Why hydrogen flames are different: Effects of preferential diffusion on dynamics and stabilization

WEBINAR DUTCH SECTION OF THE COMBUSTION INSTITUTE, SEPTEMBER 24, 2021

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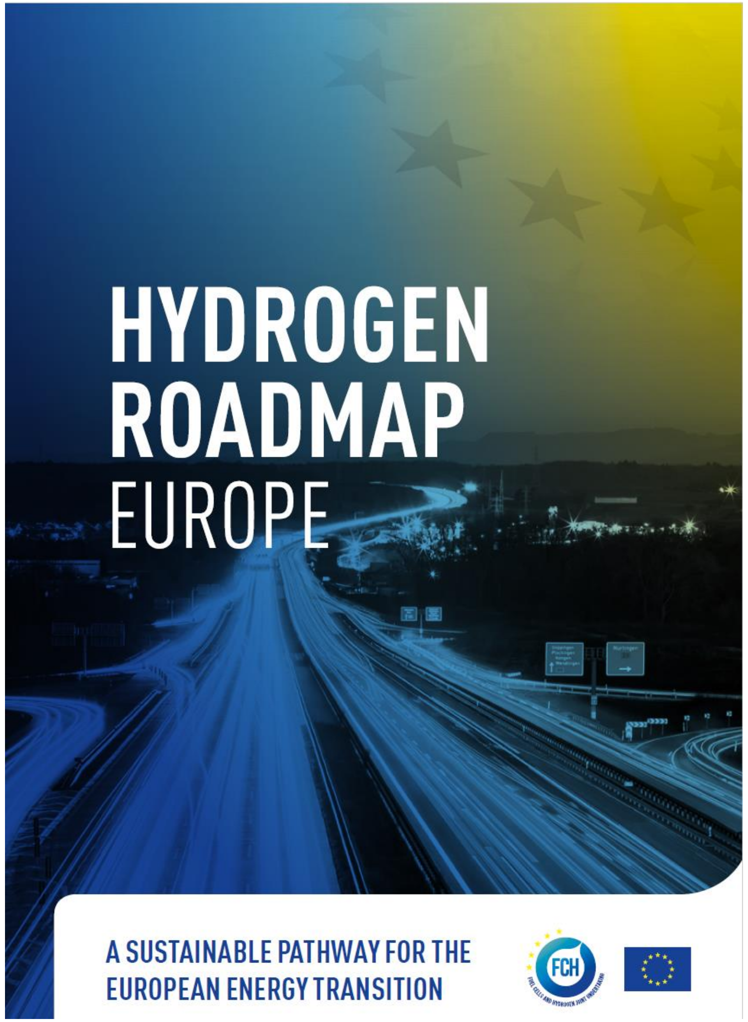
Hydrogen

Hydrogen receives a lot of attention

- No CO_2 is emitted when H_2 is burnt
- Simplest fuel to produce from renewable electricity
- Large scale energy storage
- Large potential for
    - Residential and industrial heat
    - Power generation
    - Transport sector

> “The world is moving ahead on the need to decarbonise and the need to commit to climate neutrality — so in that context the importance of hydrogen increases on almost a daily basis” — *Frans Timmermans, EC EVP for the European Green Deal*



HYDROGEN
ROADMAP
EUROPE

A SUSTAINABLE PATHWAY FOR THE
EUROPEAN ENERGY TRANSITION

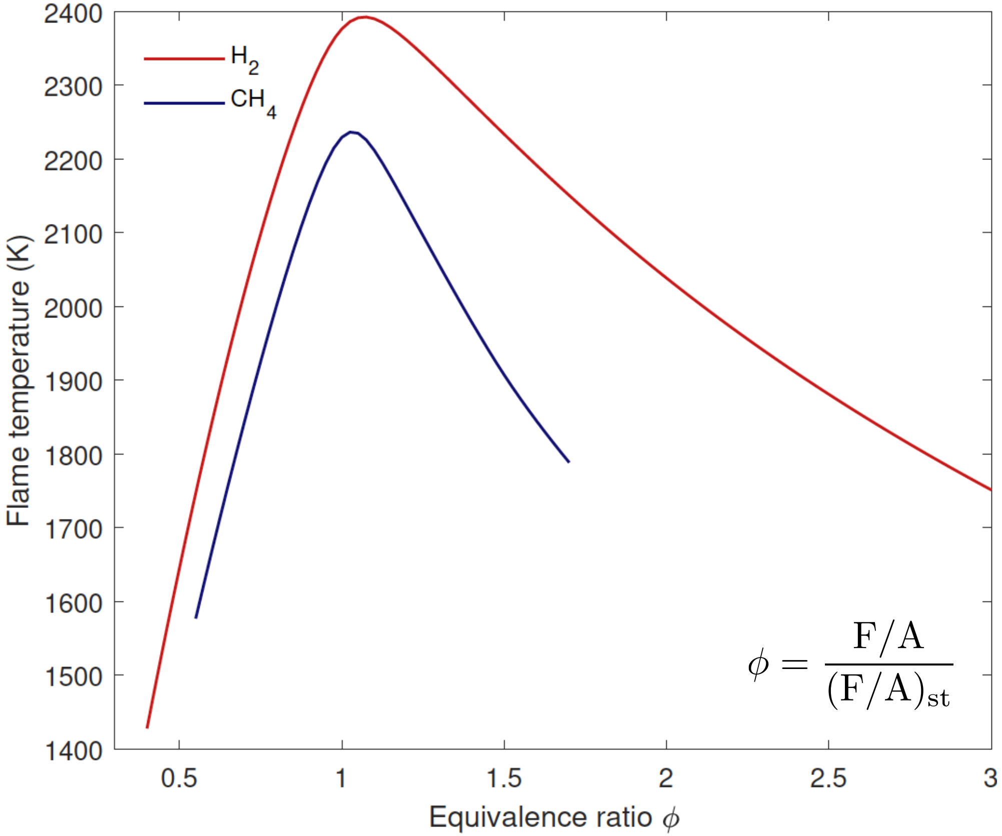
Hydrogen fuel

- Hydrogen as (partial) replacement for natural gas (CH_4)
- Existing combustion equipment is usually not suited for H_2

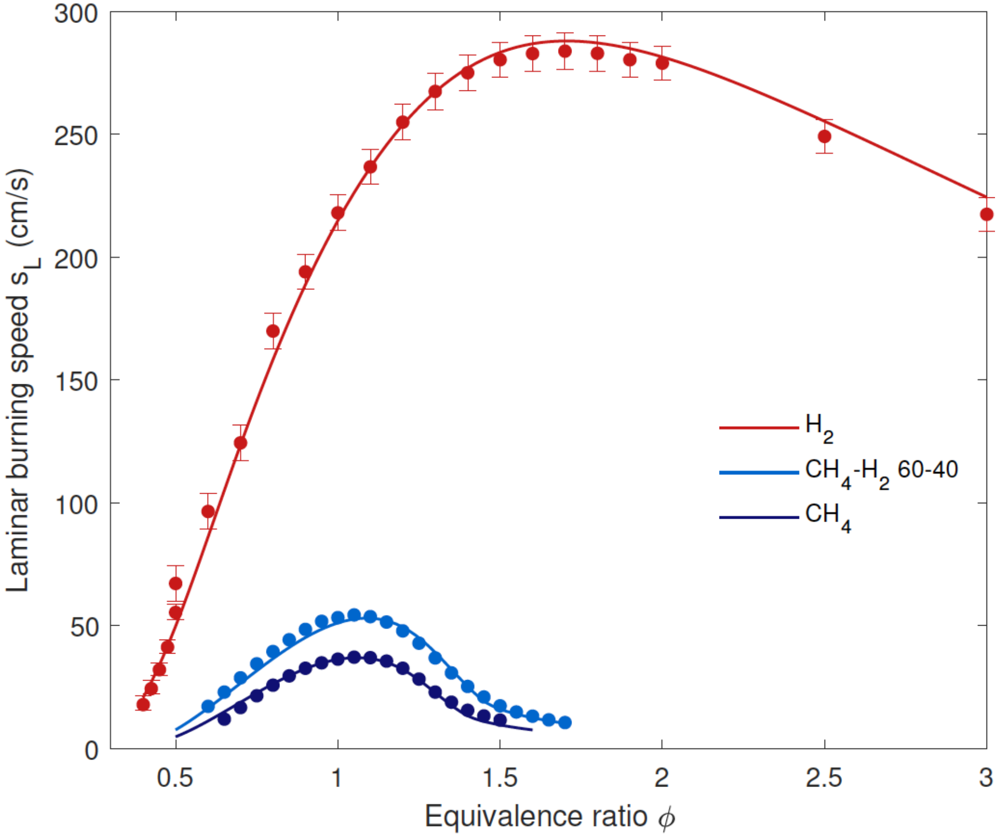
Property	Methane	Hydrogen
Heating value (LHV, MJ/kg)	50	120
Density (kg/m^3)	0.657	0.089
Stoich. Air-Fuel Ratio (mol/mol)	9.5	2.38
Flame temperature (K)	2220	2380
Laminar burning velocity (m/s)	0.37	2.18 (2.84)
Flammability limit (fuel mol%)	5 – 15	4 – 75
Autoignition temperature (K)	~800	~850
Minimum ignition energy (mJ)	0.20	0.02
Diffusivity in air (cm^2/s)	0.21	0.76

At standard conditions

Flame properties



$$\phi = \frac{\mathrm{F/A}}{(\mathrm{F/A})_{\mathrm{st}}}$$



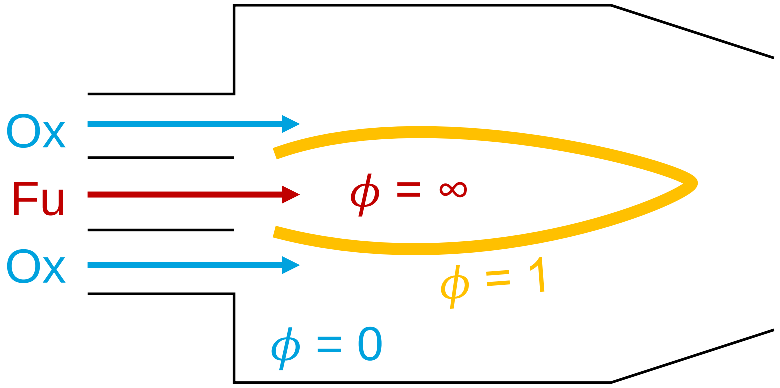
- High heat transfer rate
- High NO_x formation rate

- High power density
- Flame stabilization problems

Modes of combustion

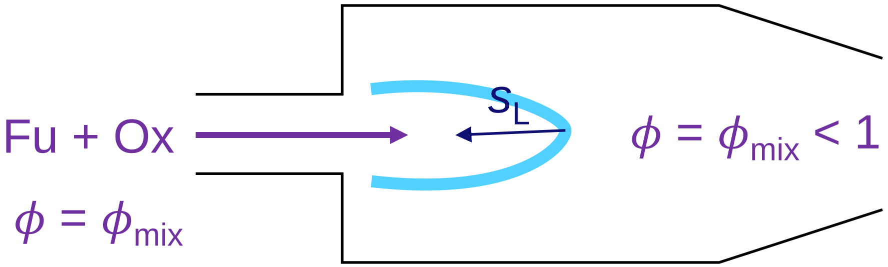
Non-premixed flames

- Reactants are initially separated
- Diffusion/mixing controlled
- Relatively slow conversion
- High flame temperature: high NO_x emissions



Premixed flames

- Reactants are mixed before they enter the reaction zone
- Explosive mixture, propagating front
- Flame stability: Flashback
- Low pollutant emissions



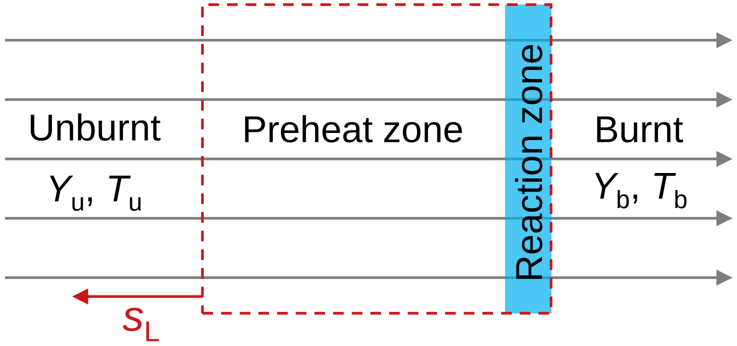
Flame stretch theory

Impact of Lewis number on flame speed

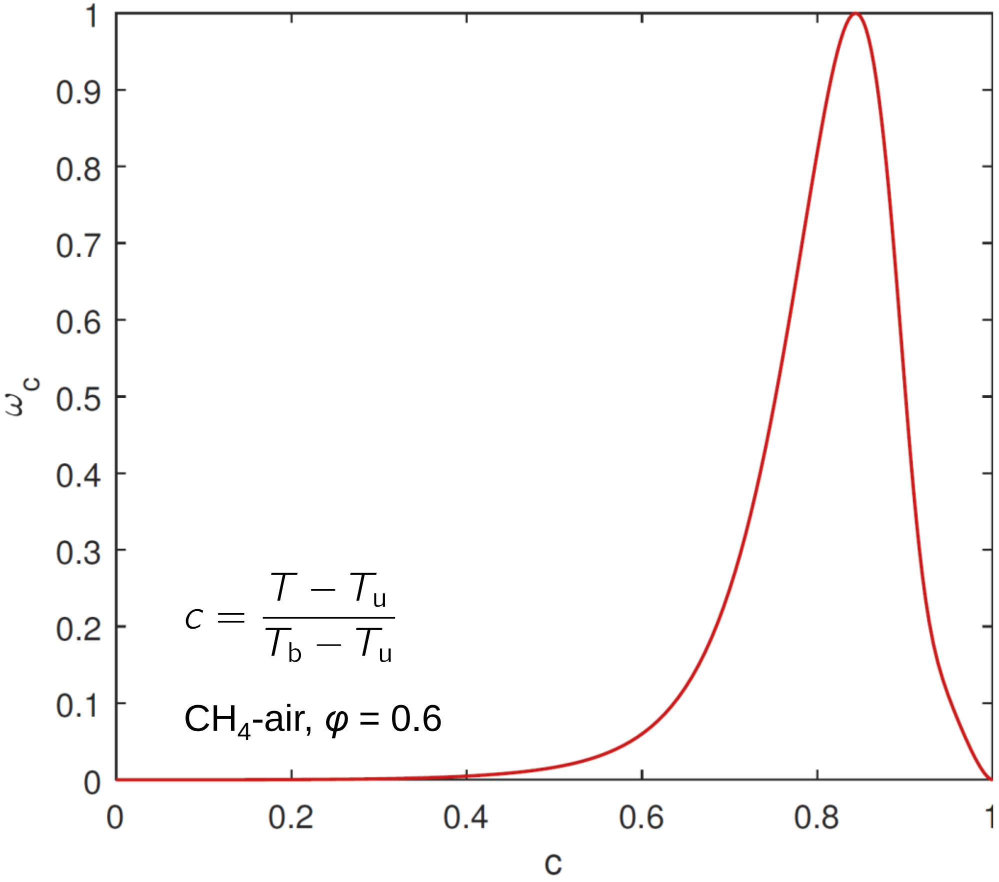
Law (2006), Combustion Physics, Cambridge University Press
Van Oijen et al. (2016) Prog. Energy Combust. Sci. 57:30-74

