CERFACS

CENTRE EUROPÉEN DE RECHERCHE ET DE FORMATION AVANCÉE EN CALCUL SCIENTIFIQUE

CFD tools specificities for H2 flames and available/recommended models in AVBP for hydrogen/air flames.

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H2 Week • IMFT • 2024.02.27

Chemistry

Chemical Kinetics

Just this once, H_2 *is simpler to model than conventional fuels*

Conventional fuels (e.g. kerosene)

- Global chemistries:
  $N_{\mathrm{spec}} \simeq 6,\ N_{\mathrm{reac}} \simeq 2$
- Analytically Reduced Chemistries
  $N_{\mathrm{spec}} \simeq 20-30,\ N_{\mathrm{reac}} \simeq 200-500$
- Detailed chemistries (e.g. CRECK [1]):
  $N_{\mathrm{spec}} \simeq 600,\ N_{\mathrm{reac}} \simeq 30000$

Hydrogen

- Global chemistries:
  $N_{\mathrm{spec}} \simeq 4,\ N_{\mathrm{reac}} \simeq 1$
- ~~Analytically Reduced Chemistries~~
- Detailed chemistries (e.g., UCSD aka San Diego [2]):
  $N_{\mathrm{spec}} \simeq 9,\ N_{\mathrm{reac}} \simeq 21$

[1] Bieleveld et al., *Proc. Comb. Inst.*, 2009

[2] Saxena & Williams, *Combust. Flame*, 2006

Transport properties

Transport properties

Laminar viscosity of the gas

- For conventional fuels, two dynamic viscosity laws are typically used:

Sutherland

$$\mu = c_1 \frac{T^{3/2}}{T + c_2} \frac{T_{ref} + c_2}{T_{ref}^{3/2}}$$

Power law

$$\mu = c_1 \left(\frac{T}{T_{ref}} \right)^b$$

These laws are based on air.

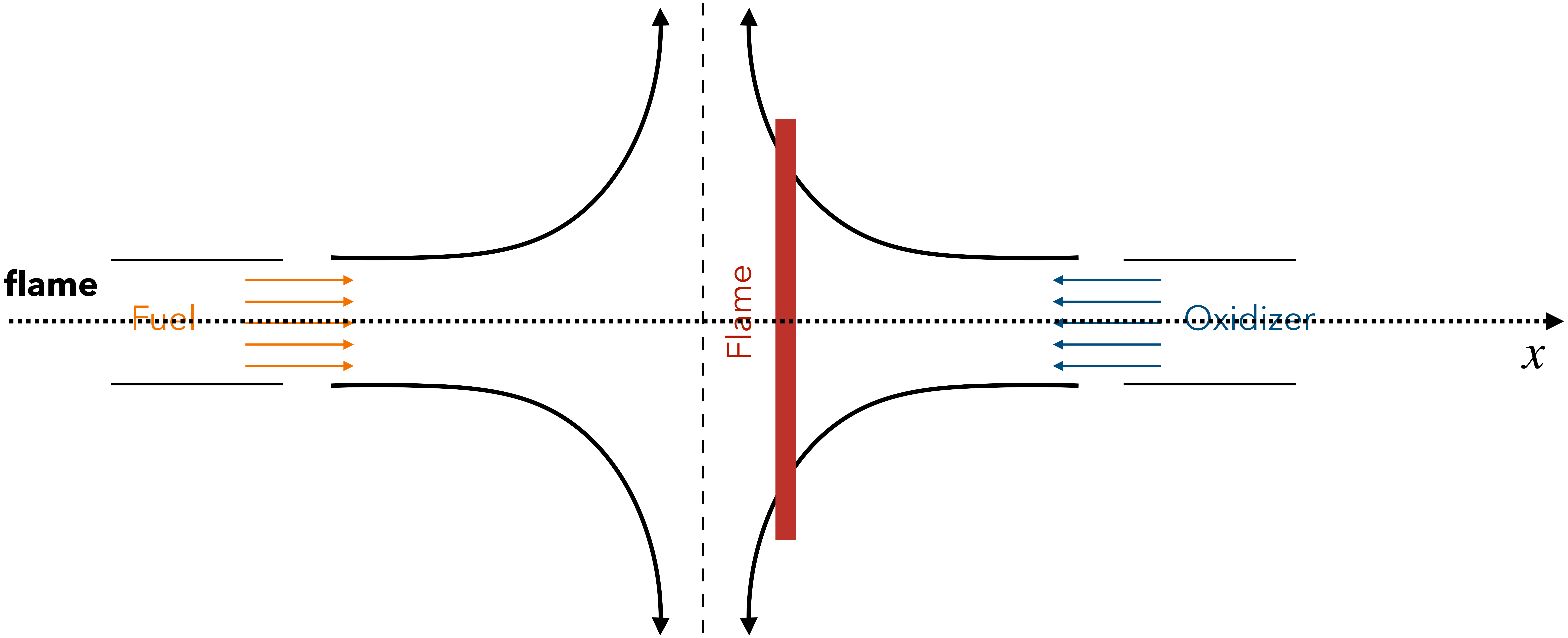
→ is this valid for H_2/Air flames?

Transport properties

Laminar viscosity of the gas

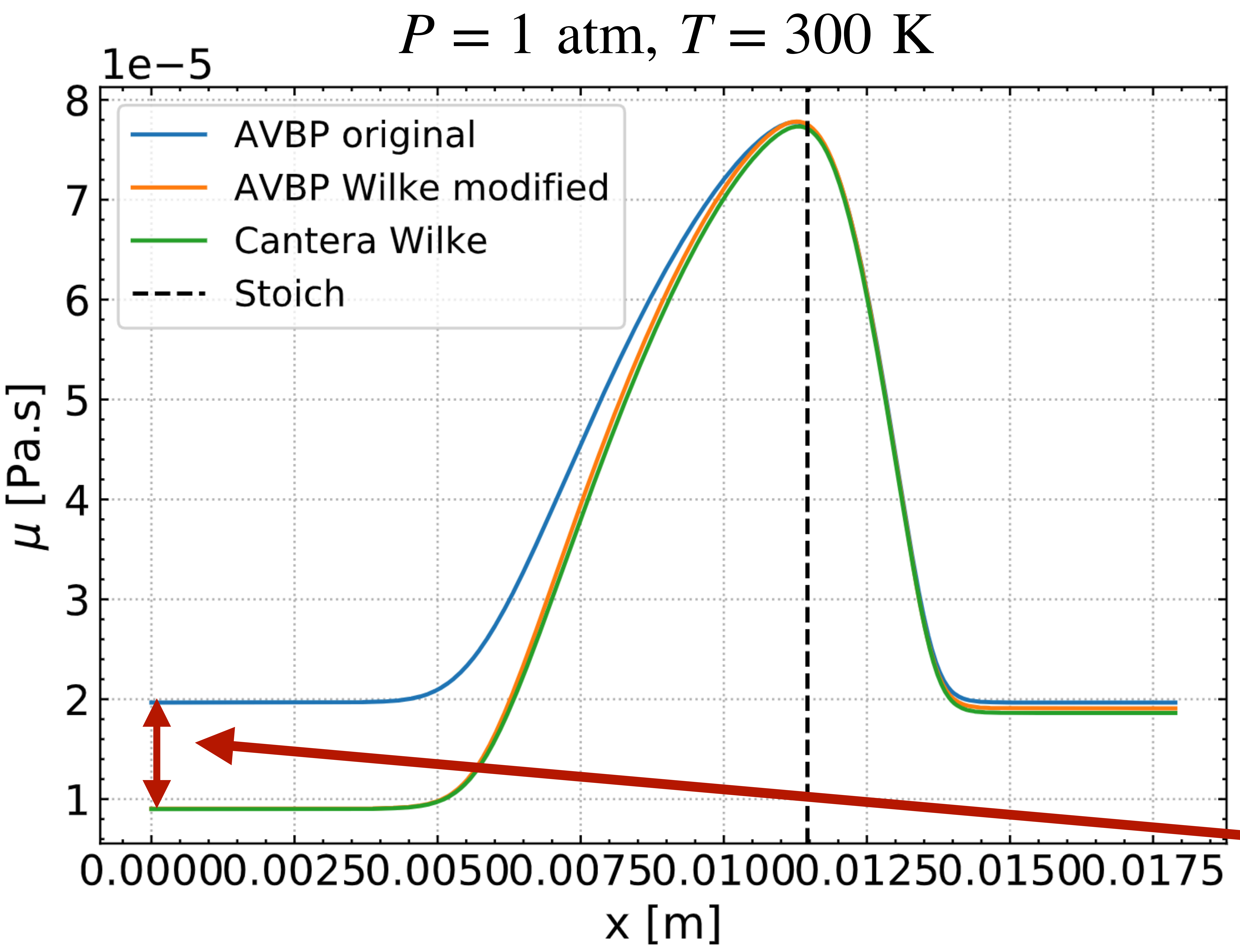
Let's consider a canonical counterflow non-premixed flame

