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

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Hydrogen

Hydrogen receives a lot of attention

- No CO₂ is emitted when H₂ 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 fuel

- Hydrogen as (partial) replacement for natural gas (CH$_4$)
- Existing combustion equipment is usually not suited for H$_2$

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

At standard conditions
Flame properties

- High heat transfer rate
- High NO$_x$ formation rate

\[ \phi = \frac{F/A}{(F/A)_{st}} \]

- 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

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

\[ c = \frac{T - T_u}{T_b - T_u} \]

CH$_4$-air, $\phi = 0.6$
Premixed flame structure

- Governing equations steady 1D case

\[
\frac{d}{dx} \left( \rho u \right) = 0 \\
\rho u \frac{dY}{dx} - \rho D \frac{d^2 Y}{dx^2} = -\omega \\
\rho u c_p \frac{dT}{dx} - \lambda \frac{d^2 T}{dx^2} = q_c \omega
\]

- Solution (preheat zone)

\[
\rho u = m = \text{const} \\
T = T_u + (T_b - T_u) \exp(x/\delta_f) \\
Y = Y_u - Y_u \exp(Le \times \delta_f)
\]

Flame thickness: \( \delta_f = \lambda / (mc_p) \)

Lewis number: \( Le = \lambda / (\rho Dc_p) \)
Integral analysis

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

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

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

\[
m_u = m_b = m^0_b
\]

\[
m_u Y_u = \int_u^b \omega \, dx
\]

\[
m^0_b = \frac{1}{Y_u} \int_u^b \omega \, dx
\]

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

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

- Integral analysis

\[
\begin{align*}
    m_b - m_u &= -\int \rho K \, dx \\
    -m_u Y_u &= -\int \omega \, dx - \int \rho KY \, dx \\
    &= -m_b^0 Y_u - \int \rho KY \, dx
\end{align*}
\]

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

\[
\frac{m_b}{m_b^0} = 1 - \frac{1}{m_b^0} \int \rho KY \, dx = 1 - K_a
\]

Karlovitz integral, $K_a$
Non-unity Lewis number effects

- Enthalpy profiles \( h = q_c Y + c_p T \)

Dashed curves: \( Le = 0.5 \)

Integrating enthalpy equation

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

Combining with continuity equation

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

Le < 1 and \( K > 0 \) \( \Rightarrow \) \( h_b > h_u = h_b^0 \)

\( \Rightarrow \) \( T_b > T_b^0 \)

\( \Rightarrow \) \( \omega > \omega^0 \)
Preferential diffusion effects

- In general, all species have different $Le_i \neq 1$
- Results in changes in element mass fractions $\Delta Z_j$ at the burnt side
- Affects the equilibrium composition at the burnt side $T_b, Y_{i,b}$
- And thus, the reaction rates $\omega$ and the mass consumption rate $m_b^0 = m_b^0(h_b, Z_{j,b})$
- Mass burning rate

$$m_b = m_b^0(h_b, Z_{j,b}) [1 - Ka]$$

- Linearized for weak stretch, $Ka \ll 1$

$$\frac{m_b}{m_b^0} = 1 - Ka + \Delta h_b \frac{\partial \ln(m_b^0)}{\partial h_b} + \Delta Z_{j,b} \frac{\partial \ln(m_b^0)}{\partial Z_{j,b}} = 1 - Ma Ka$$

Direct stretch effect

Indirect, preferential diffusion effects

Markstein number $Ma$
Comparison with simulations

- Stretched CH$_4$-air flames in counterflow

- Stretched CH$_4$-H$_2$-air flames

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\[ \Delta Z_{i,b} \left( \partial \ln(m^0_b) / \partial Z_{i,b} \right) \]

\[ Le_i \neq 1 \]

\[ \phi = 0.8 \]
Markstein number

\[
\frac{m_b}{m_b^{0}} = 1 - \text{Ma Ka}
\]
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)
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