Premixed laminar flames

- Propagating reaction waves
- Laminar flame speed s_L
- Reaction-diffusion structure
- Large activation energy:
  - Thin reaction zone
  - Heat and mass diffusion zone



- Reaction rates are determined by burnt mixture



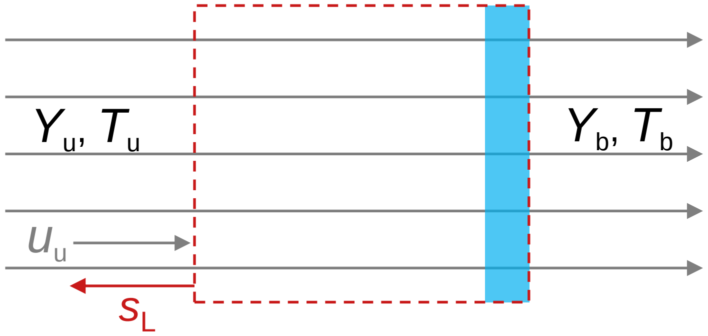
Premixed flame structure

- Governing equations steady 1D case

$$\frac{d}{dx}(\rho u) = 0$$

$$\rho u \frac{dY}{dx} - \rho D \frac{d^2 Y}{dx^2} = -\omega$$

$$\rho u c_p \frac{dT}{dx} - \lambda \frac{d^2 T}{dx^2} = q_c \omega$$



- Solution (preheat zone)

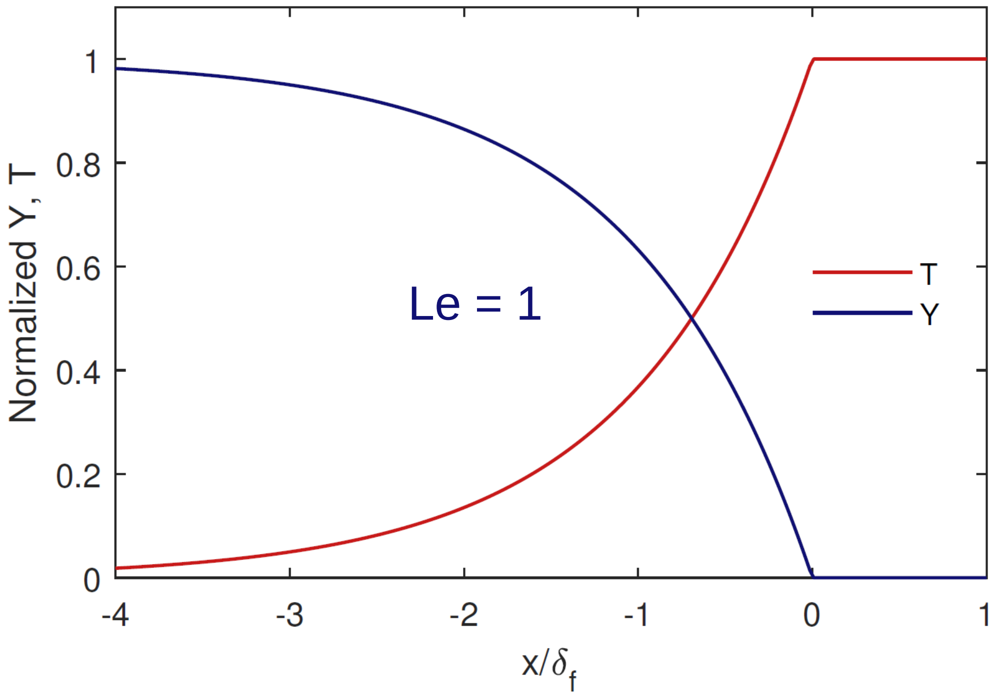
$$\rho u = m = \text{const}$$

$$T = T_\mathrm{u} + (T_\mathrm{b} - T_\mathrm{u}) \exp(x/\delta_f)$$

$$Y = Y_\mathrm{u} - Y_\mathrm{u} \exp(\mathrm{Le}\, x/\delta_f)$$

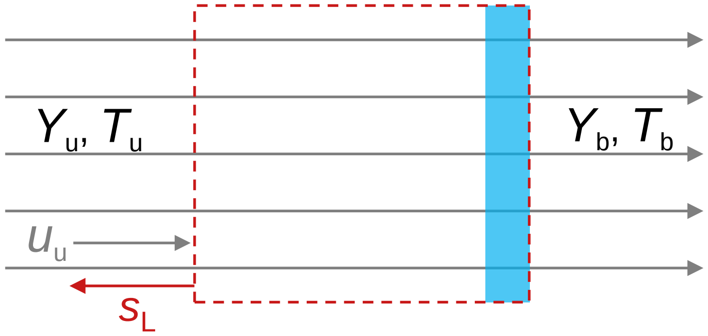
Flame thickness: $\delta_f = \lambda/(m c_p)$

Lewis number: $\mathrm{Le} = \lambda/(\rho D c_p)$



Integral analysis

- Integrate governing equations from unburnt to burnt
- Diffusive fluxes are zero and $Y_b = 0$



$$\frac{d}{dx}(\rho u) = 0$$

$$\rho u \frac{dY}{dx} - \rho D \frac{d^2 Y}{dx^2} = -\omega$$

$$m_u = m_b = m_b^0$$

$$m_u Y_u = \int_u^b \omega \, dx$$

$$m_b^0 = \frac{1}{Y_u} \int_u^b \omega \, dx$$

Mass burning rate (unstretched)

Mass consumption rate

Flame stretch

- Classical definition: Fractional rate of change of area of flame surface element

$$K_A = \frac{1}{A}\frac{dA}{dt}$$

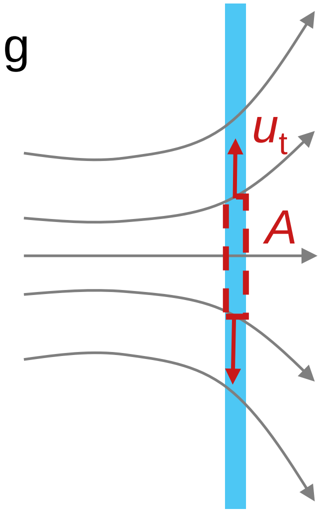
- Flow straining, flame curvature, flame motion

- De Goey & Ten Thije: Fractional rate of change of mass in flame volume element

$$K = \frac{1}{M}\frac{dM}{dt}$$

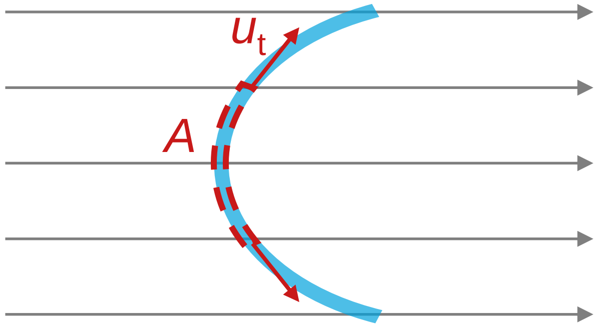
- Stretch rate defined in whole flame structure including preheat zone

Flow straining



$$K_A = \nabla_t \cdot u_t$$

Flame curvature



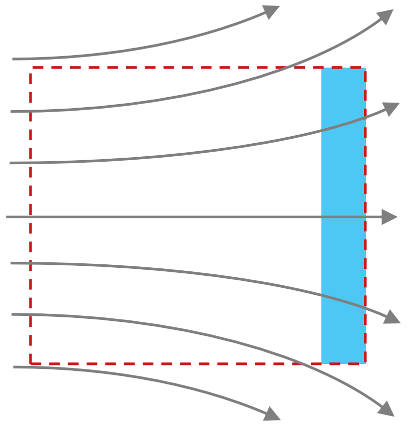
Mass burning rate of stretched flames

Flame stretch has an impact on the burning velocity

- Consider flat strained flame

$$\frac{d}{dx}(\rho u) = -\rho K$$

$$\frac{d}{dx}(\rho u Y) = \rho D \frac{d^2 Y}{dx^2} - \omega - \rho K Y$$



$$K = \nabla_t \cdot u_t = \frac{\partial v}{\partial y}$$

- Integral analysis

$$\left.\begin{aligned} m_b - m_u &= -\int \rho K \, dx \\ -m_u Y_u &= -\int \omega \, dx - \int \rho K Y \, dx \\ &= -m_b^0 Y_u - \int \rho K Y \, dx \end{aligned}\right\}$$

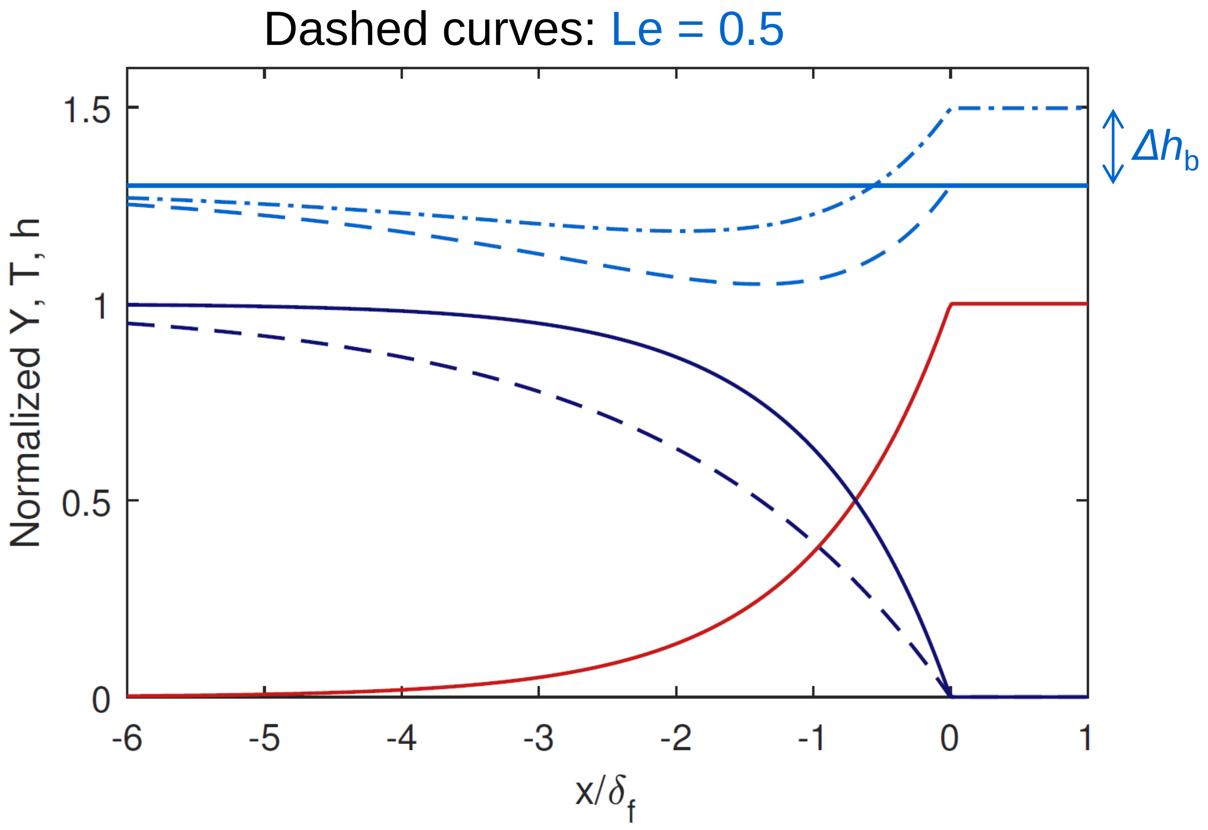
$$m_b = m_b^0 - \int \rho K (1 - Y/Y_u) dx$$

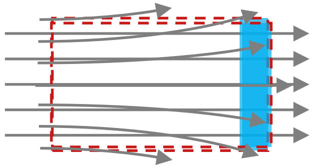
$$\frac{m_b}{m_b^0} = 1 - \frac{1}{m_b^0} \int \rho K \mathcal{Y} \, dx = 1 - \mathrm{Ka}$$

Karlovitz integral, Ka

Non-unity Lewis number effects

- Enthalpy profiles $h = q_c Y + c_p T$





Integrating enthalpy equation

$$m_b h_b - m_u h_u = -\int \rho K h \, dx$$

Combining with continuity equation

$$h_b - h_u = -\frac{1}{m_b} \int \rho K (h - h_u) \, dx$$

$$\boxed{\begin{aligned} \mathrm{Le} < 1 \text{ and } K > 0 &\Longrightarrow h_b > h_u = h_b^0 \\ &\Longrightarrow T_b > T_b^0 \\ &\Longrightarrow \omega > \omega^0 \end{aligned}}$$

Preferential diffusion effects

- In general, all species have different $\mathrm{Le}_i \neq 1$
- Results in changes in element mass fractions ΔZ_j at the burnt side
- Affects the equilibrium composition at the burnt side T_b, $Y_{i,\mathrm{b}}$
- And thus, the reaction rates ω and the mass consumption rate $m_\mathrm{b}^0 = m_\mathrm{b}^0(h_\mathrm{b}, Z_{j,\mathrm{b}})$
- Mass burning rate