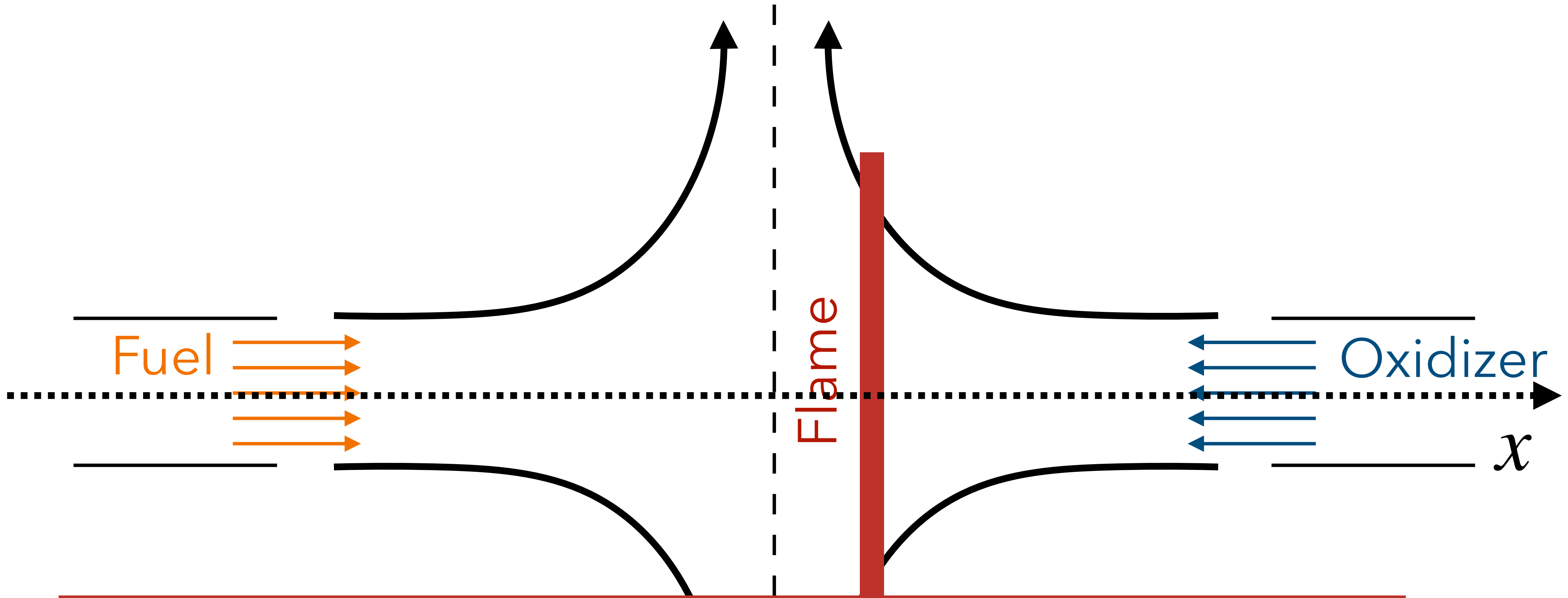
1D non-premixed flame



Transport properties

Laminar viscosity of the gas





Laminar viscosity is wrong by a factor 2 in pure hydrogen

Transport properties

Laminar viscosity of the gas

Wilke's binary mixing between Pure H2 and Air

$$\boxed{\mu = \frac{X_{H_2}\mu_{H_2}}{X_{H_2} + (1 - X_{H_2})\phi_{H_2,\mathrm{air}}} + \frac{(1 - X_{H_2})\mu_{\mathrm{air}}}{X_{H2}\phi_{air,H_2} + 1 - X_{H_2}}}$$

$$\phi_{air,H_2} = \frac{\left[1 + \left(\frac{\phi_{air}}{\phi_{H_2}}\right)^{1/2}\left(\frac{W_{H_2}}{W_{air}}\right)^{1/4}\right]^2}{2\sqrt{2}\left(1 + \frac{W_{air}}{W_{H_2}}\right)^{1/2}}$$

Power law

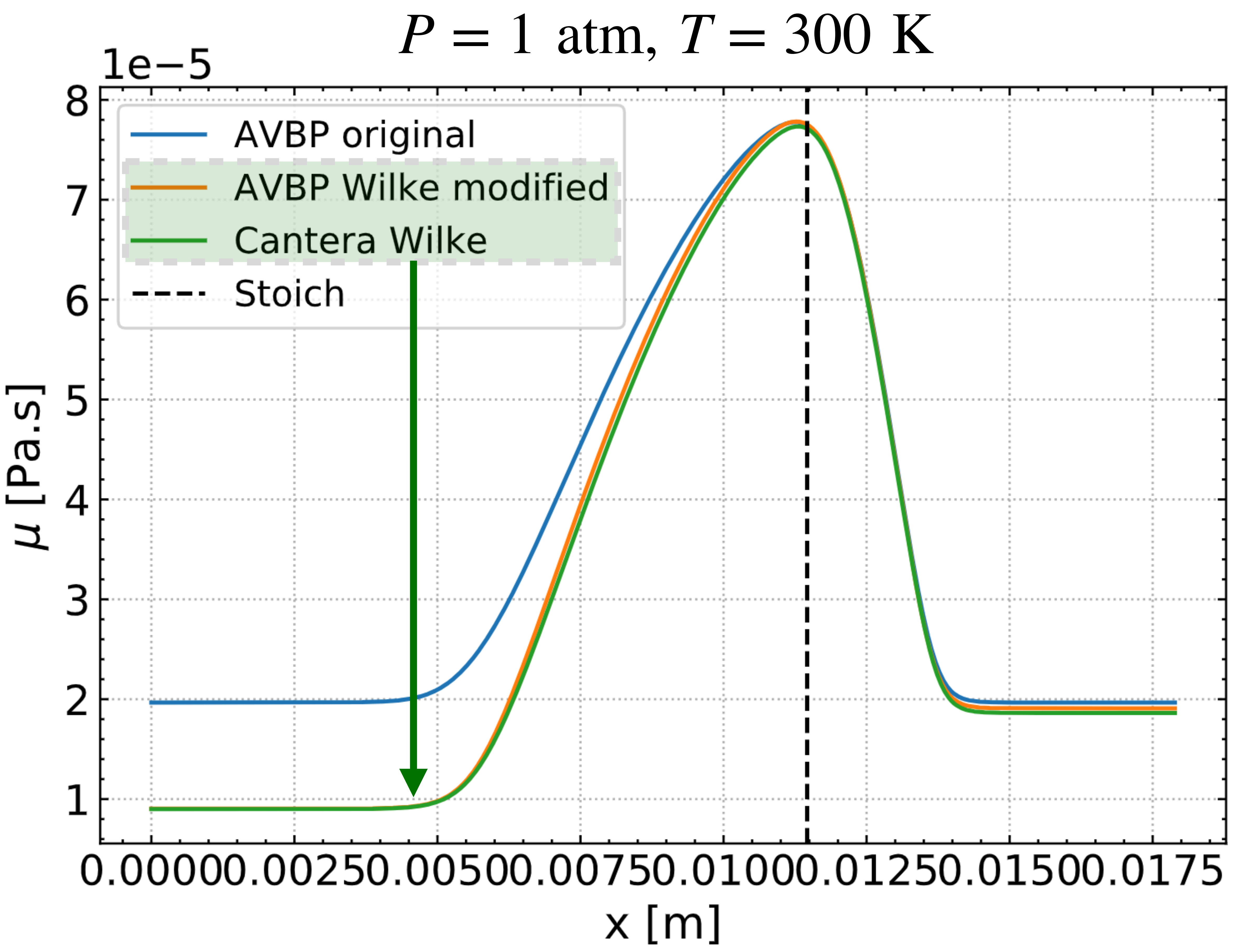
$$\mu_{H_2}(T) = \mu_{ref,H_2}\left(\frac{T}{T_{ref,H_2}}\right)^{\alpha_{H_2}}$$

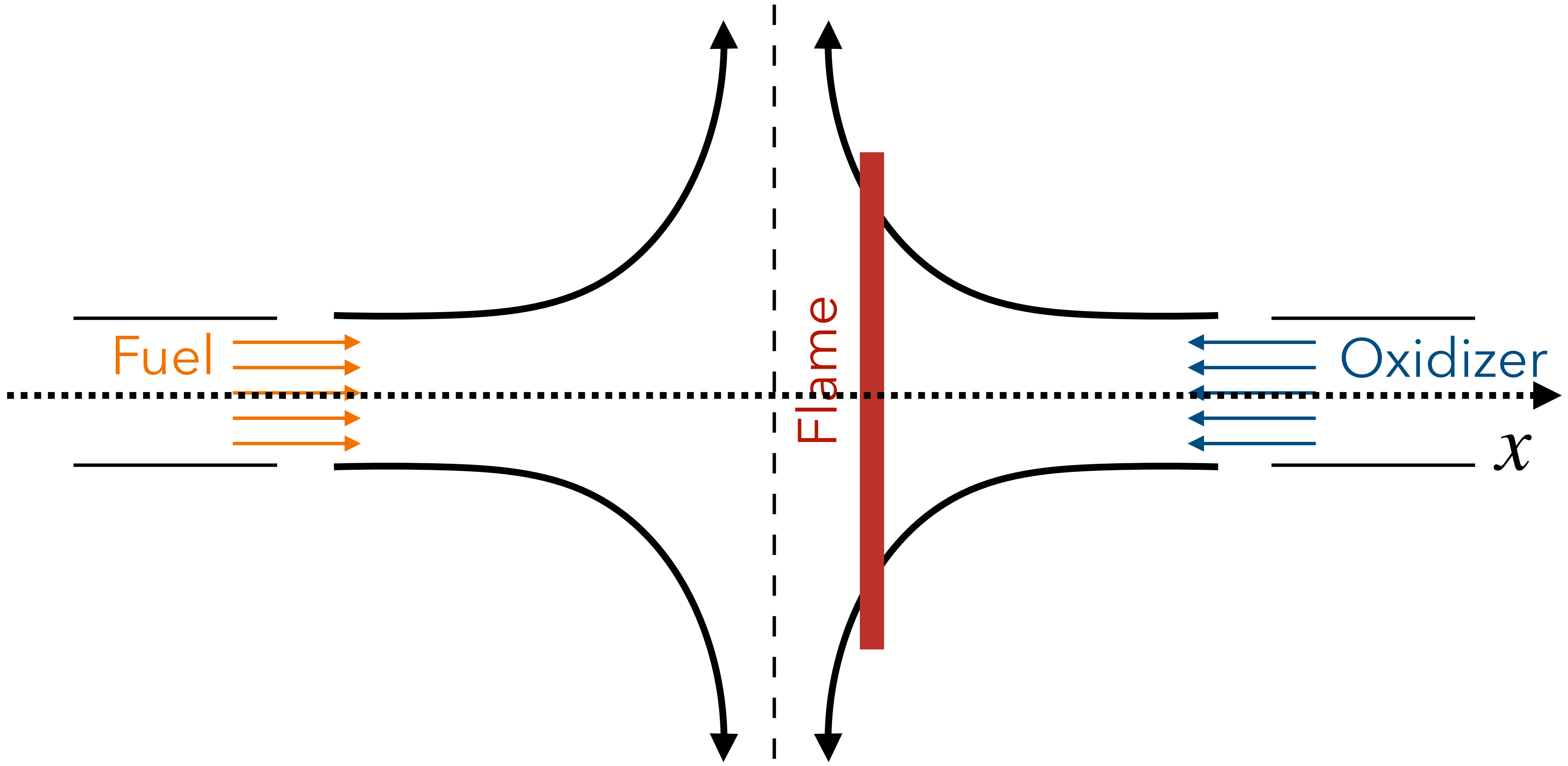
$$\mu_{air}(T) = \mu_{ref,air}\left(\frac{T}{T_{ref,air}}\right)^{\alpha_{air}}$$