**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: \(\phi \uparrow\)
- Resulting in local higher burning rate: \(s_L \uparrow\)
- Affects stabilization a lot

\[
\begin{align*}
\text{CH}_4 & \quad \phi = 0.58 \\
V_0 & = 30 \text{ cm/s} \quad V_0 = 40 \text{ cm/s} \quad V_0 = 60 \text{ cm/s} \quad V_0 = 80 \text{ cm/s} \\
\text{0.6 CH}_4 + 0.4 \text{ H}_2 & \quad \phi = 0.52 \\
V_0 & = 30 \text{ cm/s} \quad V_0 = 40 \text{ cm/s} \quad V_0 = 60 \text{ cm/s} \quad V_0 = 80 \text{ cm/s}
\end{align*}
\]

- 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 $\text{H}_2$

- 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\textsubscript{2} enriched flames: Neck formation
- Different blow-off mechanisms observed

H$_2$-air flame shapes approaching lean limit ($V = 1$ m/s)

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

- Combustion at extremely low $\phi$
- Simulations show same behavior

Vance et al. (2021) Energies, 14, 1977
H₂-air lean limit flames

At higher mixture velocity, lean limit is lower!

Stable flames

Blow-off
Hydrogen flame flashback

- Simulations of H$_2$-air flame on slot burner
- Decrease inlet velocity $V_{in}$ at constant $\phi$ until flame flashes back
- Enhanced burning rate due to preferential diffusion effects, leads to early flashback

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

- Unsteady simulation
- Flashback when velocity is lowered
Flamelet-Generated Manifolds for H₂ 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

$$Y_i(x) \rightarrow Y_i(y_1) \quad y_1 = \mathcal{Y}$$

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

\[ Y_i(x, h_b) \rightarrow Y_i(y_1, y_2) \quad y_1 = \mathcal{Y}, y_2 = h \]
FGM for H₂ flames

- Strong preferential diffusion effects in H₂ 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, $\Delta Z_j$'s and $\Delta h$ 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 $K$
  
  $$Y_i(x, K) \rightarrow Y_i(y_1, y_2)$$

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

  $$y_1 = \mathcal{Y}$$
  $$y_2 = \mathcal{Z} = \frac{1}{2} Z_C + Z_H$$

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Preferential diffusion effects in FGM

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

\[
\frac{\partial \rho_y}{\partial t} + \nabla \cdot (\rho uy) - \nabla \cdot \left( \frac{\lambda}{c_p} \nabla y \right) = \nabla \cdot \left[ \frac{\lambda}{c_p} \sum \alpha_i \left( \frac{1}{Le_i} - 1 \right) \nabla Y_i \right] + \omega_y
\]

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

\[
\frac{\partial \rho y}{\partial t} + \nabla \cdot (\rho uy) - \nabla \cdot \left( \frac{\lambda}{c_p} \nabla y \right) = \nabla \cdot (D \nabla y) + \omega_y
\]

- New approach assuming constant Lewis numbers

\[
\frac{\partial \rho y}{\partial t} + \nabla \cdot (\rho uy) - \nabla \cdot \left( \frac{\lambda}{c_p} \nabla y \right) = \nabla \cdot \left( \frac{\lambda}{c_p} \nabla \beta \right) + \omega_y \quad \beta = \sum \alpha_i \left( \frac{1}{Le_i} - 1 \right) Y_i
\]

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$_2$, $\varphi = 0.7$)
- 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

2D FGM with Le effects

1D FGM without Le effects

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

Unsteady phenomena

- Response to sudden jump in stretch rate

- 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

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

Other modeling challenges

- Reactions occur at rather low temperature
  \[ H_2 + O \rightarrow OH + H \quad \text{H production, 1400 K} \]
  \[ H_2 + OH \rightarrow H_2O + H \]
  \[ O_2 + H + M \rightarrow HO_2 + M \quad \text{H consumption, close to } T_u \]
- Peak heat release rate at \( T < 1000 \text{ K} \)
- 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
Why hydrogen flames are different

- High diffusivity of $\text{H}_2$ leads to strong preferential diffusion effects
- Response of lean $\text{H}_2$ 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
Thanks to all my colleagues at TU/e who participated in this research