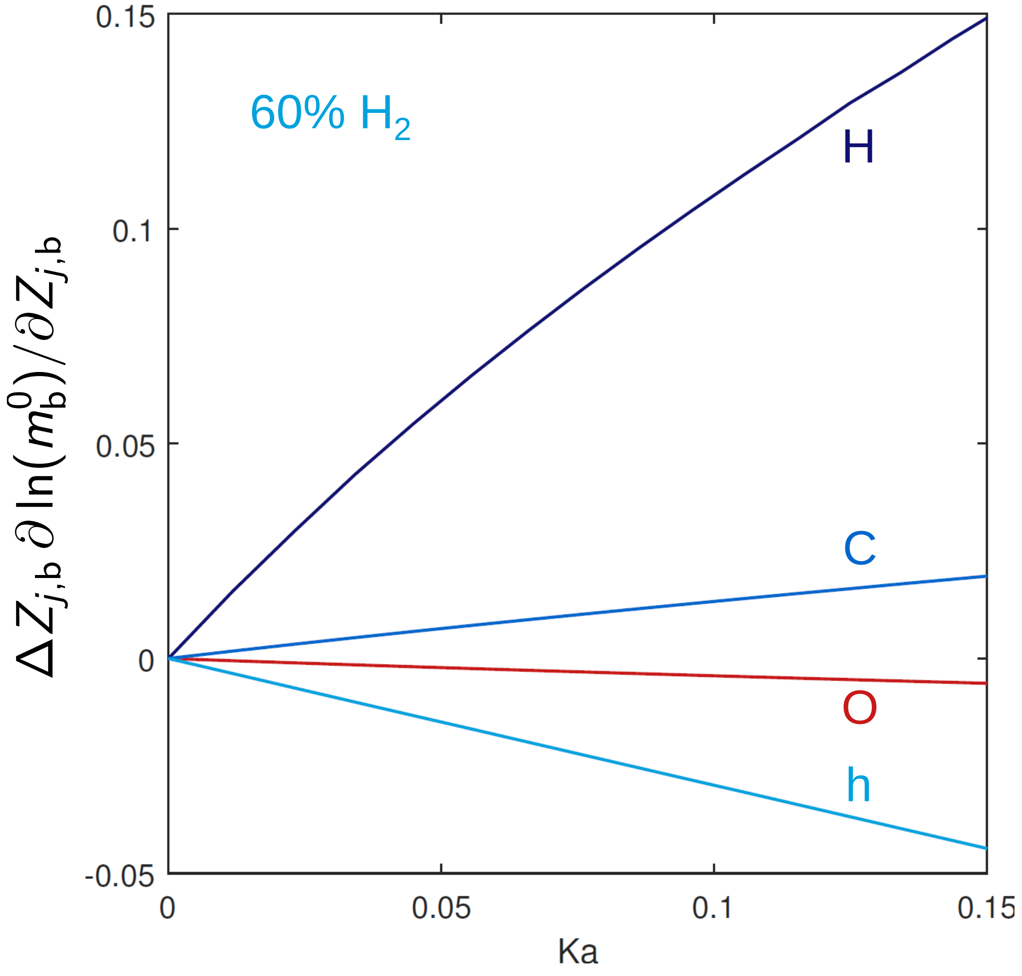
$$m_\mathrm{b} = m_\mathrm{b}^0(h_\mathrm{b}, Z_{j,\mathrm{b}})\,[1 - \mathrm{Ka}]$$

- Linearized for weak stretch, $\mathrm{Ka} \ll 1$

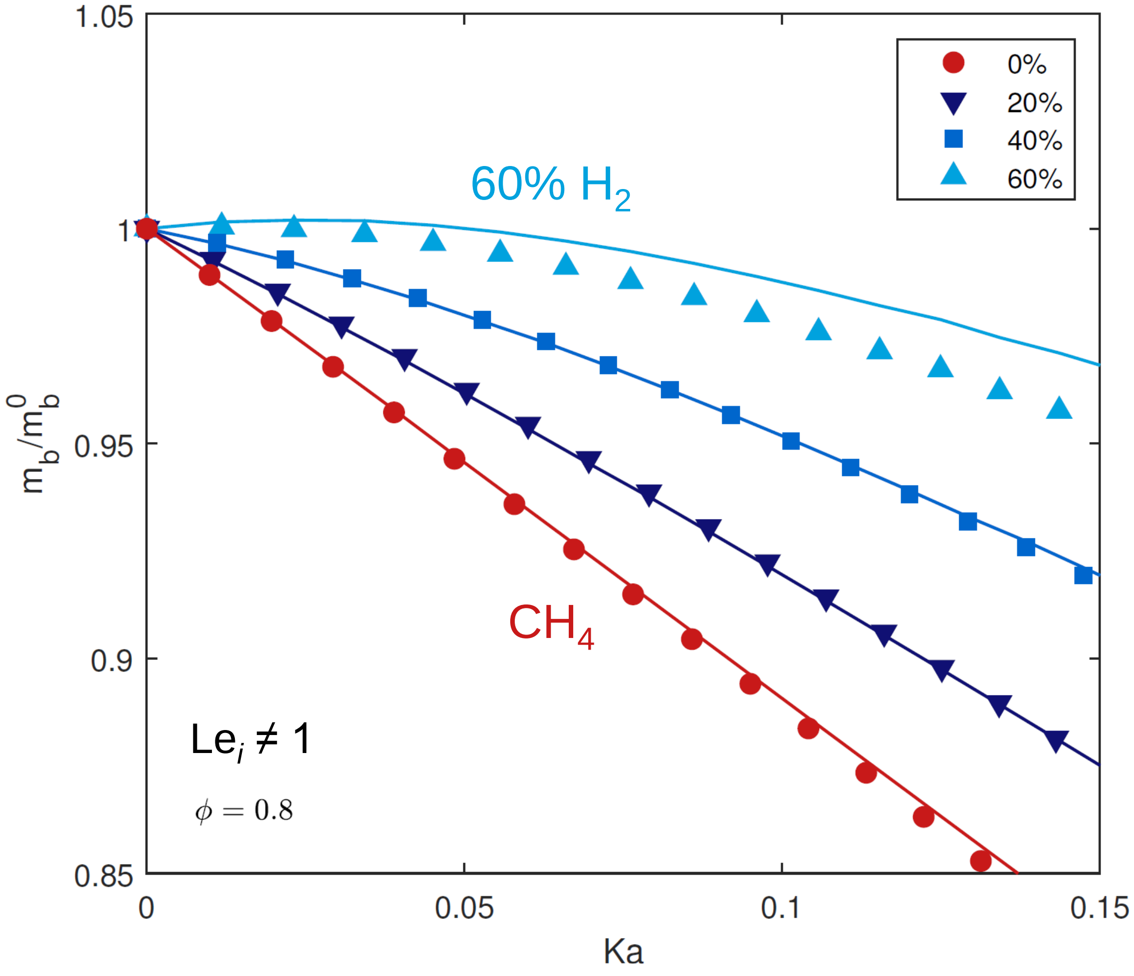
$$\frac{m_\mathrm{b}}{m_\mathrm{b}^0} = 1 - \underset{\substack{\uparrow \\ \text{Direct stretch effect}}}{\mathrm{Ka}} + \underbrace{\Delta h_\mathrm{b} \frac{\partial \ln(m_\mathrm{b}^0)}{\partial h_\mathrm{b}} + \Delta Z_{j,\mathrm{b}} \frac{\partial \ln(m_\mathrm{b}^0)}{\partial Z_{j,\mathrm{b}}}}_{\text{Indirect, preferential diffusion effects}} = 1 - \underset{\substack{\uparrow \\ \text{Markstein number Ma}}}{\mathrm{Ma}}\,\mathrm{Ka}$$

Comparison with simulations

- Stretched CH_4-air flames in counterflow

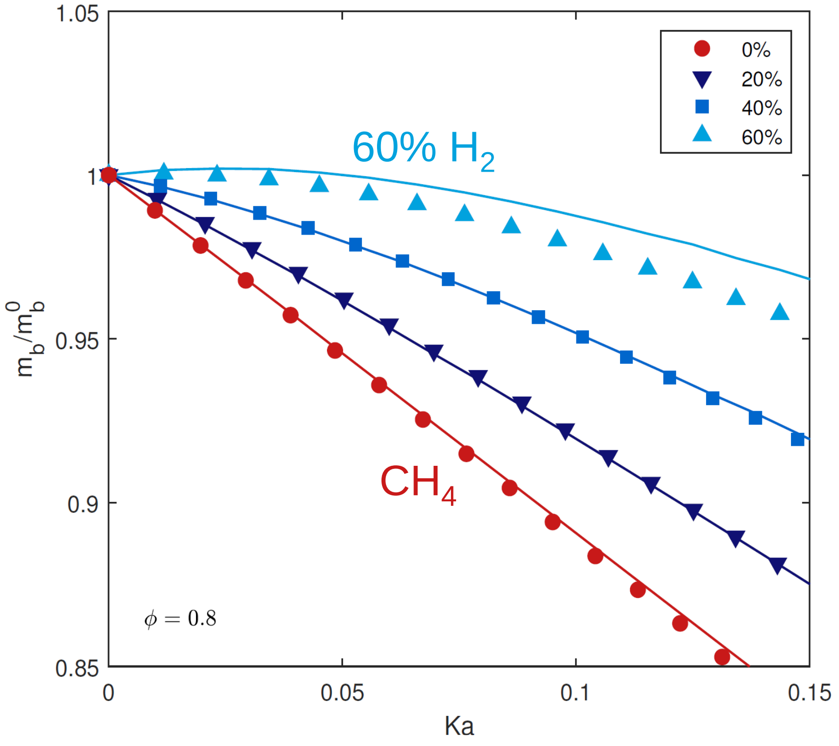


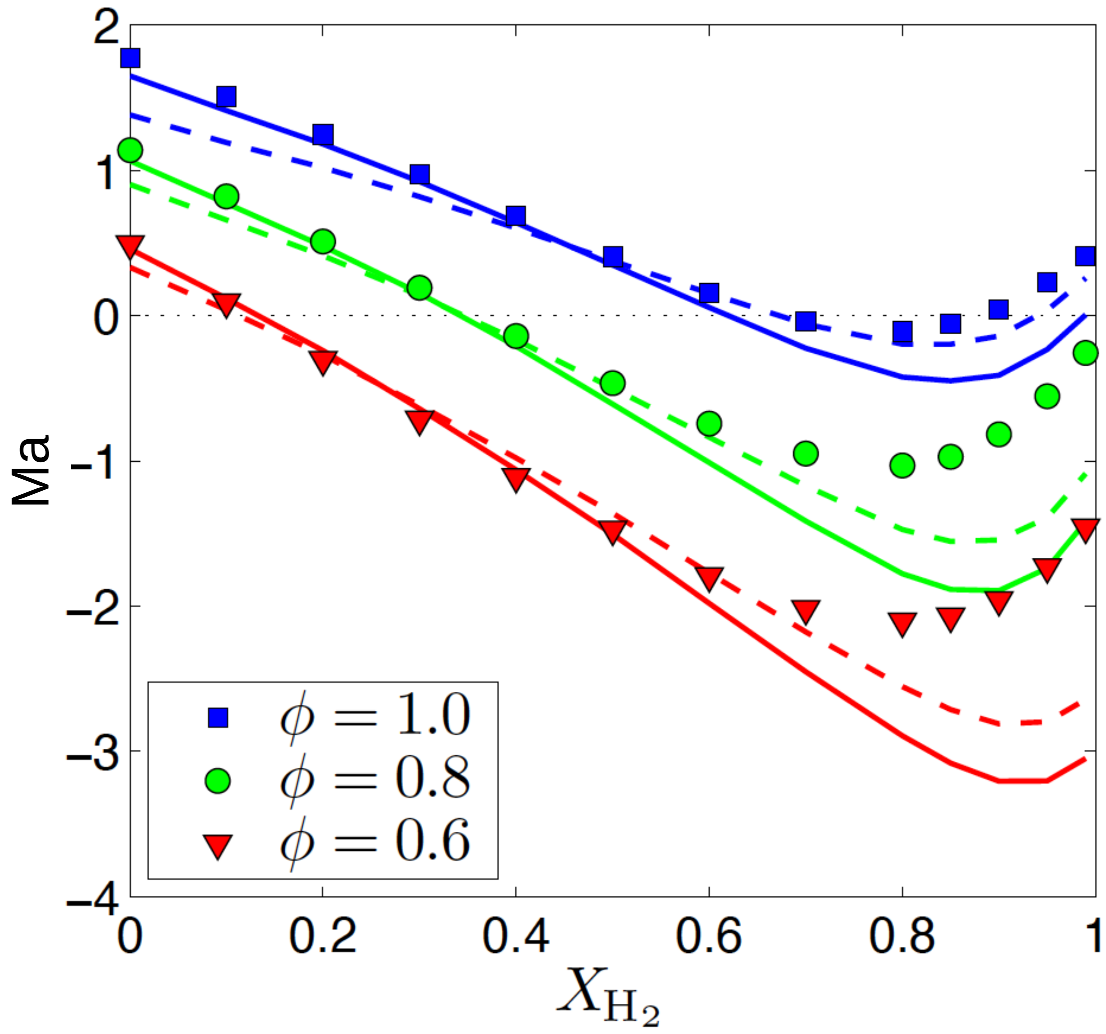
- Stretched CH_4-H_2-air flames



Markstein number

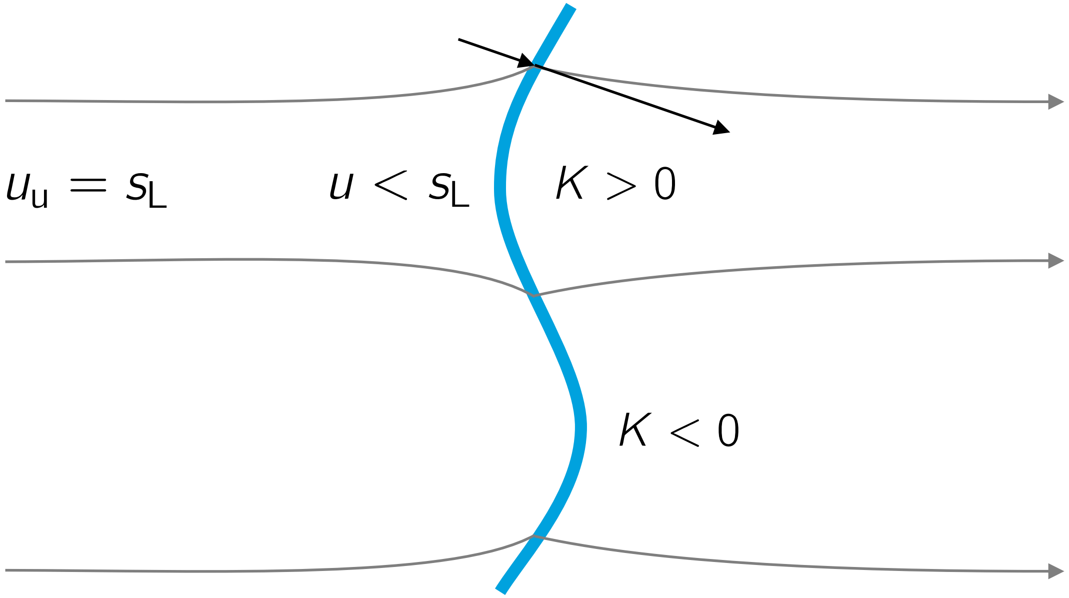
$$\frac{m_b}{m_b^0} = 1 - \mathrm{Ma}\,\mathrm{Ka}$$