D. Laera & F. Garnier

Transport properties

Laminar viscosity of the gas

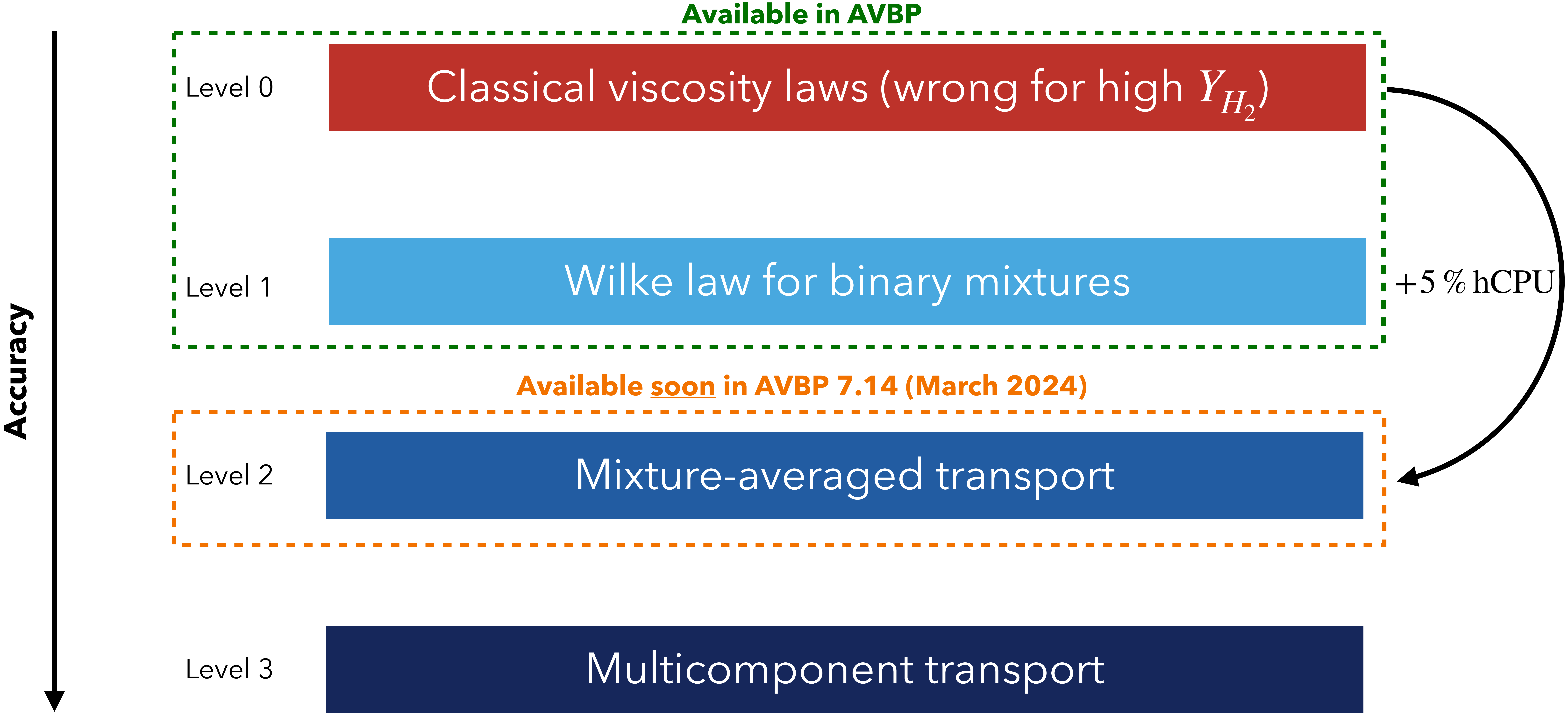




D. Laera & F. Garnier

Transport properties

Laminar viscosity



Available in AVBP

Level 0 — Classical viscosity laws (wrong for high Y_{H_2})

Level 1 — Wilke law for binary mixtures

$+5\ \%$ hCPU

Available soon in AVBP 7.14 (March 2024)

Level 2 — Mixture-averaged transport

Level 3 — Multicomponent transport

Transport properties

Species diffusion & heat conduction

Species diffusion coefficient

- Based on Schmidt number

$$D_k = \frac{\mu}{\rho \mathrm{Sc}_k}$$

- For Wilke’s law: we changed μ, Sc_k has to be adjusted to recover the correct Species diffusion coefficient

Heat conduction coefficient

- Based on Prandtl number

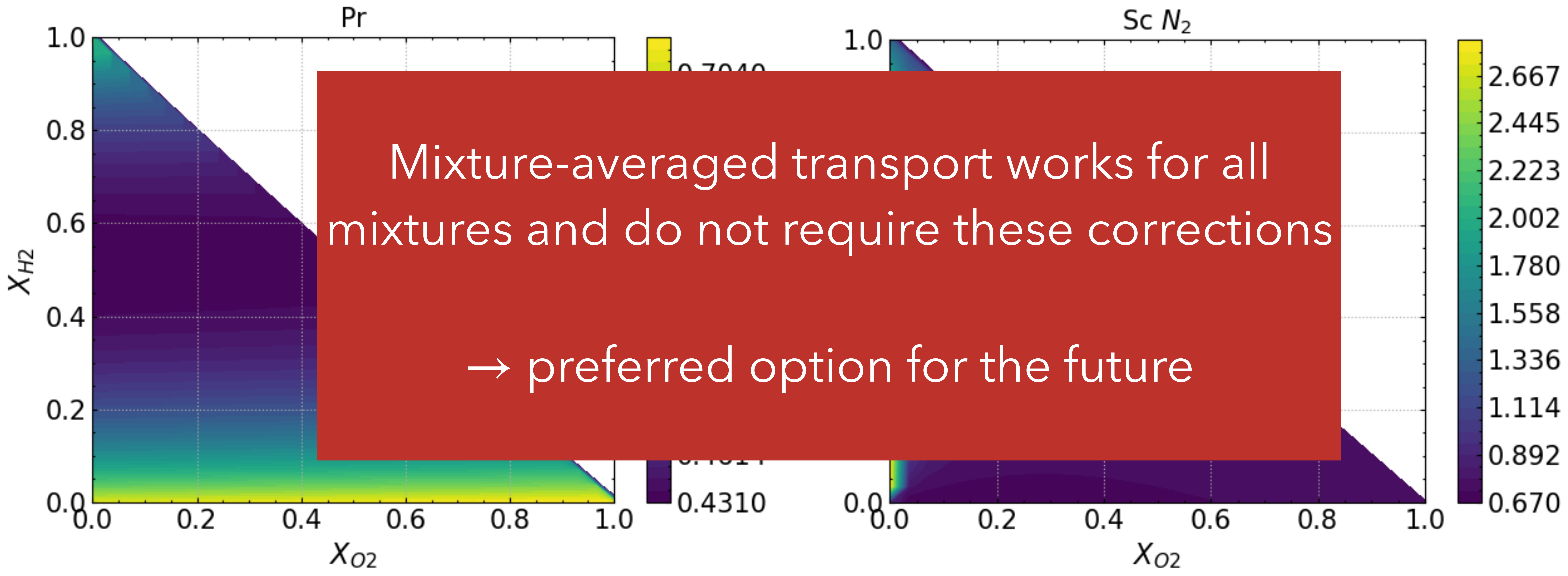
$$\lambda = \frac{\mu C_p}{\mathrm{Pr}}$$

- For Wilke’s law: we changed μ, P_r has to be adjusted to recover the correct heat condition coefficient

Transport properties

Wilke's Law: variable Prandlt & Schmidt numbers

- Pr_k and Sc_k of the major species are tabulated: H_2, O_2, N_2, H_2O



Mixture-averaged transport works for all mixtures and do not require these corrections

