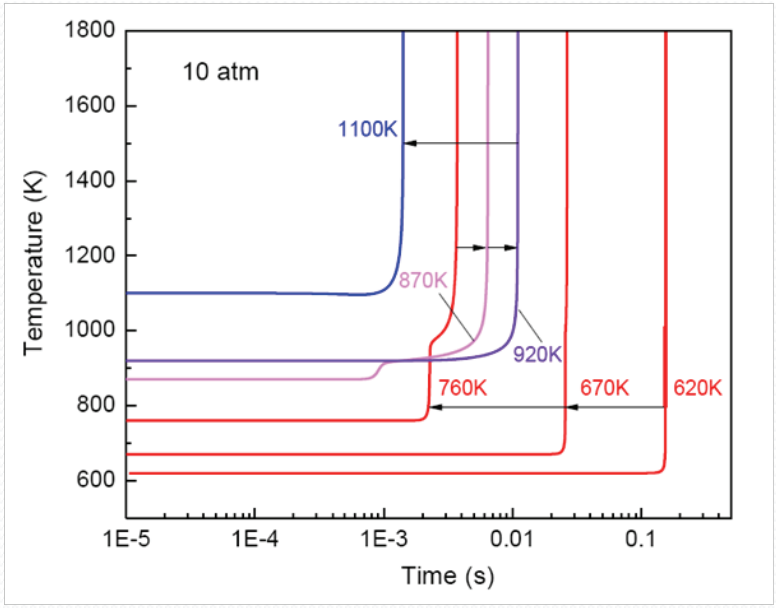


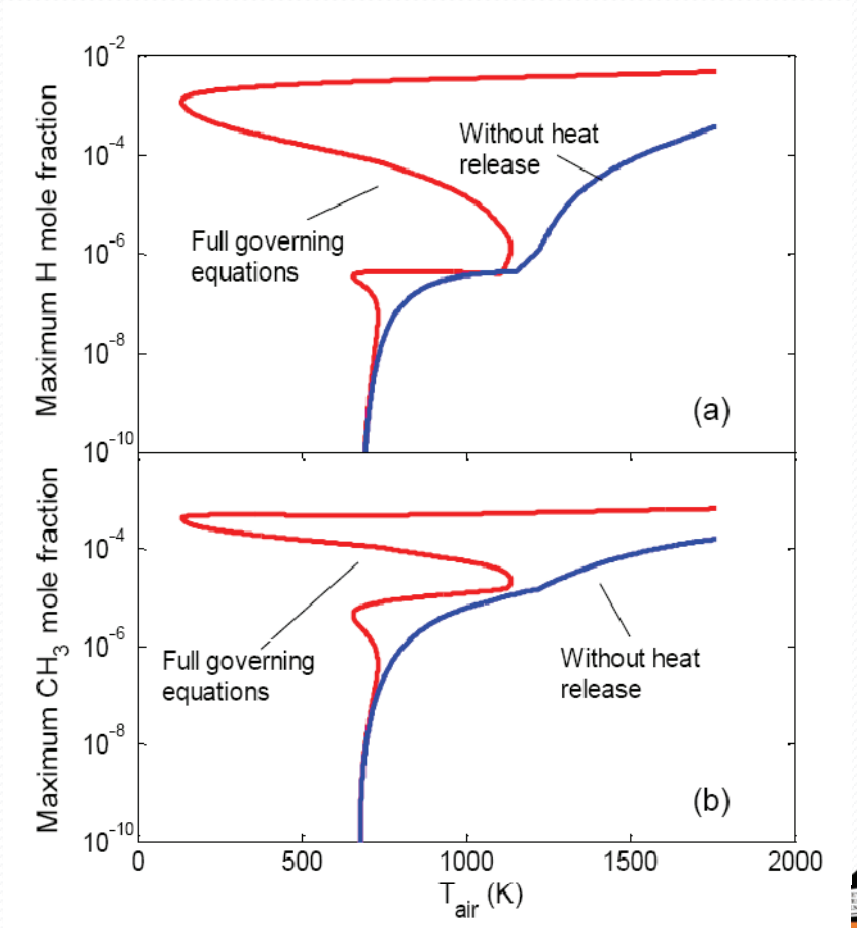
Prominent NTC Response at Elevated Pressures (100/s)



Essential Role of Heat Release

- Both primary and secondary turning points are eliminated by suppressing heat release





Summary of NTC-Affected Behavior

- NTC behavior manifested for flows at low strain rates and/or elevated pressures
- NTC response constitutes a separate (weak) flame system, with distinct (secondary) S-curve ignition-extinction states grafted onto the lower branch of the primary S-curve.
- At lower pressures ignition occurs in two stages with increasing temperature
- At sufficiently high pressures ignition occurs in a single, NTC-control stage

Work Plan

- Determination of laminar flame speeds at elevated pressures:
  - iso-Alkanes: 3-methylheptane; 2,5-dimethylhexane; 2,7-dimethyloctane
  - Aromatics: benzene; toluene; n-propylbenzene; o-xylene; m-xylene; p-xylene; 1,2,4-trimethylbenzene;

    1,3,5-Trimethylbenzene

- NTC-affected flame phenomena
  - Counterflow and stagnation flow ignition experiments on DME and heptane to substantiate theoretical findings
  - Analyze other ignition phenomena (e.g. spark ignition, jet ignition)
  - Study of the role of chemical oscillation