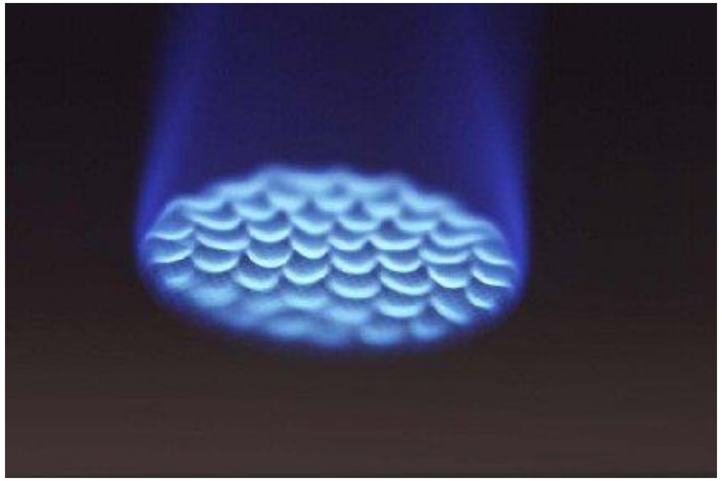


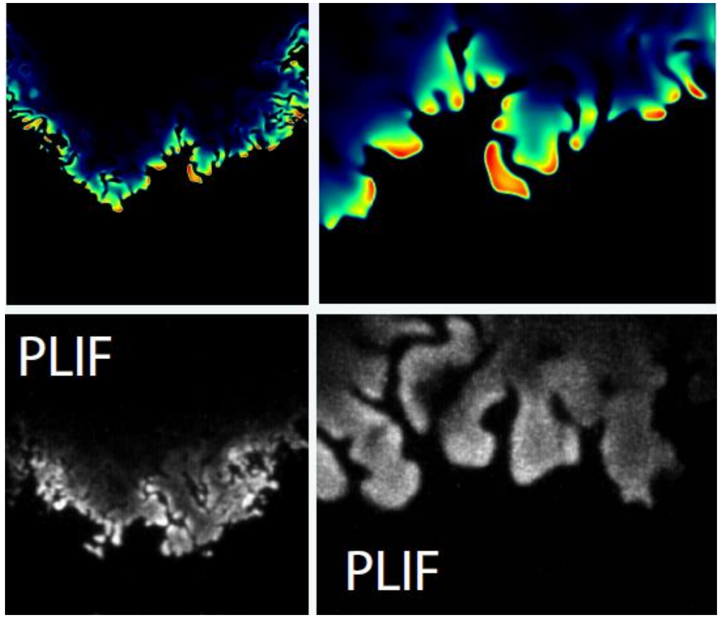


Cellular instabilities

- Density jump in flames causes hydrodynamic instability
- Direct flame stretch effect has stabilizing influence
  (Positive stretch decreases burning velocity)
- Preferential diffusion effects can counteract this (Ma < 1)
  (Positive stretch may even increase burning velocity)







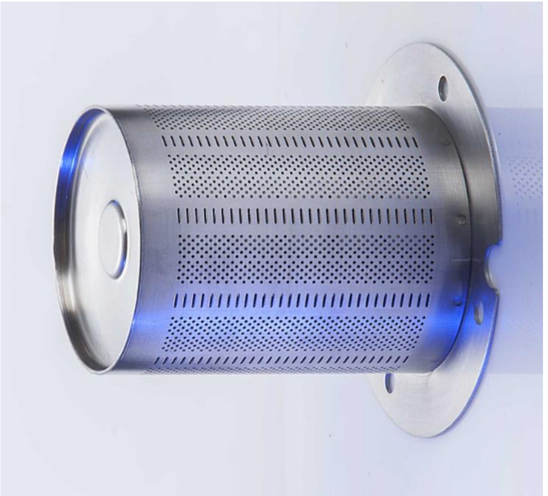
M. Day, Lawrence Berkeley Nat. Lab

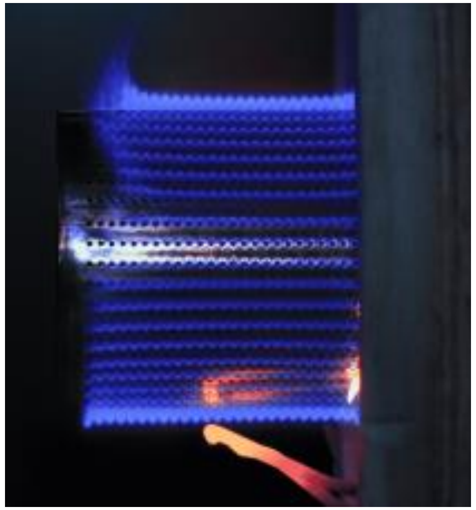
Flame stabilization

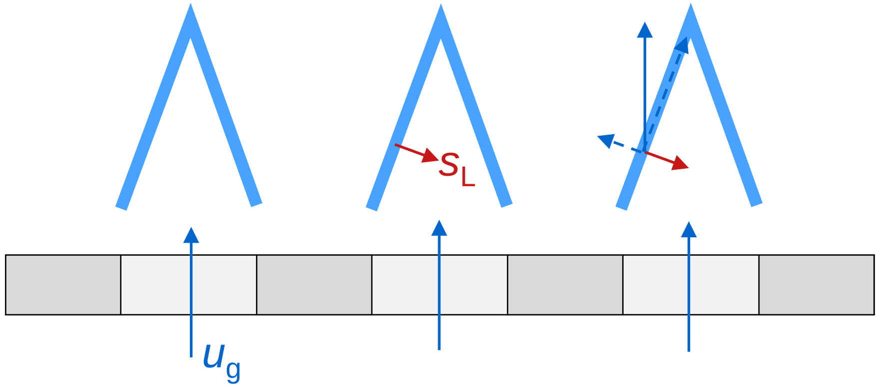
Impact of preferential diffusion effects

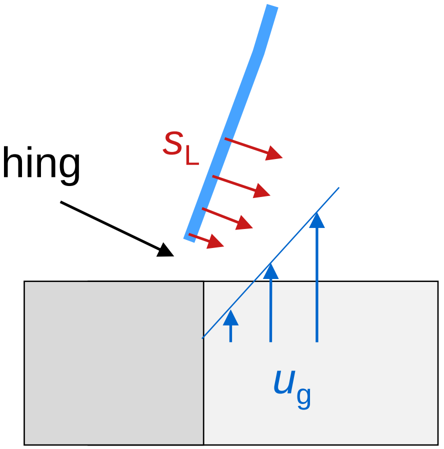
Flame stabilization on perforated plate burner

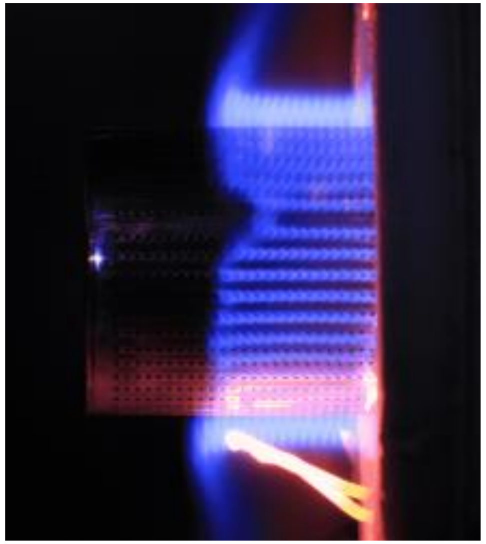
- Used in domestic heating systems
- Balance of flame speed s_L
  and gas mixture velocity u_g
- Flame flashback/blow off








Local quenching



Lewis & von Elbe (1987) Combustion, Flames, and Explosions of Gases, Academic

Experiment

- Lean CH_4-H_2-air flames on multi-slot burner (Y. Shoshin)
- Fuel mixtures with same flame speed (s_L = 10 cm/s)

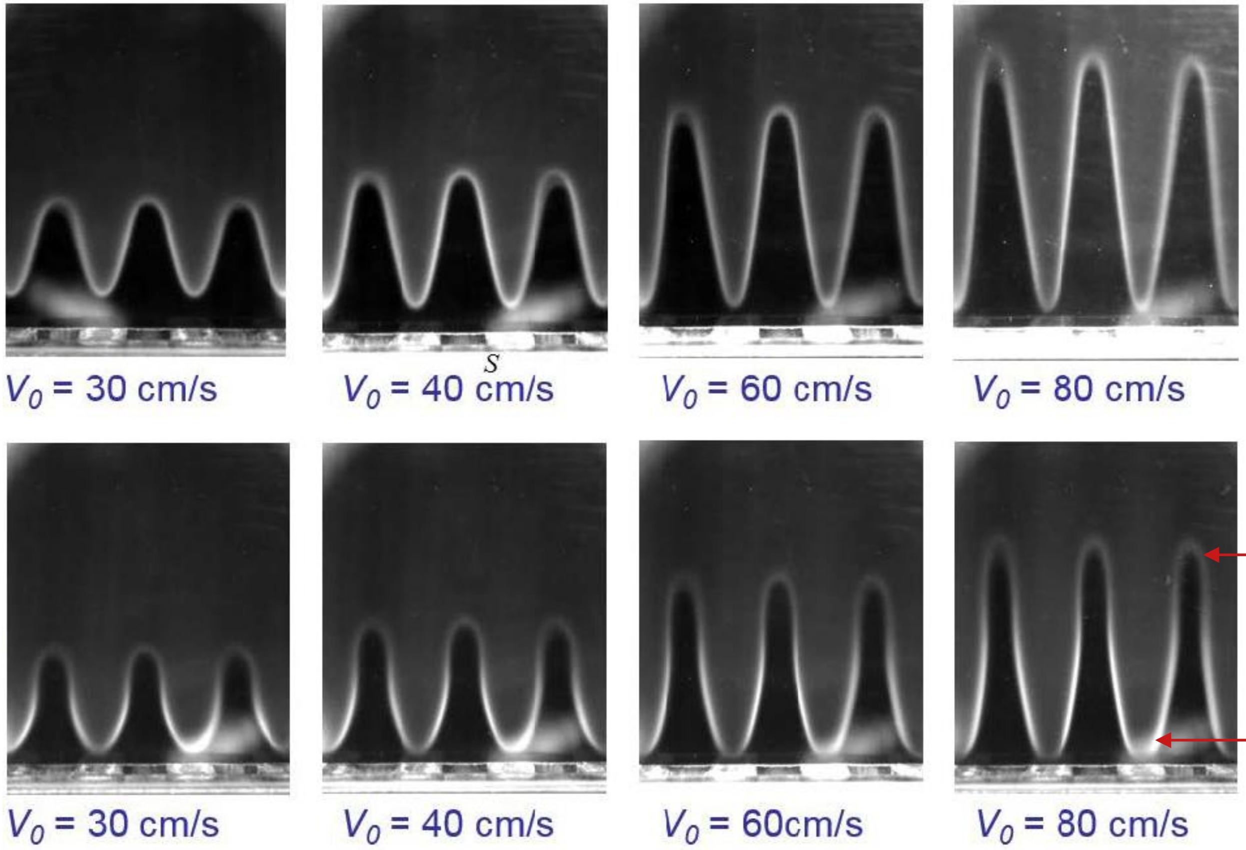
- H_2 has much higher diffusivity than methane (Le_{H2} = 0.3 vs Le_{CH4} = 1)
- Together with flame stretch and curvature this causes local enrichment: $\varphi\uparrow$
- Resulting in local higher burning rate: $s_L\uparrow$
- Affects stabilization a lot

CH_4

$\varphi = 0.58$

0.6 CH_4 + 0.4 H_2

$\varphi = 0.52$



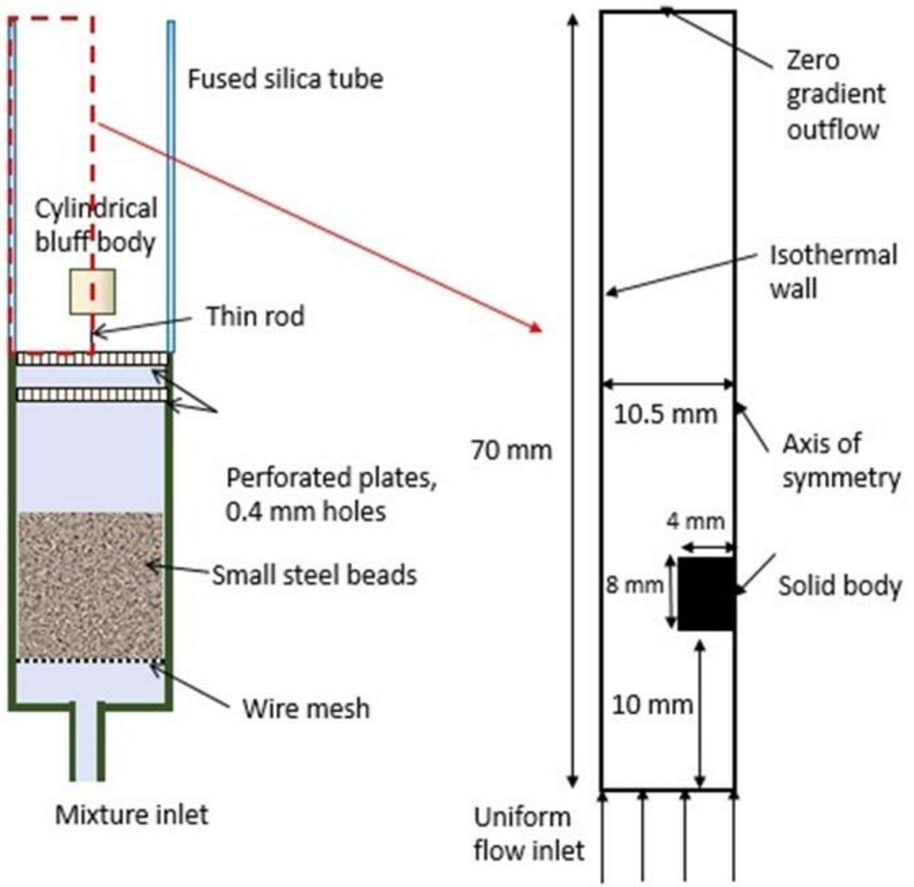
Weaker burning at flame tip ($K < 0$)

Intensified burning at flame base ($K > 0$)

Bluff body stabilized flames

- Experimental and numerical study of flames stabilized on cylindrical bluff body
- At fixed velocity u_g, equivalence ratio is decreased until blow-off occurs
- Anomalous blow-off limit behavior observed for mixtures with H_2