→ preferred option for the future

Non-adiabatic wall treatments

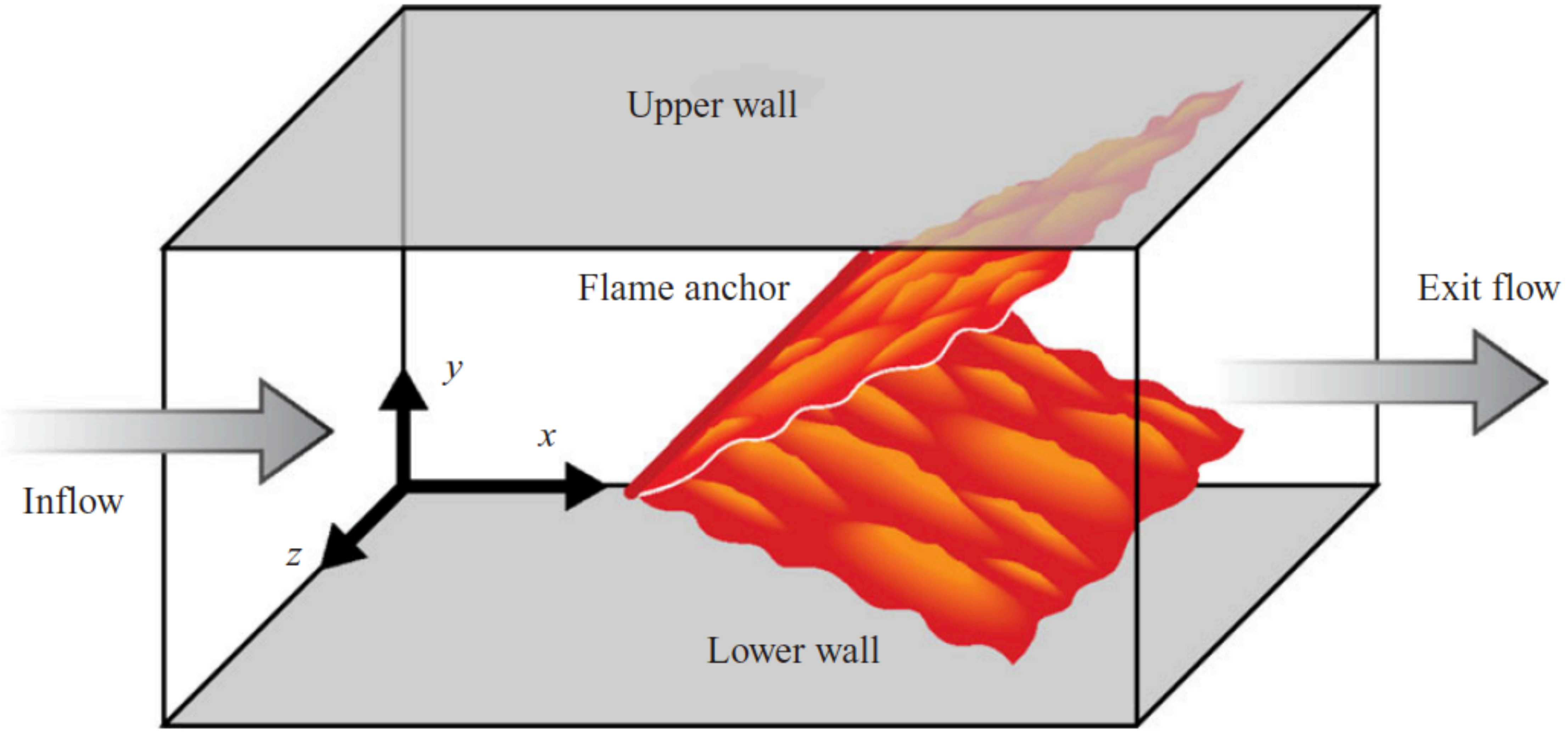
IFHC model for H_2 FWI

i Laminar FWI - standard wall treatment

- **A well-known case in the literature of H_2 FWI [1]**

$T_w = 750$ K

H_2/Air
$\phi = 1.5$
$P_{in} = 1$ bar
$T_{in} = 750$ K



$P_{out} = 1$ bar

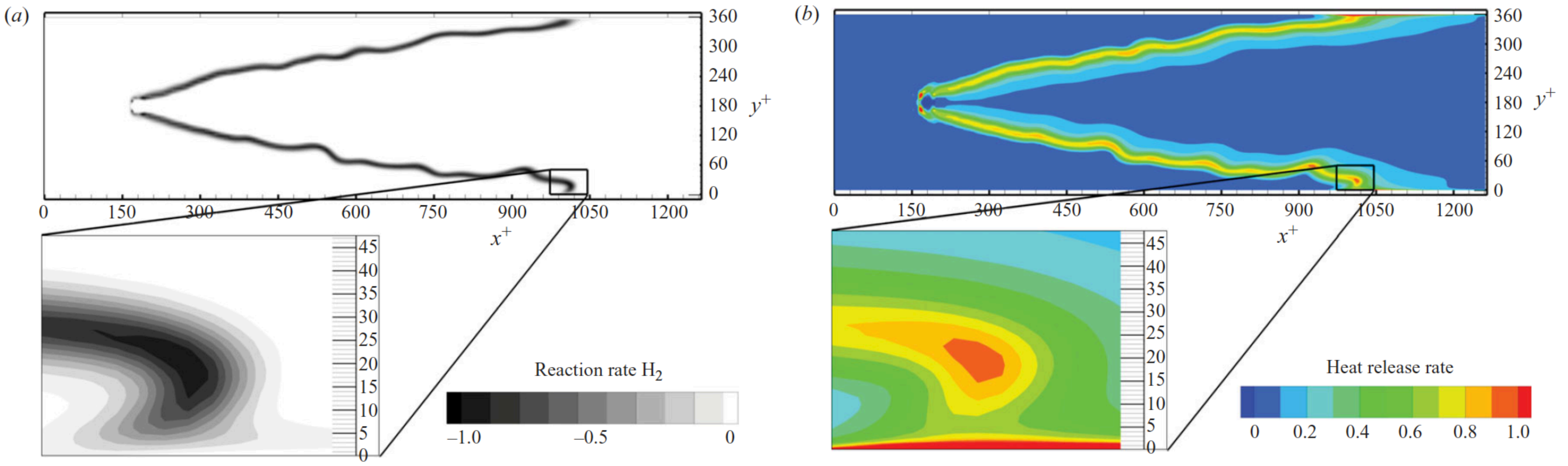
$T_w = 750$ K

[1] Gruber et al. (JFM, 2010)

IFHC model for H_2 FWI

i Laminar FWI - standard wall treatment

- **A well-known case in the literature of H_2 FWI [1]**



a) H_2 reaction rate vanishes at the wall → local quenching

b) Heat release rate peaked values at the wall → unexpected behavior

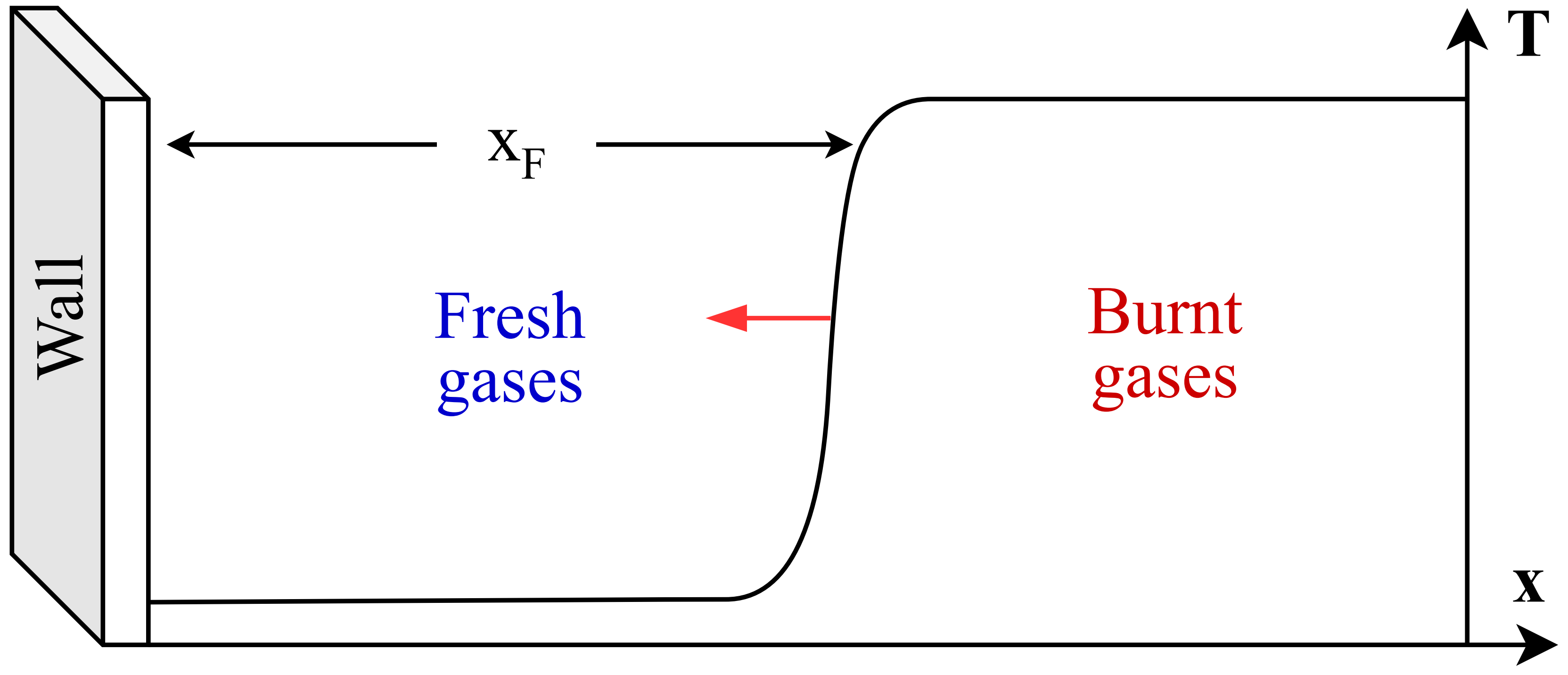
[1] Gruber et al. (JFM, 2010)

IFHC model for H_2 FWI

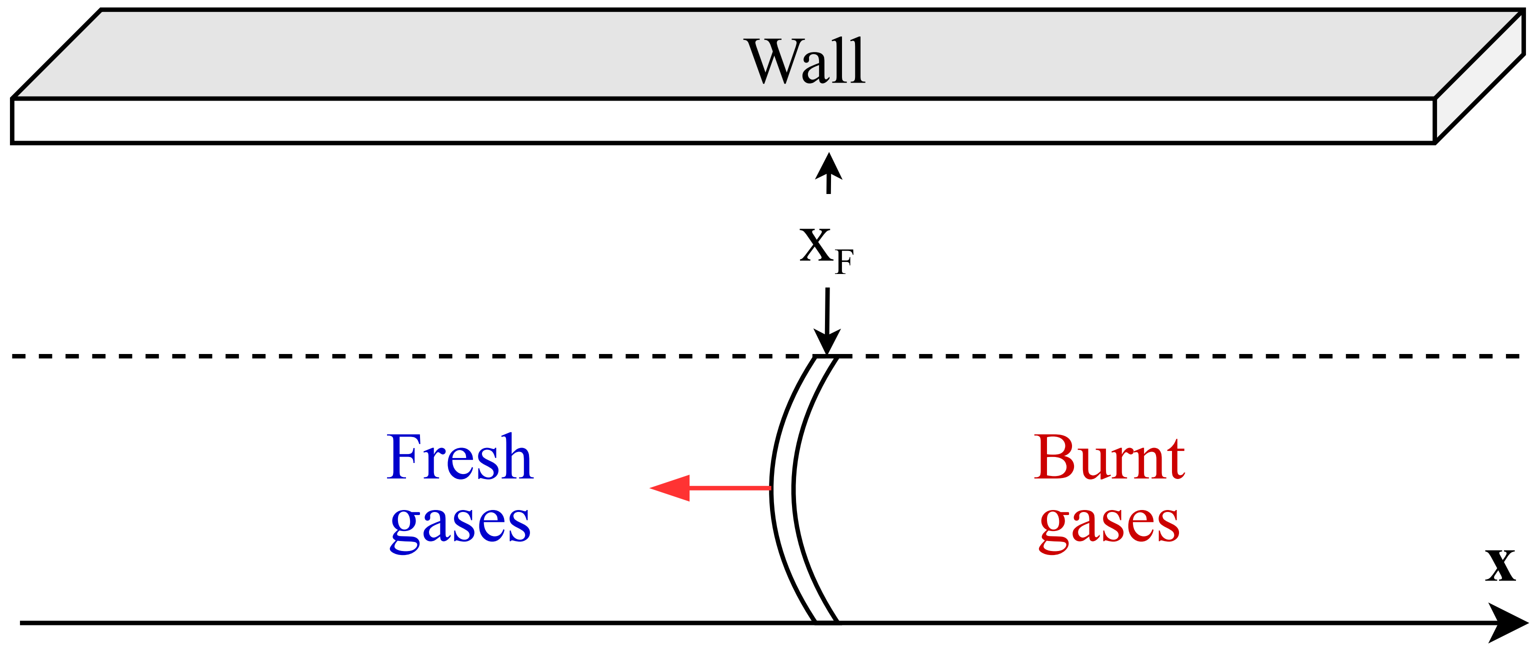
i Laminar FWI - standard wall treatment

- **FWI: complex and transient → worthwhile considering canonical cases [2]**

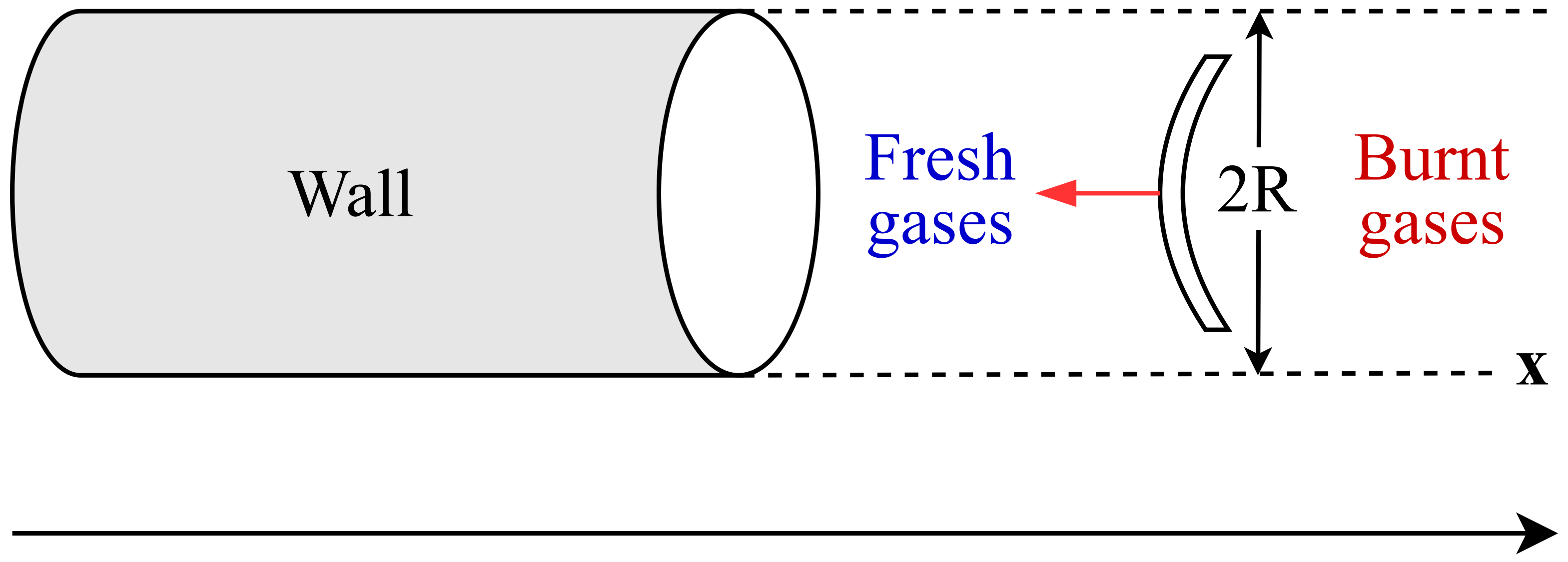
a) Head-On Quenching (HOQ)



b) Side-Wall Quenching (SWQ)



c) Tube quenching



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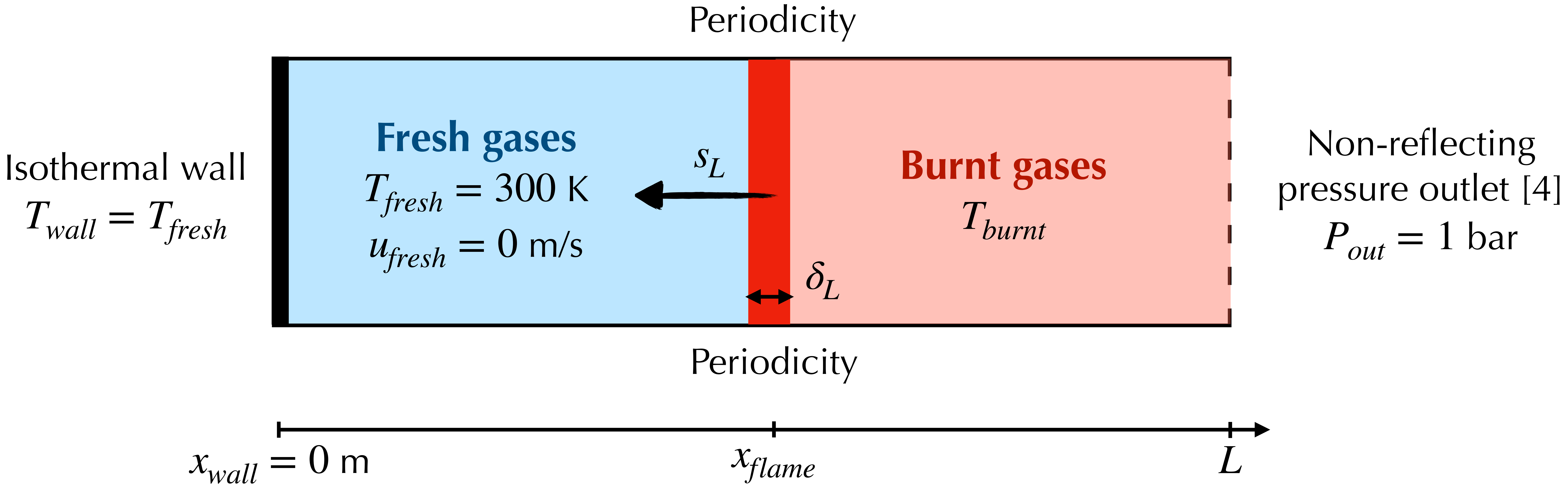
[2] Poinsot and Veynante (TNC, 2012)

IFHC model for H_2 FWI

i Laminar FWI - standard wall treatment

- **1D HOQ simulation with AVBP [3]**



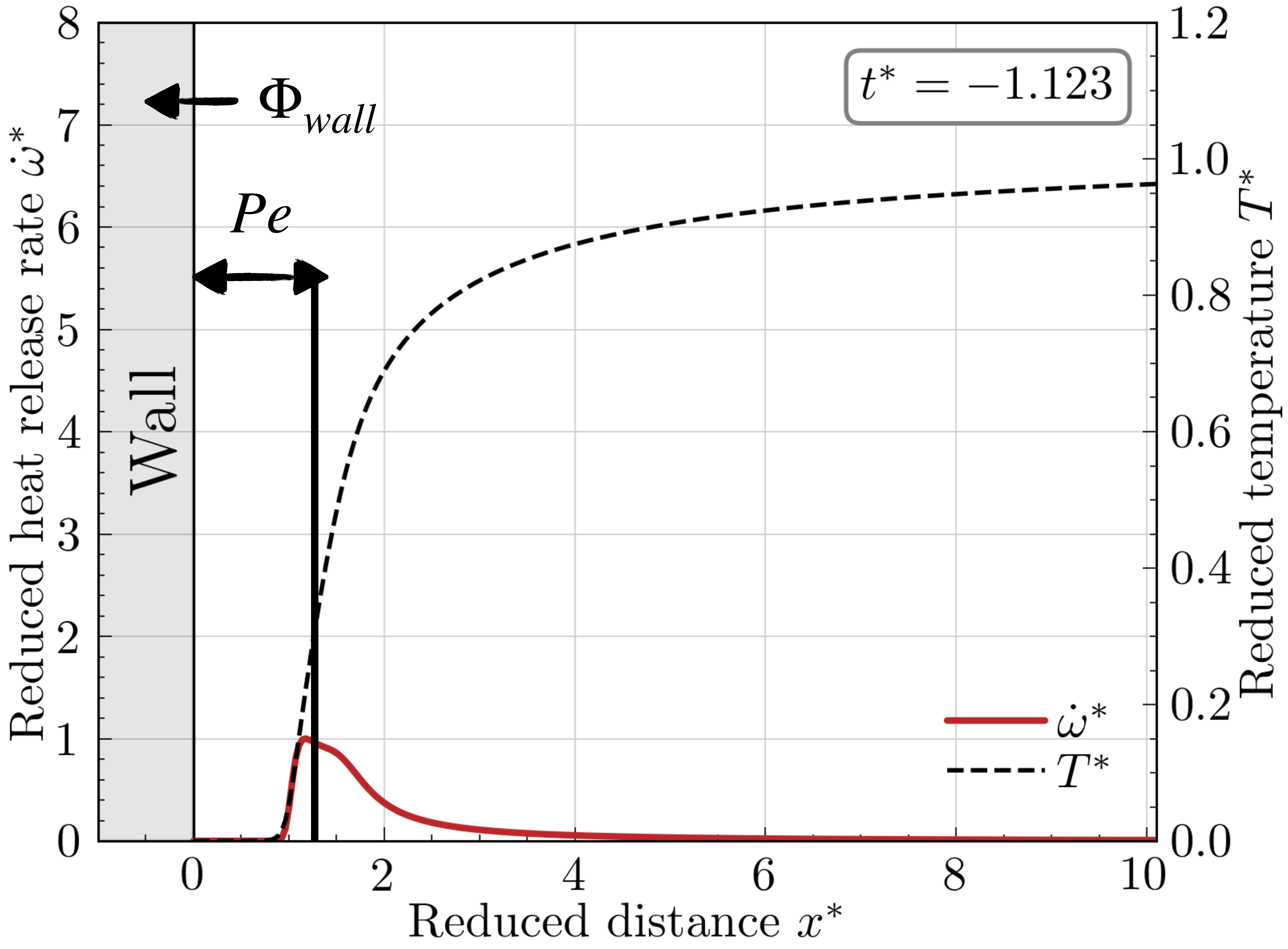
Performed with both **San Diego [5]** and **Burke [6]** schemes