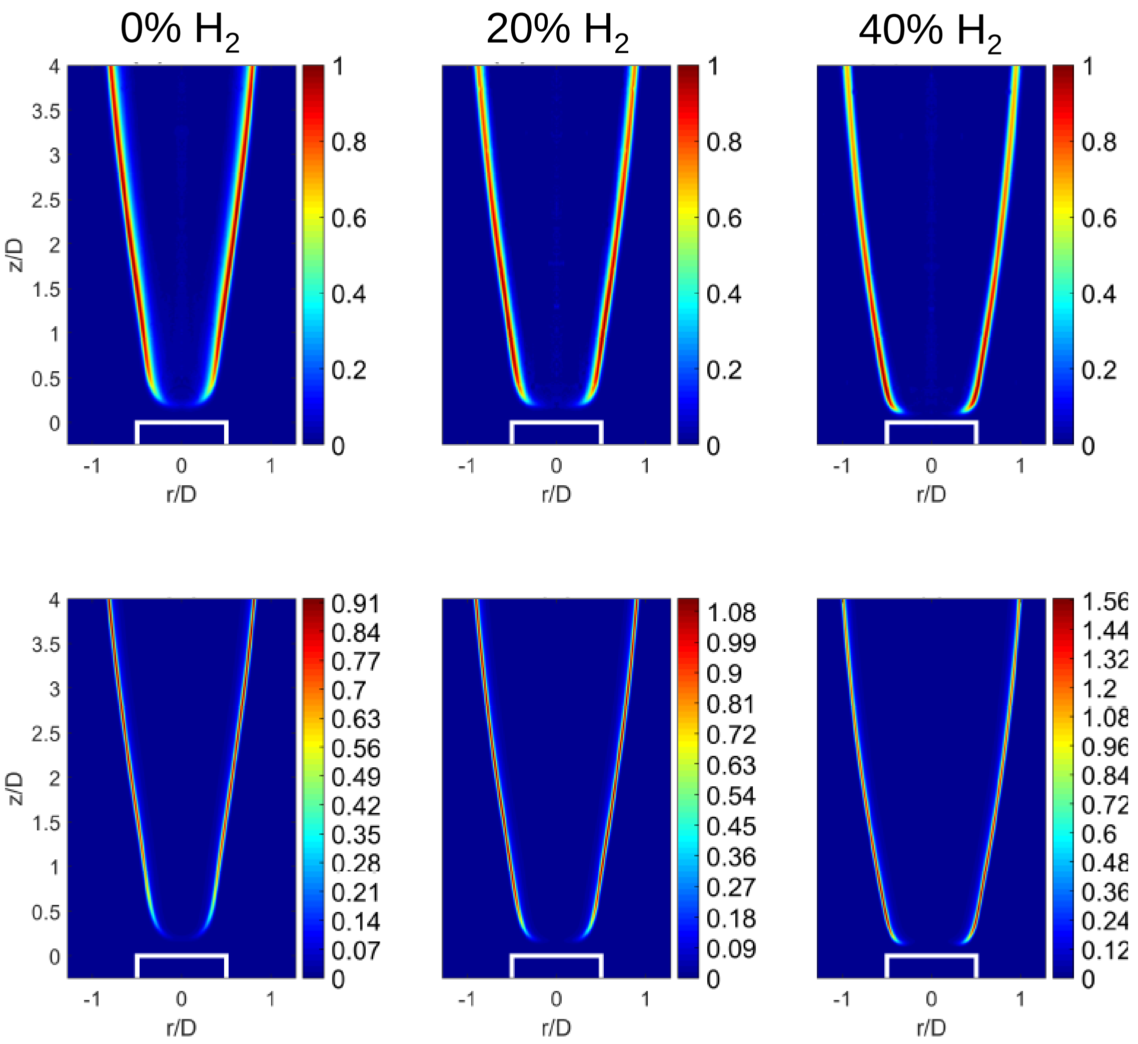
- Experiments by Y. Shoshin
- Simulations by F. Vance



Flame stabilisation: experiment vs simulation

- Three CH_4-H_2 fuel mixtures with equal flame speed, s_L = 10 cm/s
- Inlet velocity 1 m/s
- Comparison of Abel inverted CH* chemiluminescence (top) and computed heat release rate (bottom)

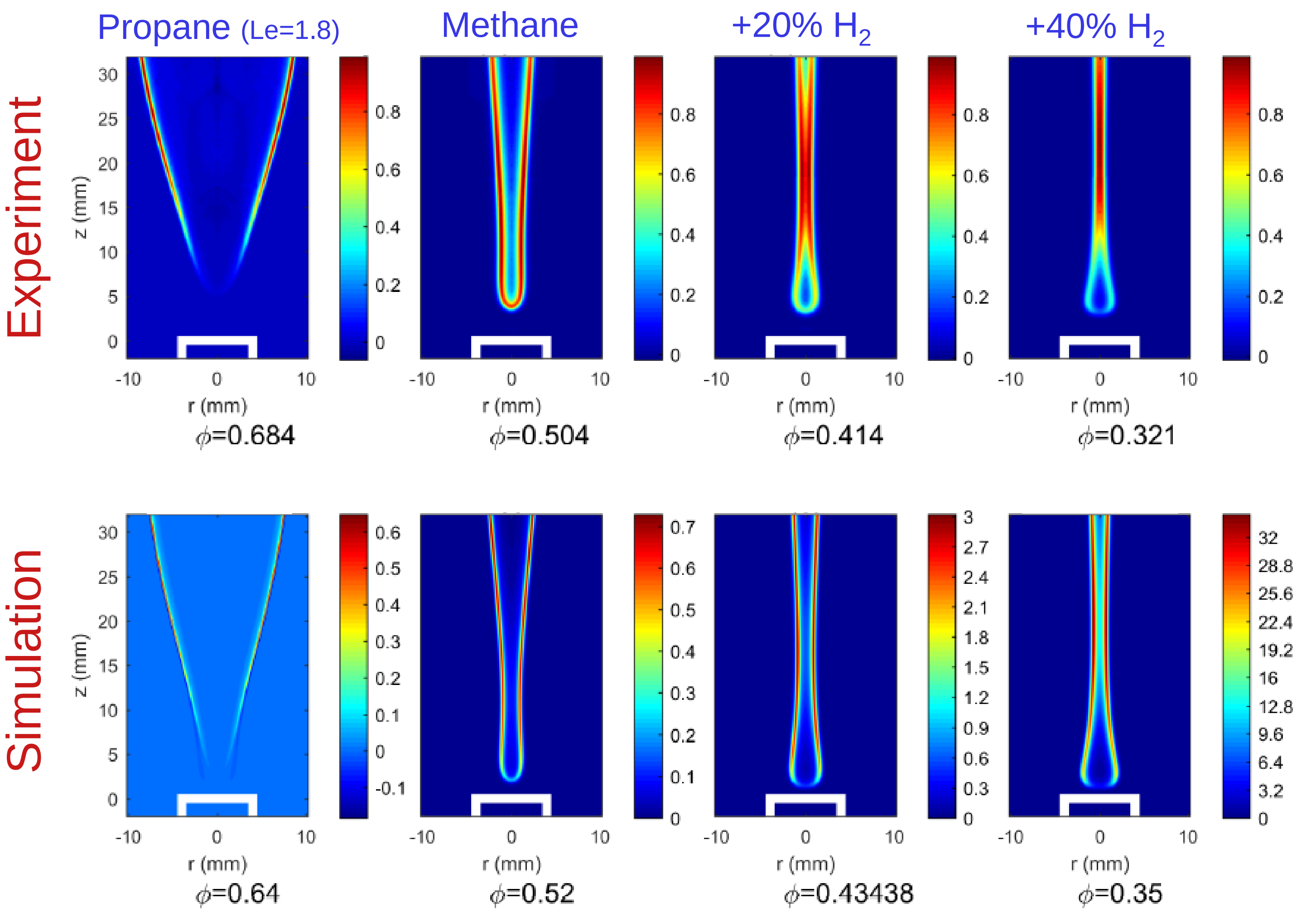
- Numerical results allow detailed quantitative analysis of stretch, heat loss and preferential diffusion effects



Vance et al. (2021) Combust. Flame, available online

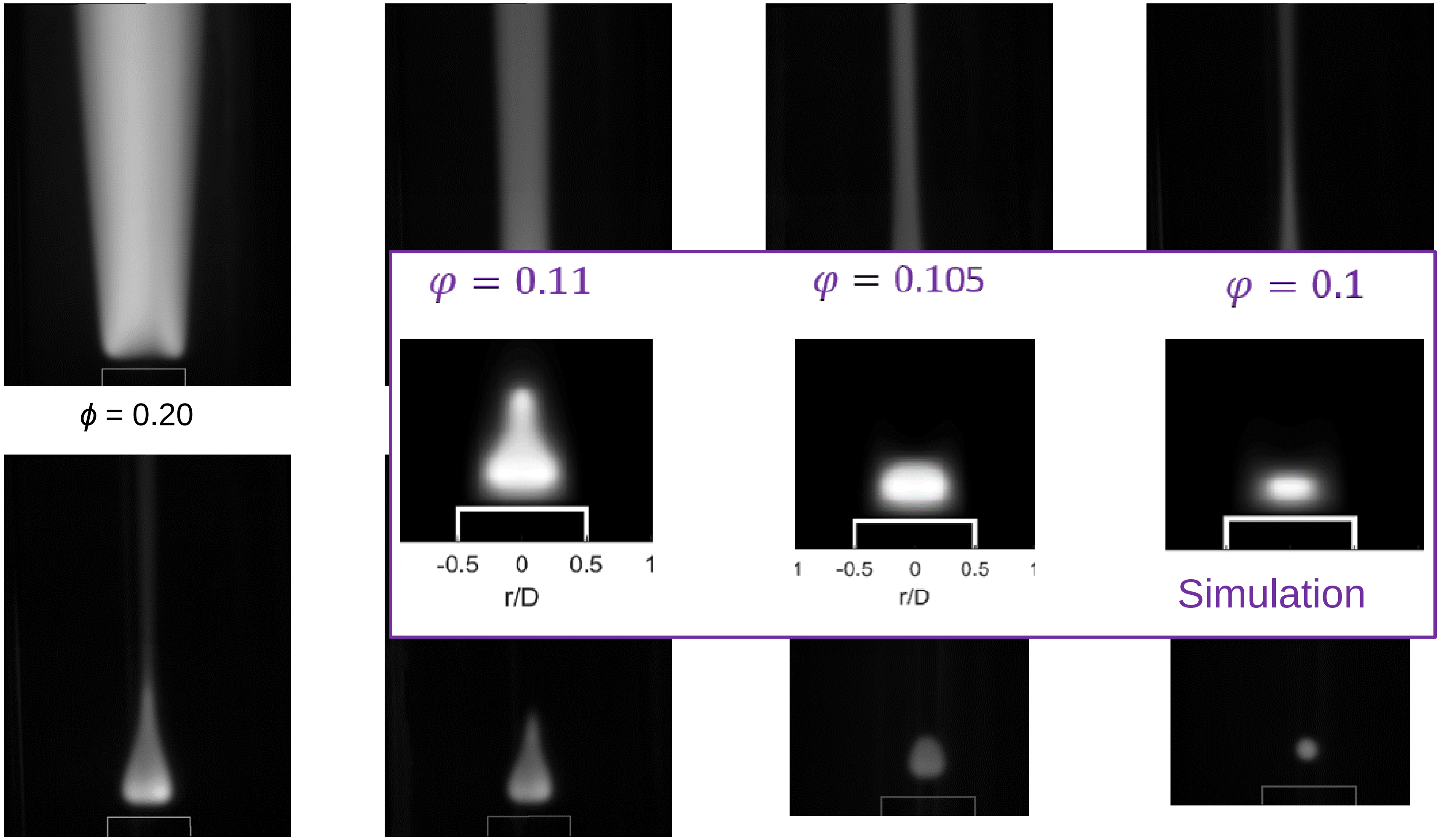
Effect of Lewis number on lean limit flame

- Lean limit flames for 4 fuels with different Lewis number
- Inlet velocity 1 m/s
- Strong Le effects enhance stability of H_2 enriched flames: Neck formation
- Different blow-off mechanisms observed



Vance et al. (2019) Proc. Combust. Inst., 37:1663-1672

H_2-air flame shapes approaching lean limit (V = 1 m/s)



ϕ = 0.20

ϕ = 0.103

ϕ = 0.1

ϕ = 0.094

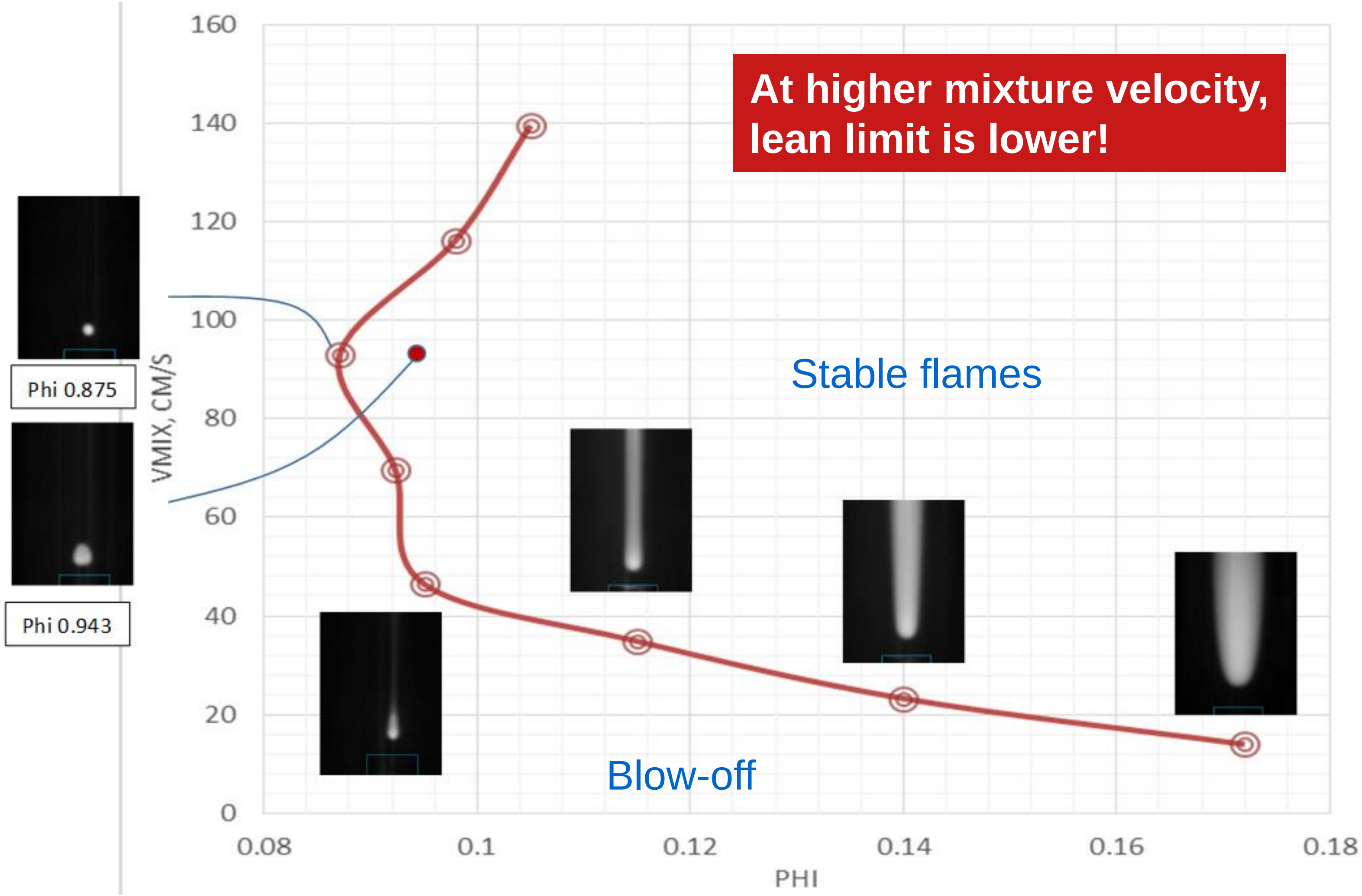
ϕ = 0.088

- Pencil-like flame
- Quenches
- Residual flame
- Flame ball!
  (PhD Zhen Zhou)