[3] Schönfeld and Rudgyard (AIAA, 1999)
[4] Poinsot and Lele (JCP, 1993)
[5] Saxena and Williams (CNF, 2006)
[6] Burke et al. (IJHE, 2010)

IFHC model for H_2 FWI

i Laminar FWI - standard wall treatment

- **Definitions**



- **Reduced variables**

$x^* = x/\delta_L \longrightarrow x^* = 0$: wall position

$t^* = (t - t_Q)s_L/\delta_L \longrightarrow t^* < 0$: free propagation; $t^* = 0$: quenching instant; $t^* > 0$: post interaction

$T^* = (T - T_f)/(T_b - T_f) \longrightarrow T^* = 0$: fresh gases; $T^* = 1$: burnt gases

$\dot{\omega}^* = \dot{\omega}_T/\dot{\omega}_T^0 \longrightarrow \dot{\omega}^* = 1$: free propagation

- **Quantities of interest**

$Pe = x_{flame}/\delta_L$

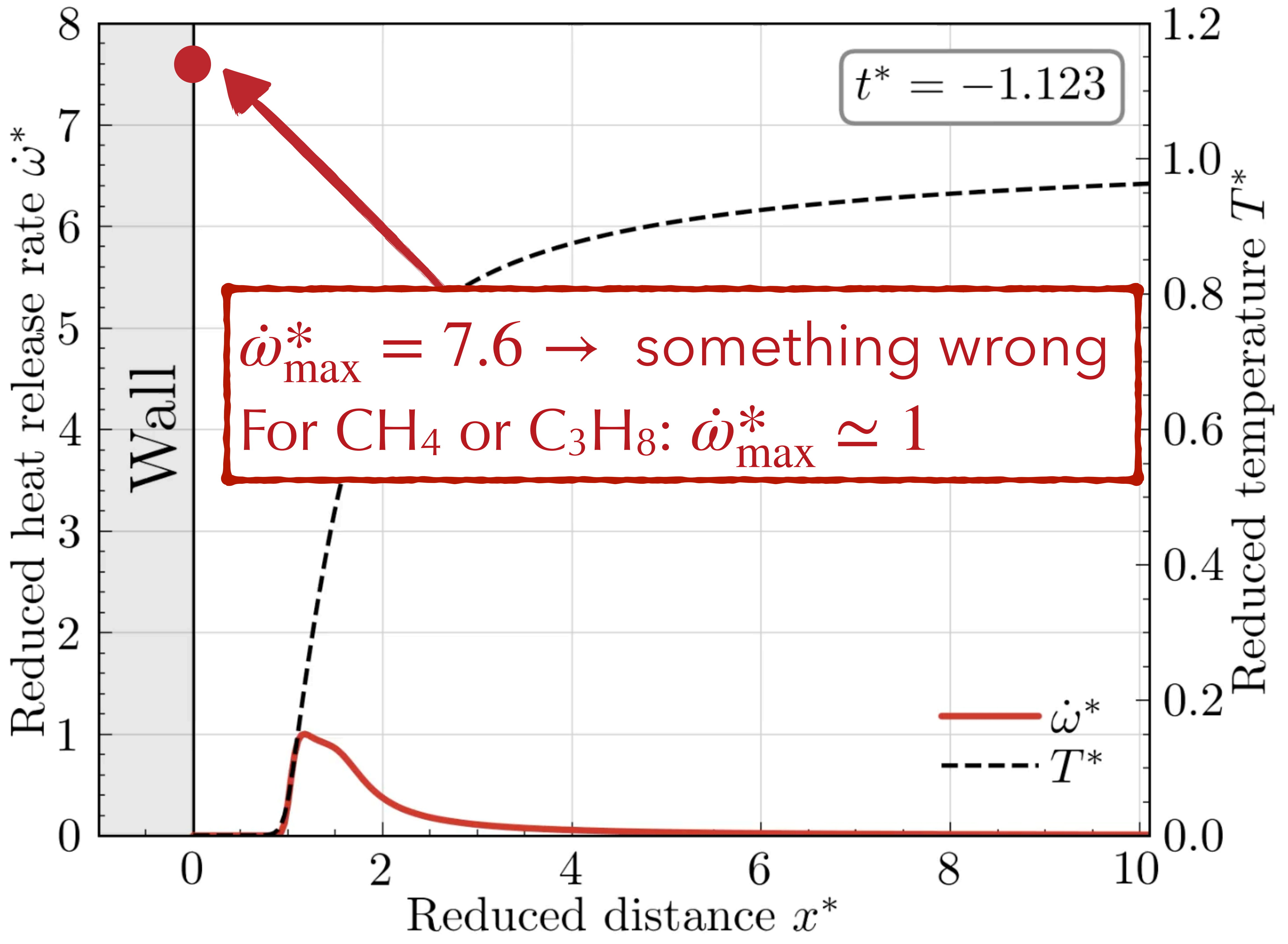
$\Phi_{wall} \longrightarrow$ easiest-to-acess physical parameter in EXP.

[21] De Nardi et al. (CNF, 2024)