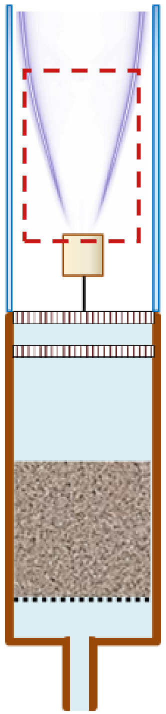
- Combustion at extremely low φ
- Simulations show same behavior

Vance et al. (2021) Energies, 14, 1977

H_2-air lean limit flames



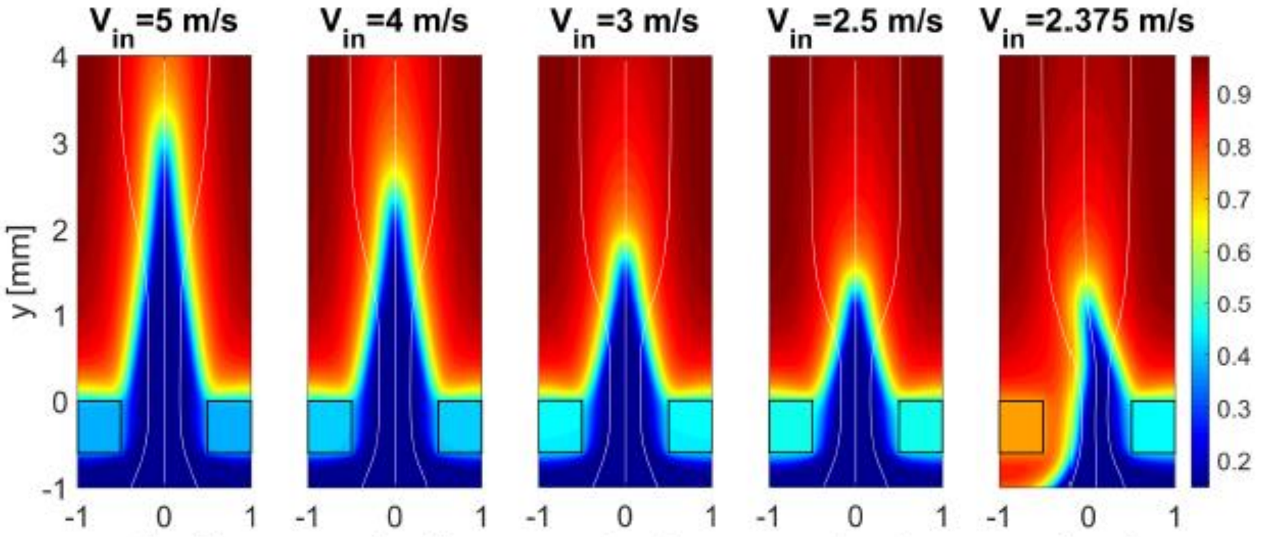
At higher mixture velocity, lean limit is lower!



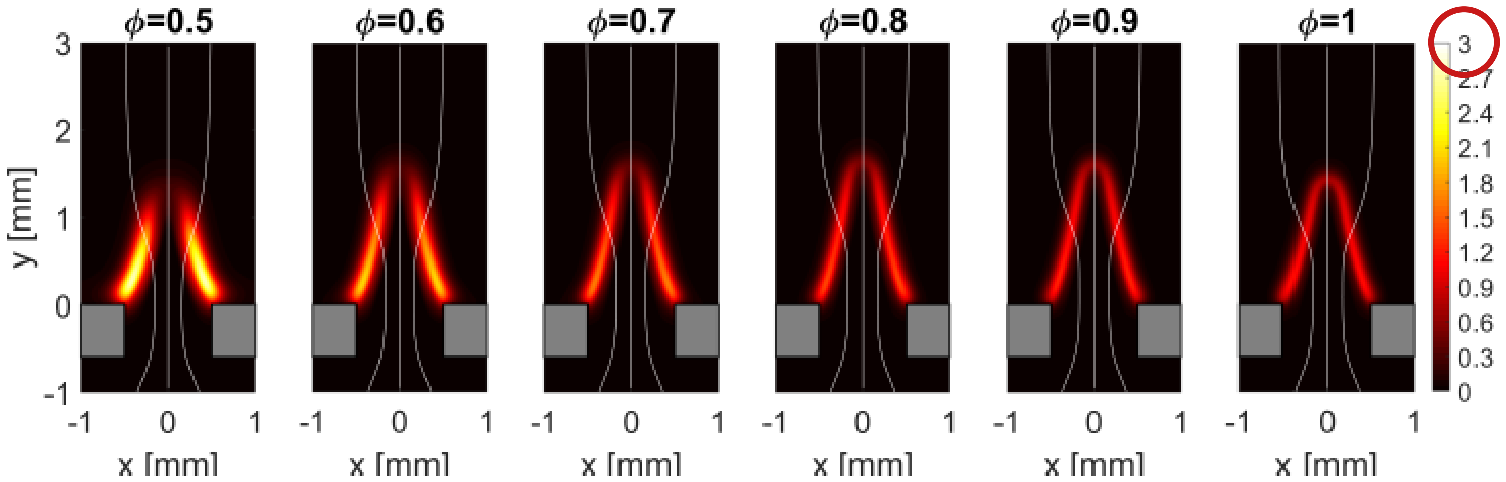
Hydrogen flame flashback

- Simulations of H_2-air flame on slot burner
- Decrease inlet velocity V_{in} at constant φ until flame flashes back
- Enhanced burning rate due to preferential diffusion effects, leads to early flashback

Normalized temperature
$\varphi = 0.7$



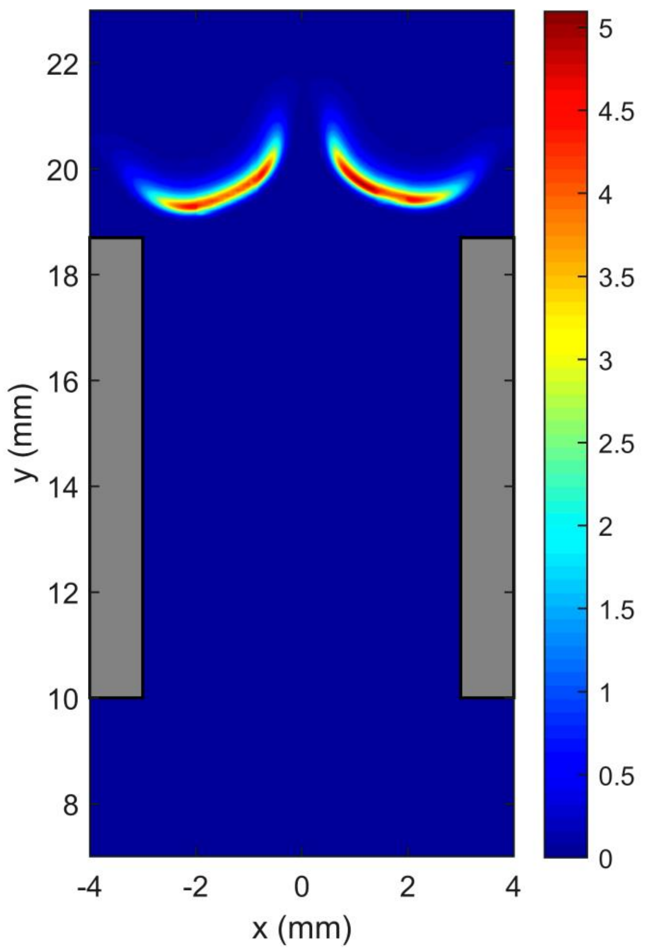
Normalized H_2 consumption rate of limit flames at different φ



Vance et al. (2021) Combust. Flame, submitted

Complex dynamics

- Unsteady simulation
- Flashback when velocity is lowered

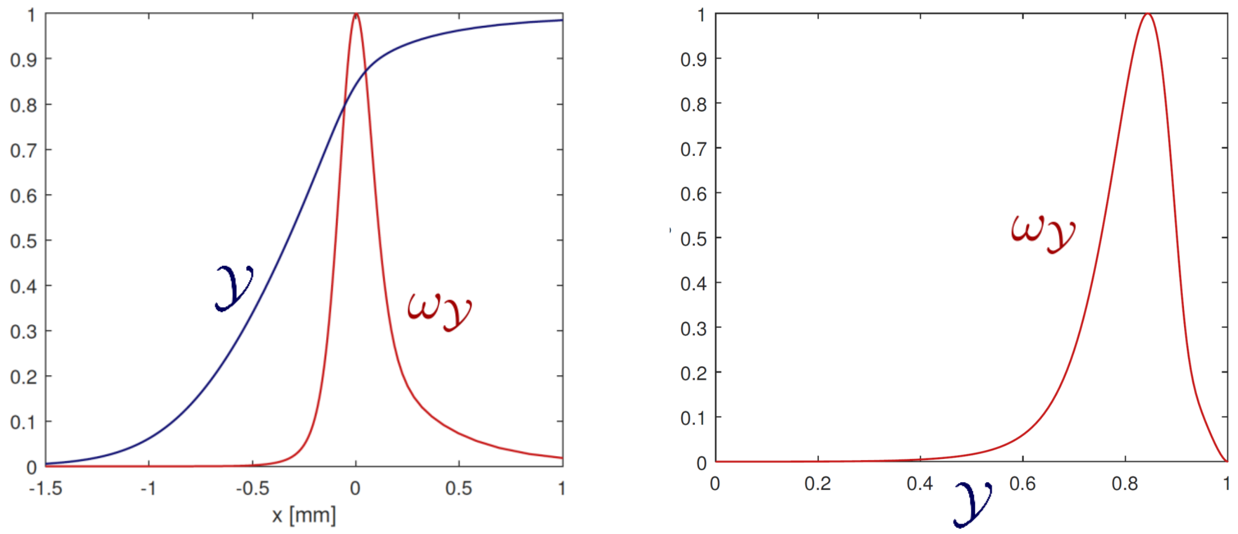


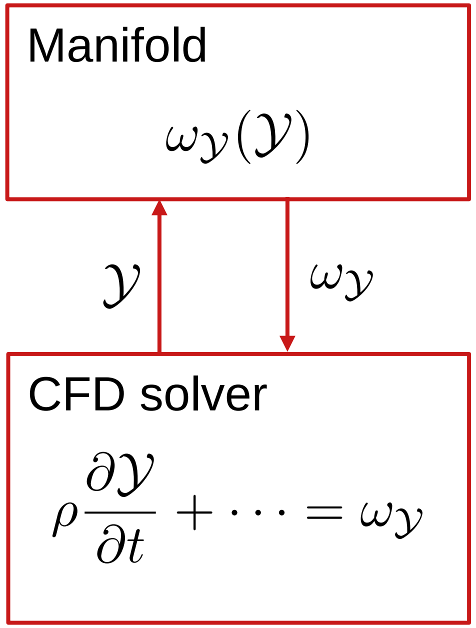
Flamelet-Generated Manifolds for H_2 flames

Flamelet-Generated Manifold (FGM) method

- Reduced order modelling of chemistry
- Combination of low-dimensional manifold and flamelet approach
- Solutions of 1D flamelet equations are used to construct a manifold
- Chemical composition (Y_i, T) is parameterized by small number of control variables y_j
- Simplest form, 1D FGM, where y_1 is reaction progress variable

$$Y_i(x) \longrightarrow Y_i(y_1) \qquad y_1 = \mathcal{Y}$$





Manifold

$$\omega_\mathcal{Y}(\mathcal{Y})$$

$\mathcal{Y}$

$\omega_\mathcal{Y}$

CFD solver

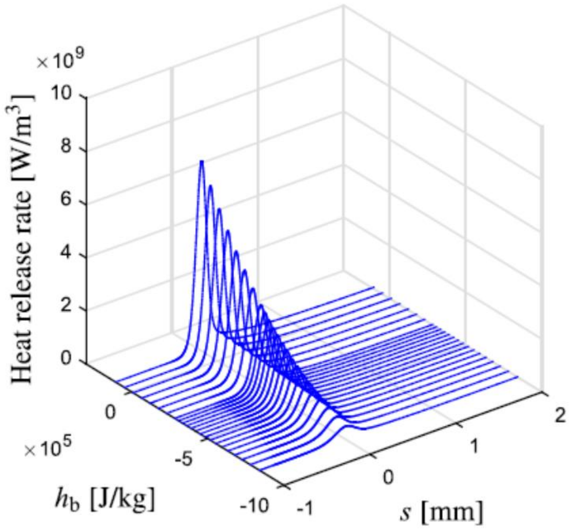
$$\rho\frac{\partial \mathcal{Y}}{\partial t} + \cdots = \omega_\mathcal{Y}$$

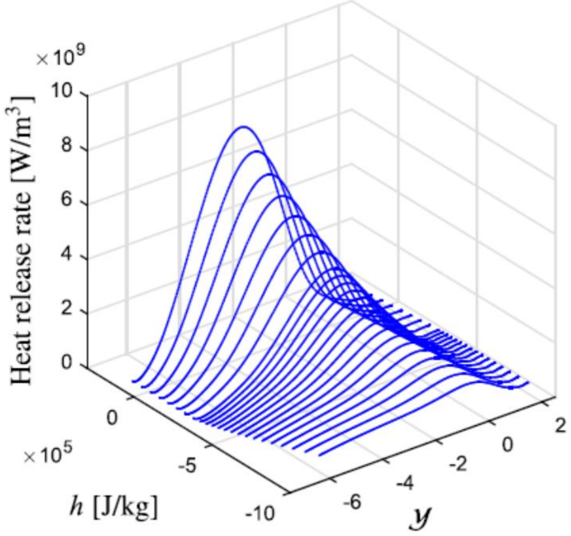
Van Oijen & De Goey (2000) Combust. Sci. Technol. 161:113-138
Van Oijen et al. (2016) Prog. Energy Combust. Sci. 57:30-74

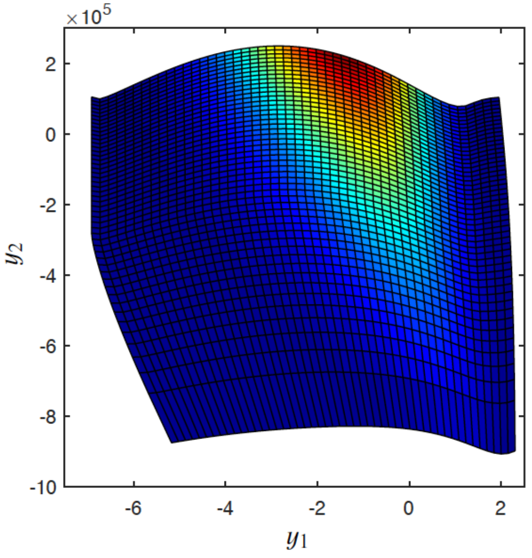
Multi-dimensional FGM

- In 1D FGM, enthalpy and element mass fractions are fixed, but in most applications, this is not the case due to, e.g., heat loss, mixture stratification, dilution, etc.
- Additional manifold dimensions (parameters) are needed to account for the effect of these changes on chemistry
- For non-adiabatic effects, enthalpy is added as manifold coordinate
- Series of flamelet solutions for different enthalpy

$$Y_i(x, h_\mathrm{b}) \longrightarrow Y_i(y_1, y_2) \qquad y_1 = \mathcal{Y}, y_2 = h$$



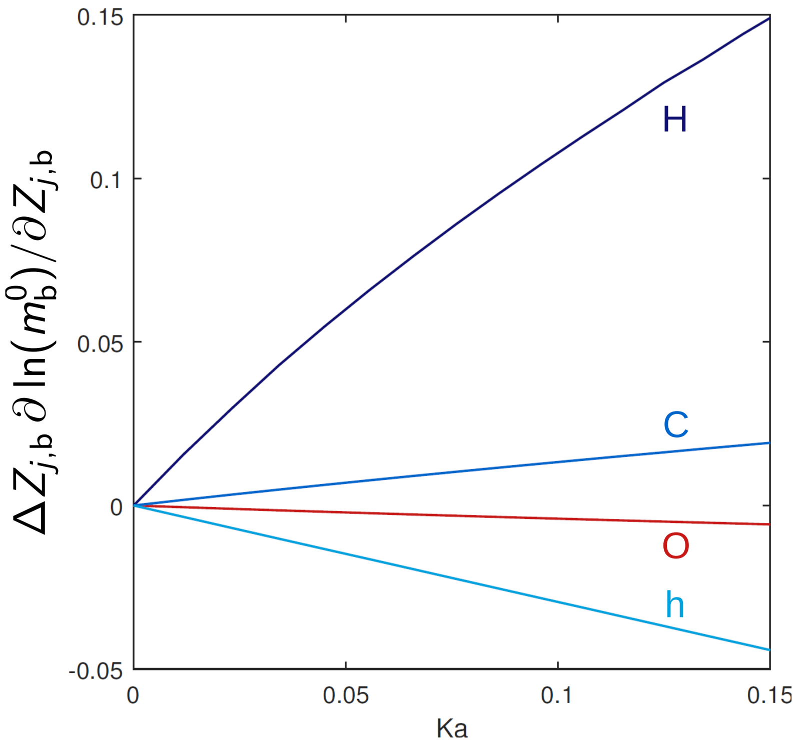




FGM for H_2 flames

- Strong preferential diffusion effects in H_2 flames cause changes in Z_j and h
- In principle all Z_j's and h should be added to the manifold as additional independent parameters

- For weak stretch, ΔZ_j's and Δh are not independent but couple: One additional dimension is sufficient



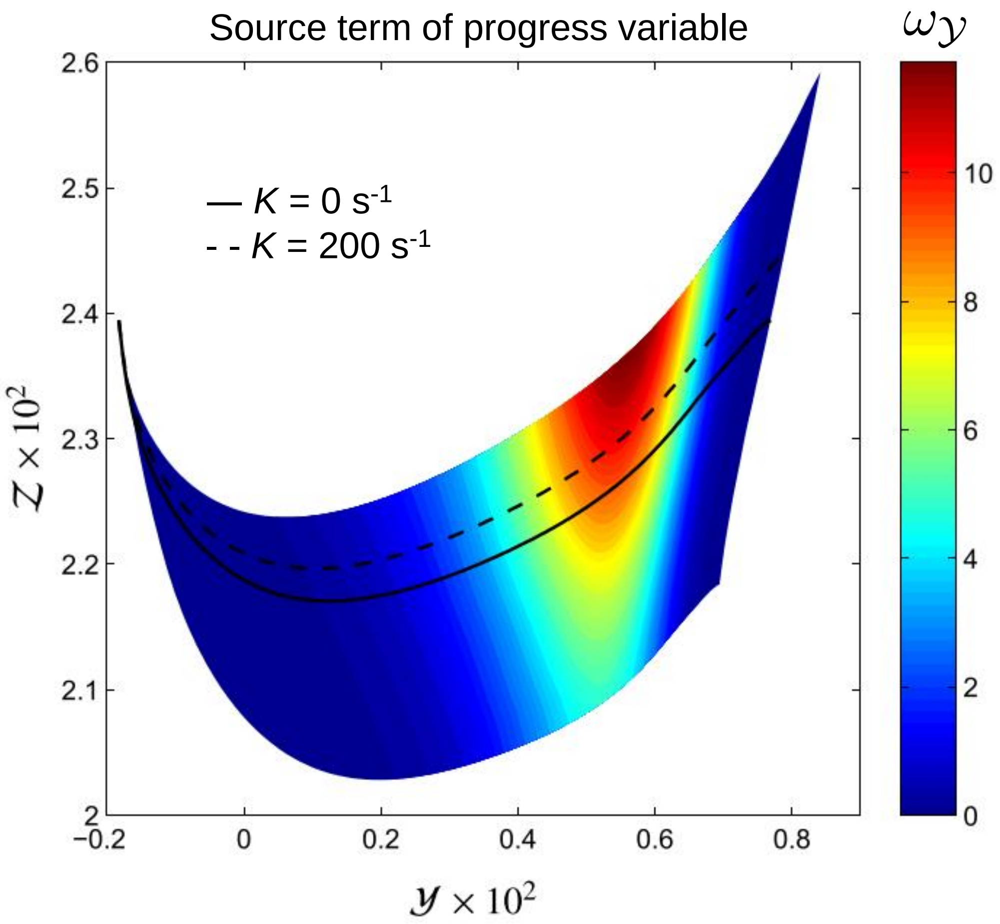
Preferential diffusion effects in FGM

- In practice: Include a stretch term in flamelet equations and solve for a range of stretch rates *K*

$$Y_i(x, K) \longrightarrow Y_i(y_1, y_2)$$

- This results in a 2D FGM parameterized by two control variables
  - Reaction progress variable
  - Element mass fraction

$$y_1 = \mathcal{Y}$$
$$y_2 = \mathcal{Z} = \tfrac{1}{2} Z_\mathrm{C} + Z_\mathrm{H}$$



Preferential diffusion effects in FGM

- Preferential diffusion terms in equations for control variables need to be retained
- Linear combinations of species mass fractions $y = \sum \alpha_i Y_i$

$$\frac{\partial \rho y}{\partial t} + \nabla \cdot (\rho \boldsymbol{u} y) - \nabla \cdot \left(\frac{\lambda}{c_p} \nabla y \right) = \nabla \cdot \underbrace{\left[\frac{\lambda}{c_p} \sum \alpha_i \left(\frac{1}{\mathrm{Le}_i} - 1 \right) \nabla Y_i \right]}_{} + \omega_y$$

- Assuming $\nabla Y_i = c_i \nabla y$ (Local 1D FGM)

$$\frac{\partial \rho y}{\partial t} + \nabla \cdot (\rho \boldsymbol{u} y) - \nabla \cdot \left(\frac{\lambda}{c_p} \nabla y \right) = \nabla \cdot (\mathcal{D} \nabla y) + \omega_y$$

- New approach assuming constant Lewis numbers

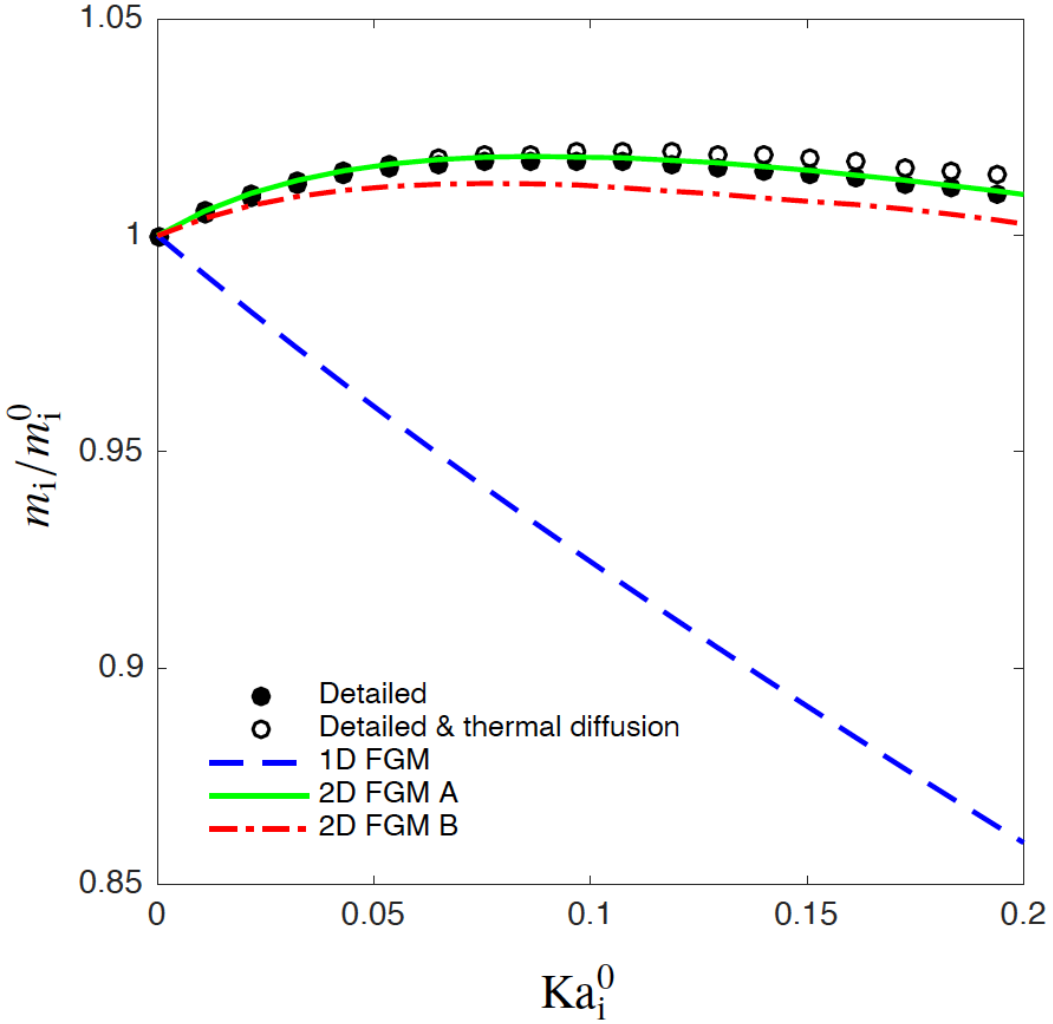
$$\frac{\partial \rho y}{\partial t} + \nabla \cdot (\rho \boldsymbol{u} y) - \nabla \cdot \left(\frac{\lambda}{c_p} \nabla y \right) = \nabla \cdot \left(\frac{\lambda}{c_p} \nabla \beta \right) + \omega_y \qquad \beta = \sum \alpha_i \left(\frac{1}{\mathrm{Le}_i} - 1 \right) Y_i$$

Significant improvement for H_2 flames

Mukundakumar et al. (2021) Combust. Theory Model, online available

Validation of FGM against detailed chemistry

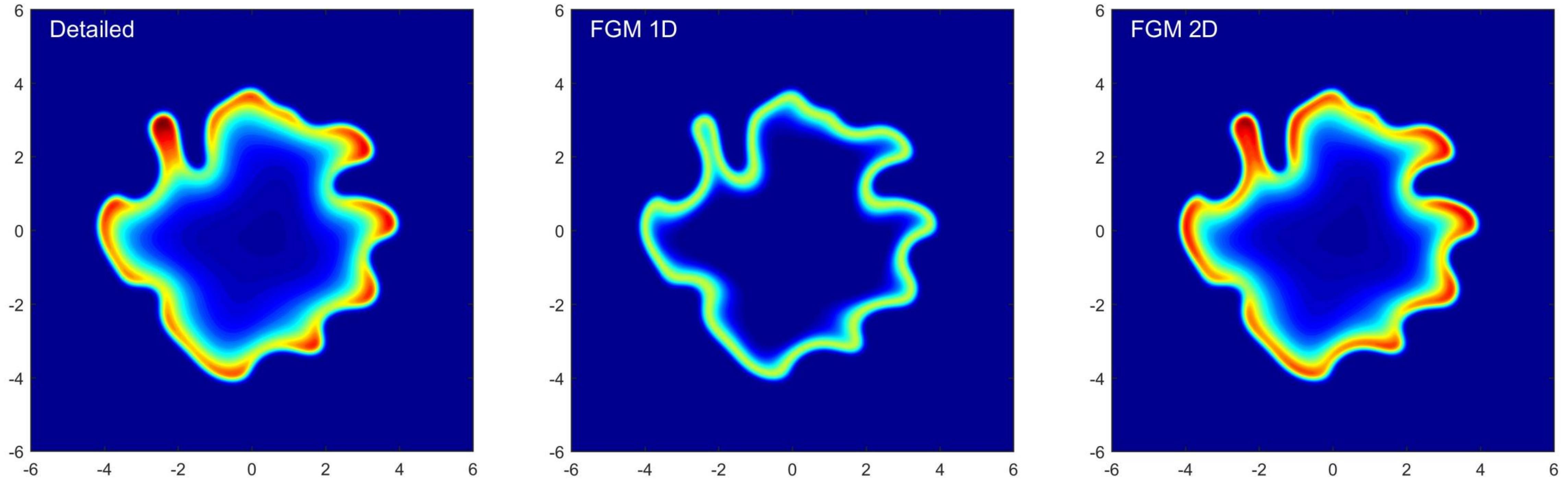
- 1D strained flames, CH_4-H_2-air (40% H_2, $\varphi = 0.7$)