IFHC model for H_2 FWI

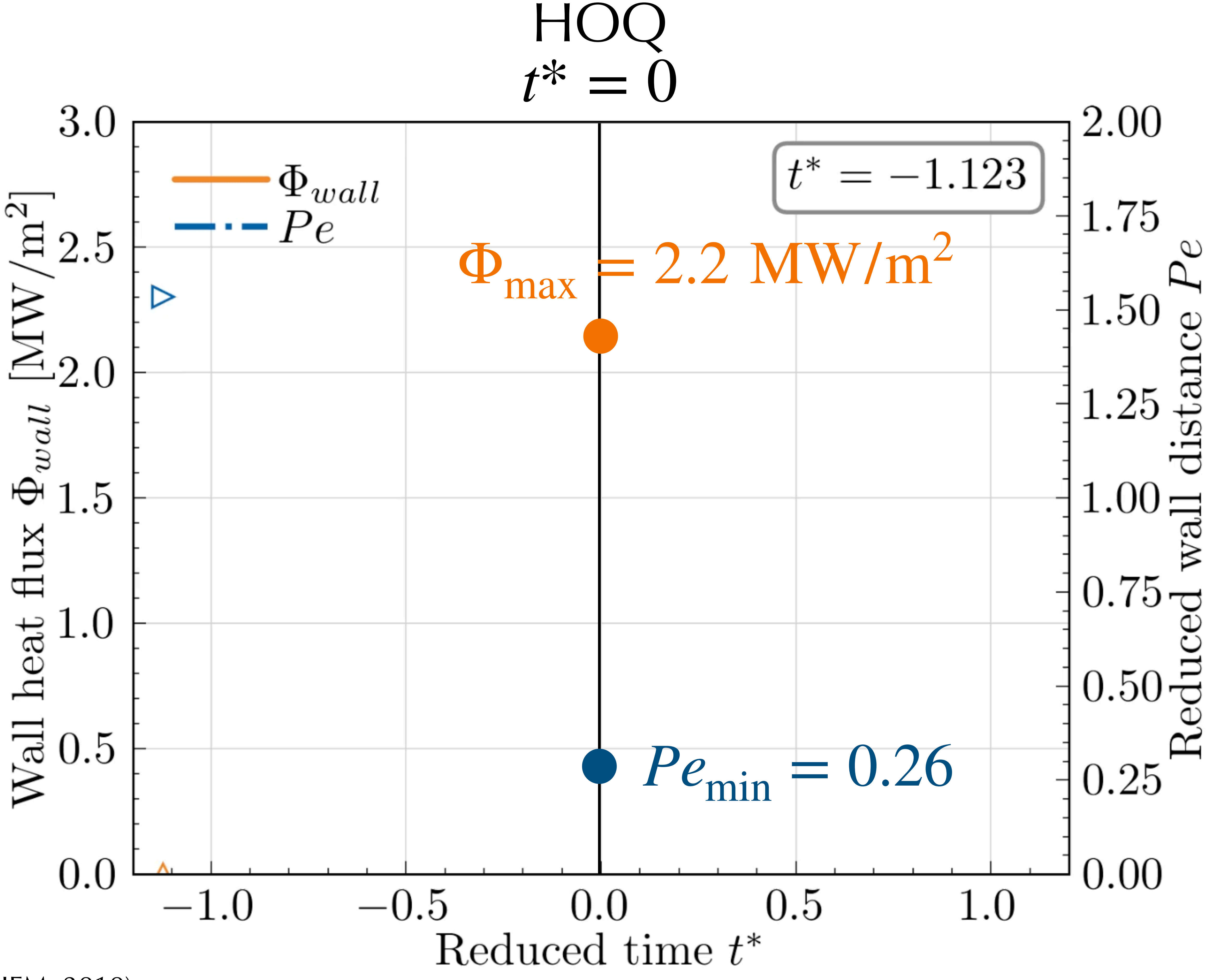
i Laminar FWI - standard wall treatment

- **Results for a standard inert wall treatment**



$\dot{\omega}^*_{\max} = 7.6 \rightarrow$ something wrong

For CH_4 or C_3H_8: $\dot{\omega}^*_{\max} \simeq 1$



$\Phi_{\max} = 2.2$ MW/m^2

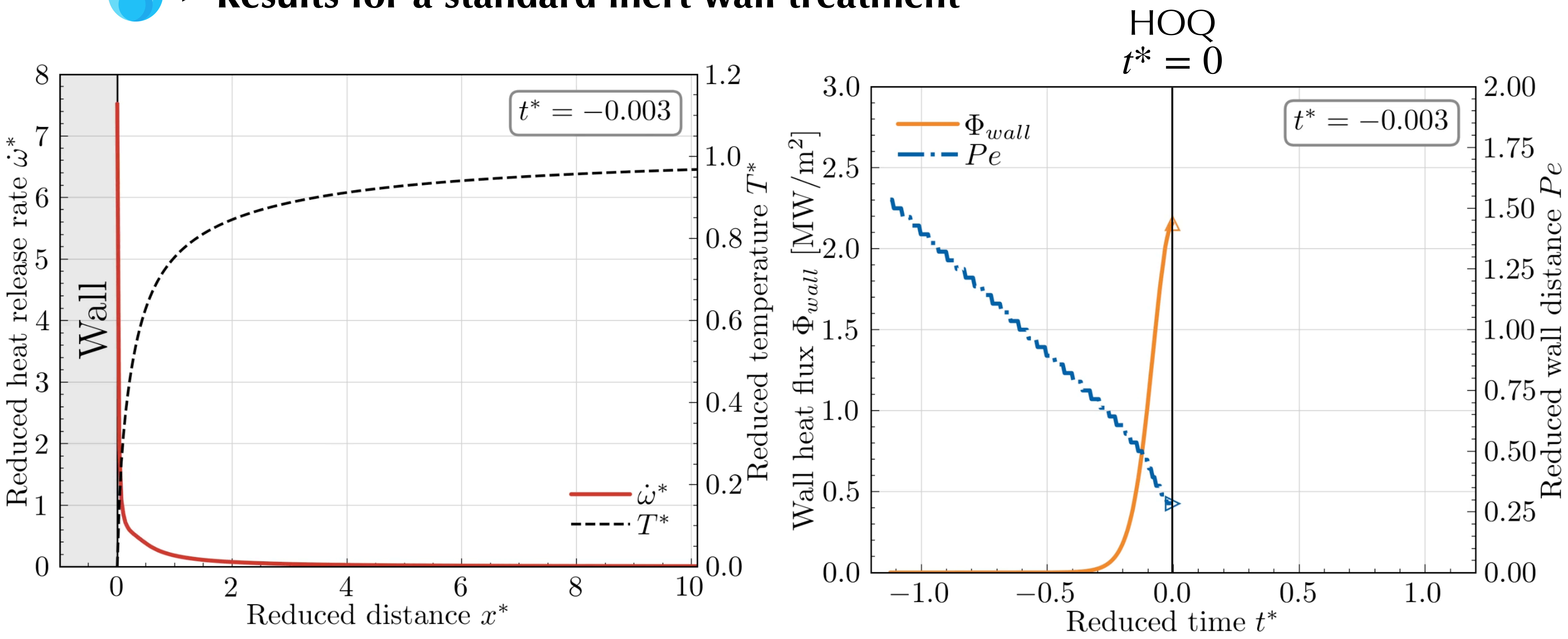
$Pe_{\min} = 0.26$

[1] Gruber et al. (JFM, 2010)
[7] Owston et al. (IJHE, 2007)
[9] Zhao et al. (CNF, 2022)
[11] Lai et al. (IJHFF, 2022)
[21] De Nardi et al. (CNF, 2024)
[8] Mari et al. (CNF, 2016)
[10] Dabireau et al. (CNF, 2003)
[12] Popp and Baum (CNF, 1997)

IFHC model for H_2 FWI

i Laminar FWI - standard wall treatment

- **Results for a standard inert wall treatment**

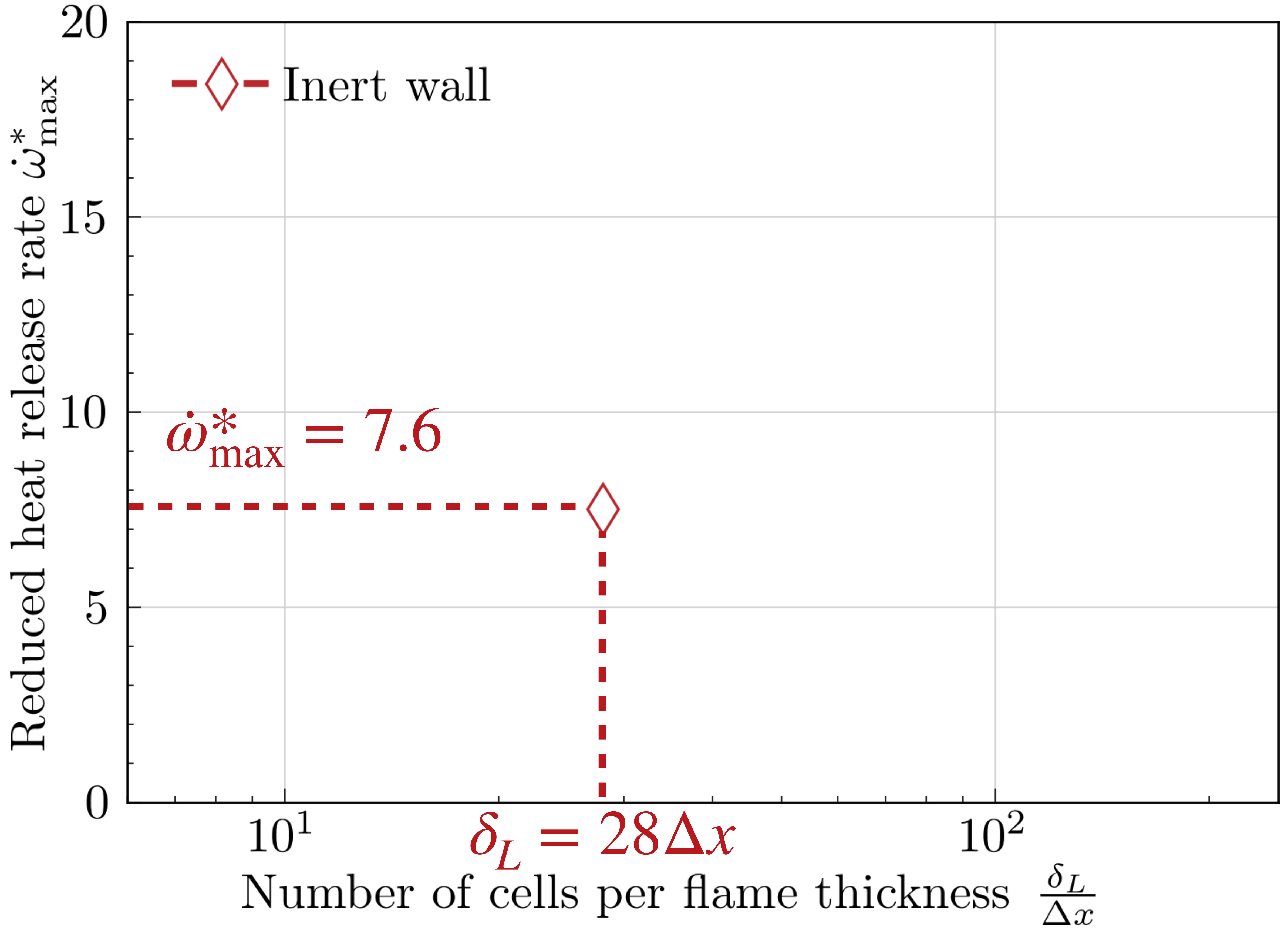


[21] De Nardi et al. (CNF, 2024)

IFHC model for H_2 FWI

i Laminar FWI - standard wall treatment

▸ **Results for a standard inert wall treatment**



- Gradient stiffened during FWI: $\delta_L = 28\Delta x$ recommended [13,15]
- Effect of grid resolution: $\delta_L = 7\Delta x - 224\Delta x$
- $\dot{\omega}^*$ peak at quenching does not converge when the mesh is refined

→ **Ill-posed problem**

[21] De Nardi et al. (CNF, 2024)

[10] Dabireau et al. (CNF, 2003) [12] Popp and Baum (CNF, 1997)

IFHC model for H_2 FWI

i Laminar FWI - standard wall treatment

- **Results for a standard inert wall treatment**

Far from the non-adiabatic wall

- Chain branching reactions:

$$\mathbf{H_2 + OH \longleftrightarrow H_2O + H} \qquad (1)$$

(E_a = 3.6 kcal/mol)

$$\mathbf{H_2 + O \longleftrightarrow OH + H} \qquad (2)$$

(E_a = 6.3 kcal/mol)

- Recombination reactions:

$$\mathbf{H + O_2\ (+\ M) \longleftrightarrow HO_2\ (+\ M)} \qquad (3)$$

(E_a = 0 kcal/mol)

Close to the non-adiabatic wall

- Cooling of the preheat zone → rate of (1) and (2) decreases
- **Very fast diffusion** of **H radical** seems to be the problem ($Le_H \simeq 0.1$)
- Only way to consume these radicals is through (3) → **highly exothermic**

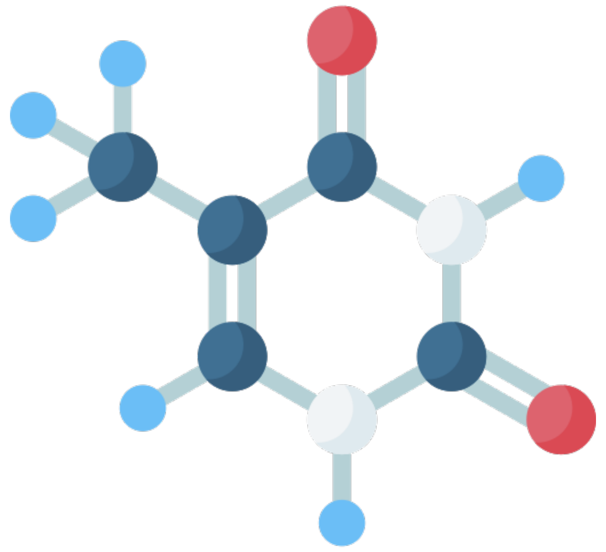
[21] De Nardi et al. (CNF, 2024)

[5] Saxena and Williams (CNF, 2006)
[6] Burke et al. (IJHE, 2010)
[13] O'connaire et al. (IJHE, 2004)
[14] Li et al. (IJHE, 2004)

IFHC model for H_2 FWI

ii Can we assume wall as inert?