- 1D FGM only account for direct stretch effect
- 2D FGMs capture preferential diffusion effects
- Not very sensitive to how the Z_j, h changes are included in the flamelets
  - 2D FGM A: stretched flat flamelets
  - 2D FGM B: stretched curved flamelets

Validation of FGM against detailed chemistry

- Expanding flame kernel in turbulent flow (CH$_4$-H$_2$-air, 40% H$_2$, φ = 0.7)
- 2D direct numerical simulation

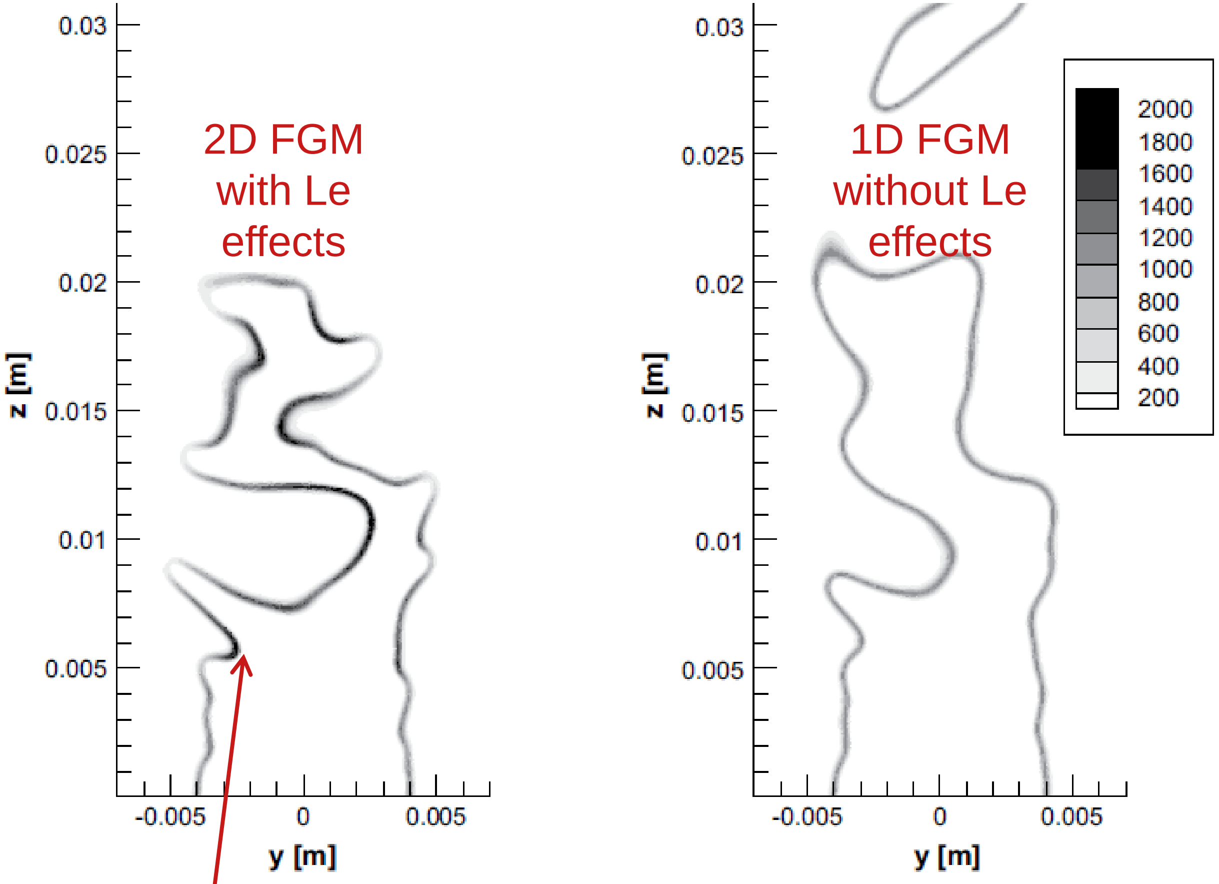


Mass fraction of H radical at t = 0.36 ms

Turbulent flame

- Lean premixed turbulent flame on slot burner
  CH_4-H_2-air, 40% H_2, $\varphi = 0.7$
- Direct numerical simulation

Snapshots of chemical source term

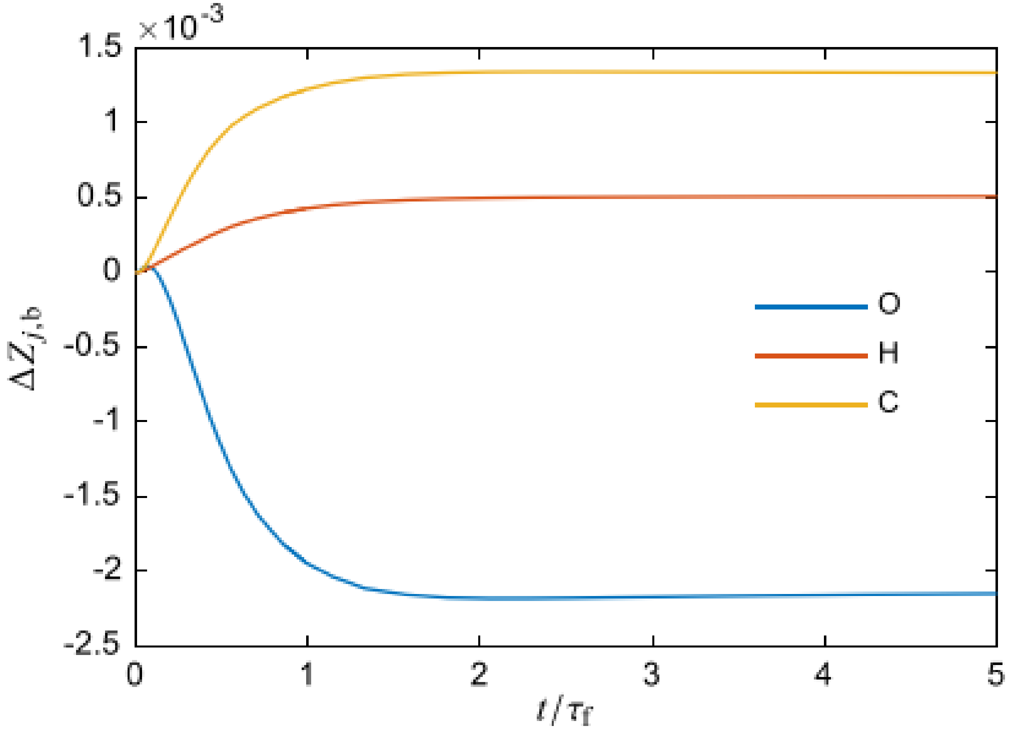


- More intense burning in convex regions
- Flame surface area/volume increases due to instabilities
- Turbulent flame speed increases by 30%!

Vreman et al. (2009) Int. J. Hydrogen Energy 34:2778-2788

Unsteady phenomena

- Response to sudden jump in stretch rate



- Time delay of $\tau_f = \delta_f / s_L$: High frequencies will be dampened

- Preferential diffusion effects are dampened in flames with fast fluctuating stretch rates
- Modelling challenge in unresolved simulations (LES/RANS)!
- DNS of highly turbulent flames required to gain insight and to develop models

De Swart et al. (2010) Flow Turbul. Combust. 85:473-511

Other modeling challenges

- Reactions occur at rather low temperature

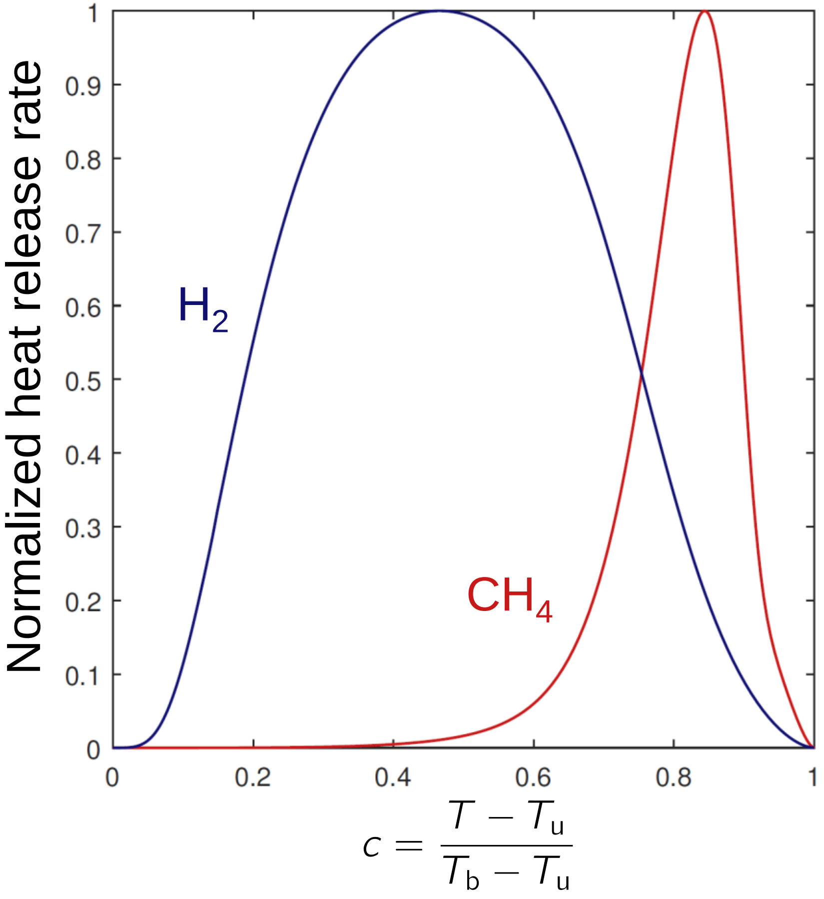
$$\begin{aligned} H_2 + O &\longrightarrow OH + H \\ H_2 + OH &\longrightarrow H_2O + H \end{aligned} \quad \text{H production, 1400 K}$$

$$O_2 + H + M \longrightarrow HO_2 + M \quad \text{H consumption, close to } T_u$$

- Peak heat release rate at T < 1000 K
- Reaction layer of H_2 flame is not thin!

FGM challenge

- Reaction rates in H-consumption layer at the leading edge are sensitive to T_b (near the H-production layer)
- Non-local dependency: Source term depends on condition downstream
- Overprediction of source term when burnt side is cooled → Too high flame speed, flashback



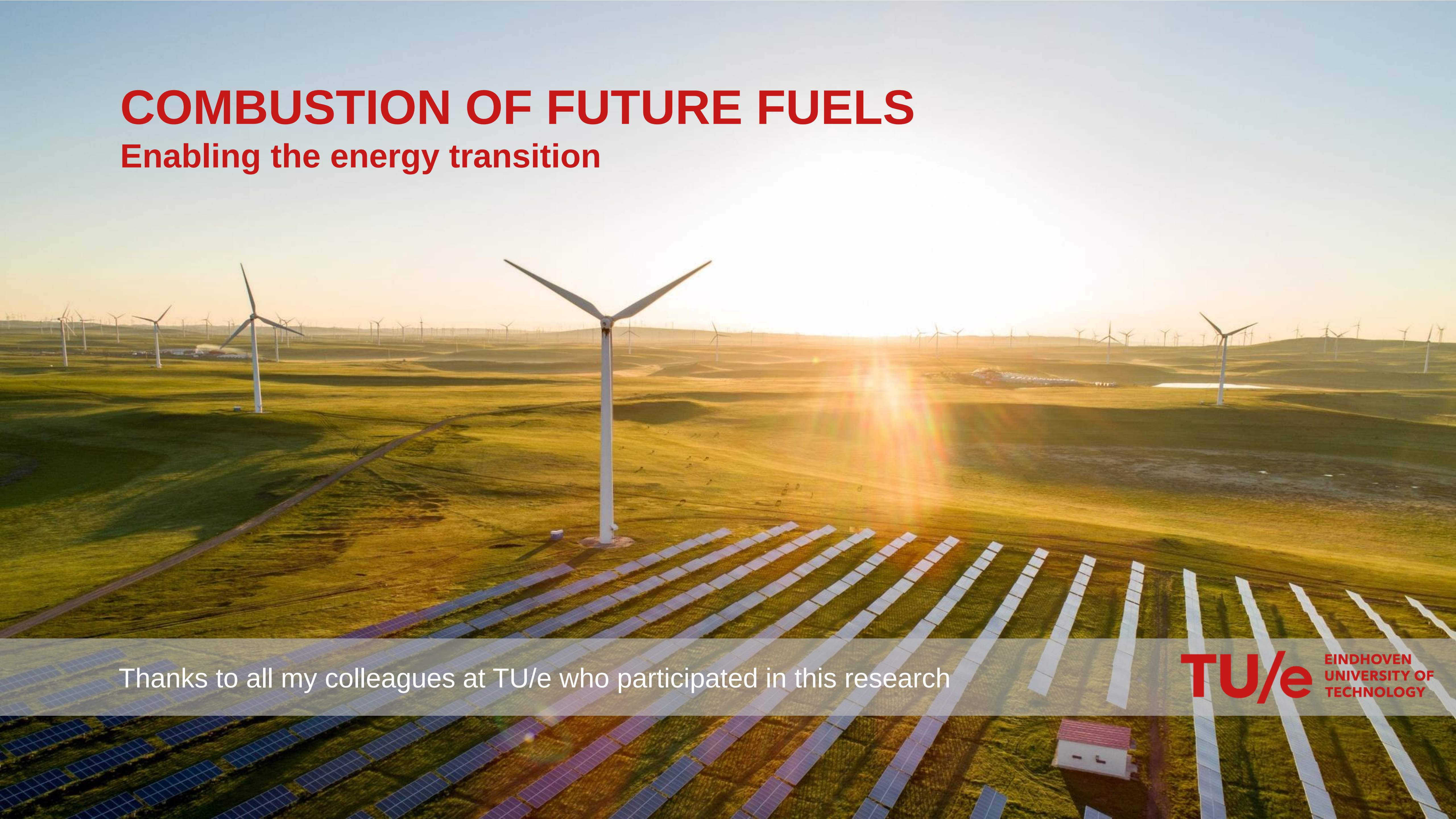
$$c = \frac{T - T_u}{T_b - T_u}$$

Why hydrogen flames are different

- High diffusivity of H_2 leads to strong preferential diffusion effects
- Response of lean H_2 flames to stretch is opposite to that of most common fuels

Lean hydrogen flames burn stronger when stretched

- Huge impact on flame dynamics and stabilization
  - Cellular instabilities
  - Increased flame surface density
  - Anomalous blow-off behavior
  - Prone to flashback due to enhanced burning rate near flame holder



COMBUSTION OF FUTURE FUELS

Enabling the energy transition

Thanks to all my colleagues at TU/e who participated in this research

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