- **Inert wall assumption?**
  - Lack of experimental validation for premixed H₂ FWI (Φ_{wall})
  - Sticking coefficient for H/O/OH to match EXP. explosion limits [20, 21]
  - Wall-coating material type → significant impact on FWI [22]



- **Heterogeneous catalysis: using detailed surface chemistry mechanisms**
  - Exist for hydrocarbons [23] and H₂ [24, 25]
  - Also suffer from a lack of experimental validation
  - Can significantly increase computational cost!

[15] Liu et al. (ACS, 2020) [17] Fan et al. (PCI, 2021) [19] Aghalayam et al. (CTM, 2022)
[16] Li et al. (ENF, 2022) [18] Popp and Baum (CNF, 1997) [20] Zhao et al. (CNF, 2022)

IFHC model for H_2 FWI

iii A simplified approach for FWI

- **Infinitely Fast Heterogeneous Catalysis (IFHC) [21]**
  - Global surface chemistry: total, irreversible, occurring in 1 timestep:

$$2H + O \rightarrow H_2O \qquad (1)$$

$$H + OH \rightarrow H_2O \qquad (2)$$

$$4H + O_2 \rightarrow 2H_2O \qquad (3)$$

  - Energy conservation:

$$\boxed{\dot{\omega}_{T,IFHC}} = -\sum_{k}^{N} \boxed{\dot{\omega}_{k,IFHC}} \times \boxed{\Delta h^{o}_{f,k}}$$

Enthalpy of formation of species k

Heat released at wall by IFHC

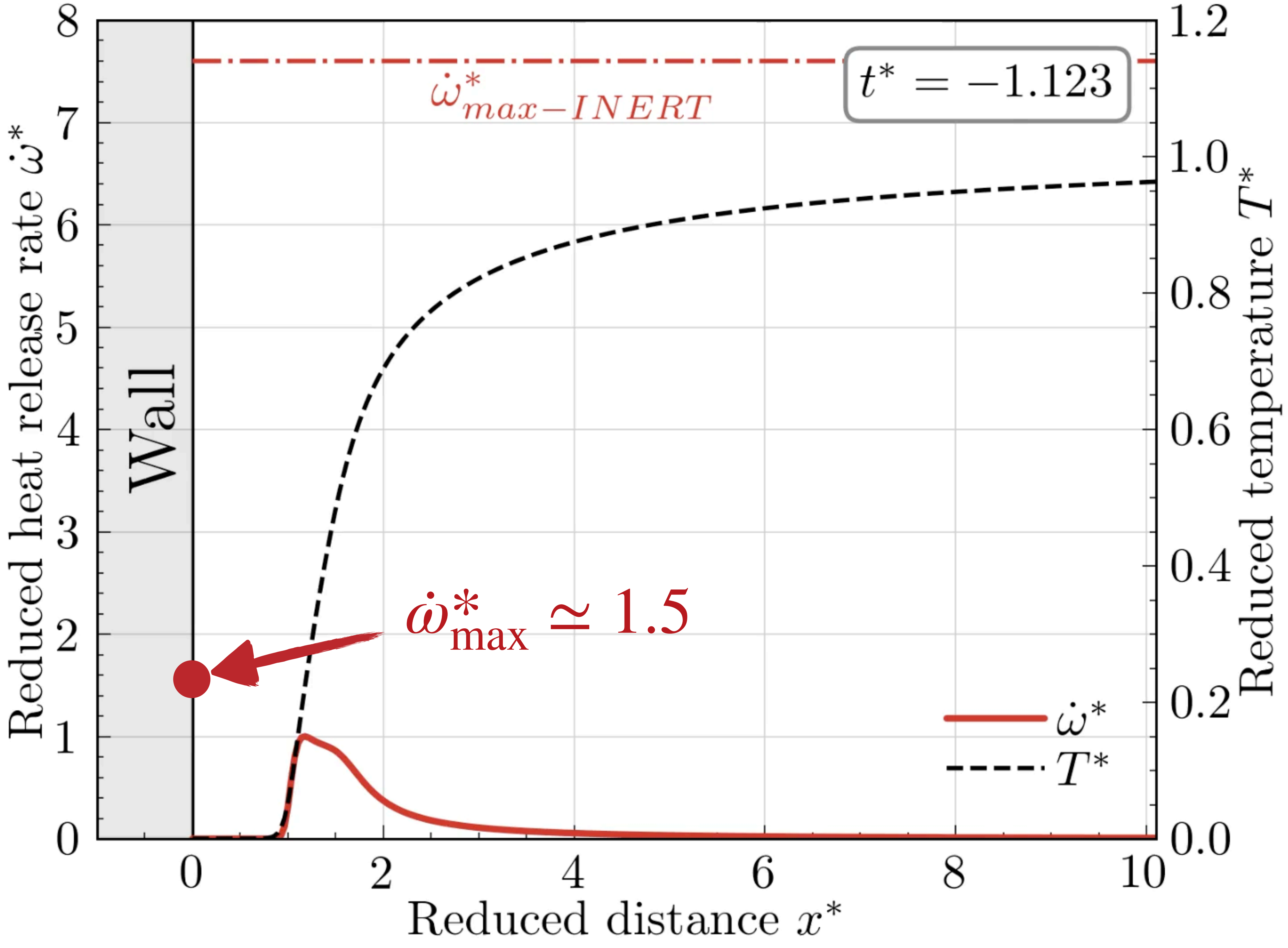
Surface reaction rate of species k

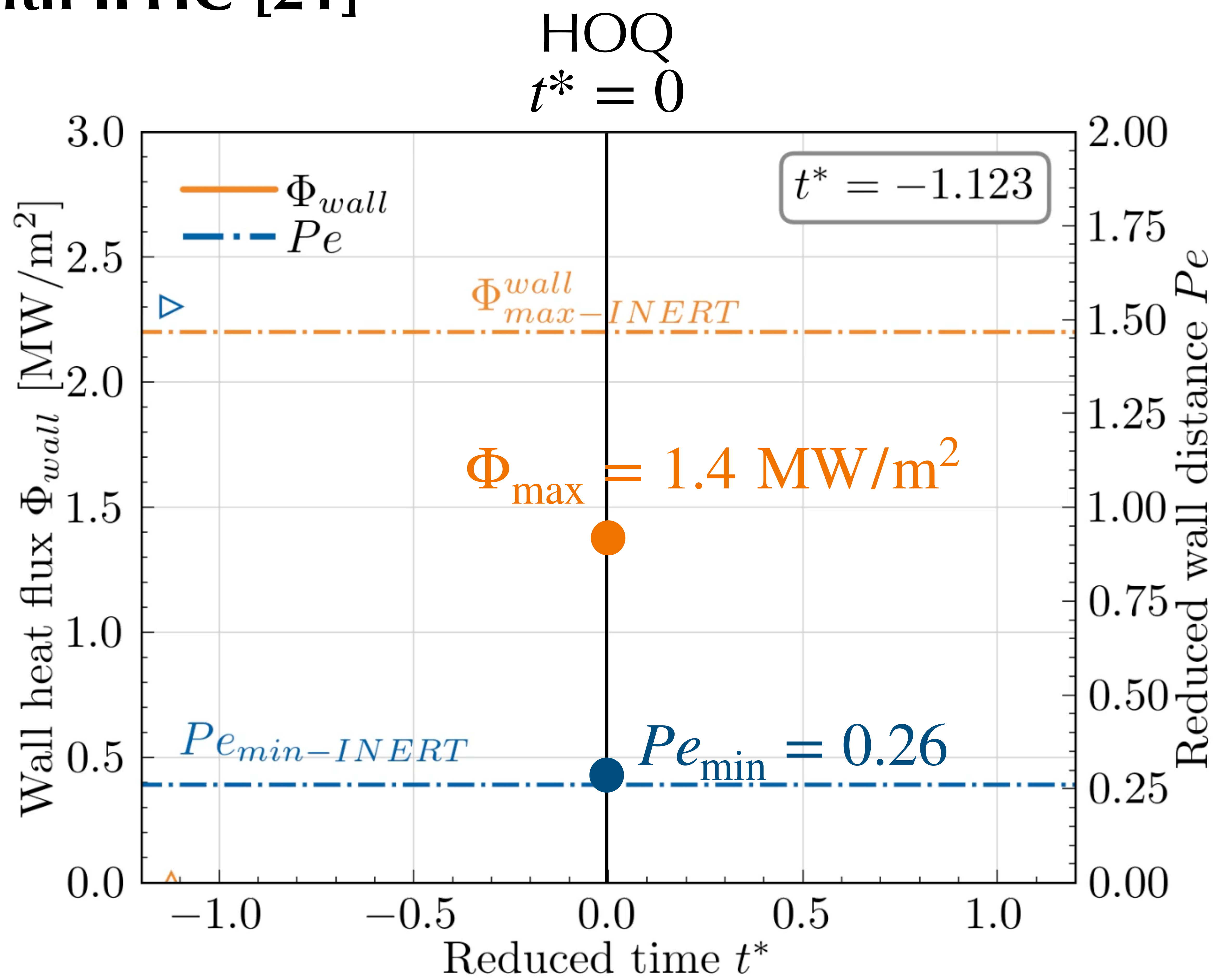
[21] De Nardi et al. (CNF, 2024)

IFHC model for H_2 FWI

iv Laminar FWI - catalytic wall using IFHC

- **Results for an isothermal wall with IFHC [21]**





[21] De Nardi et al. (CNF, 2024)

IFHC model for H_2 FWI

iv Laminar FWI - catalytic wall using IFHC

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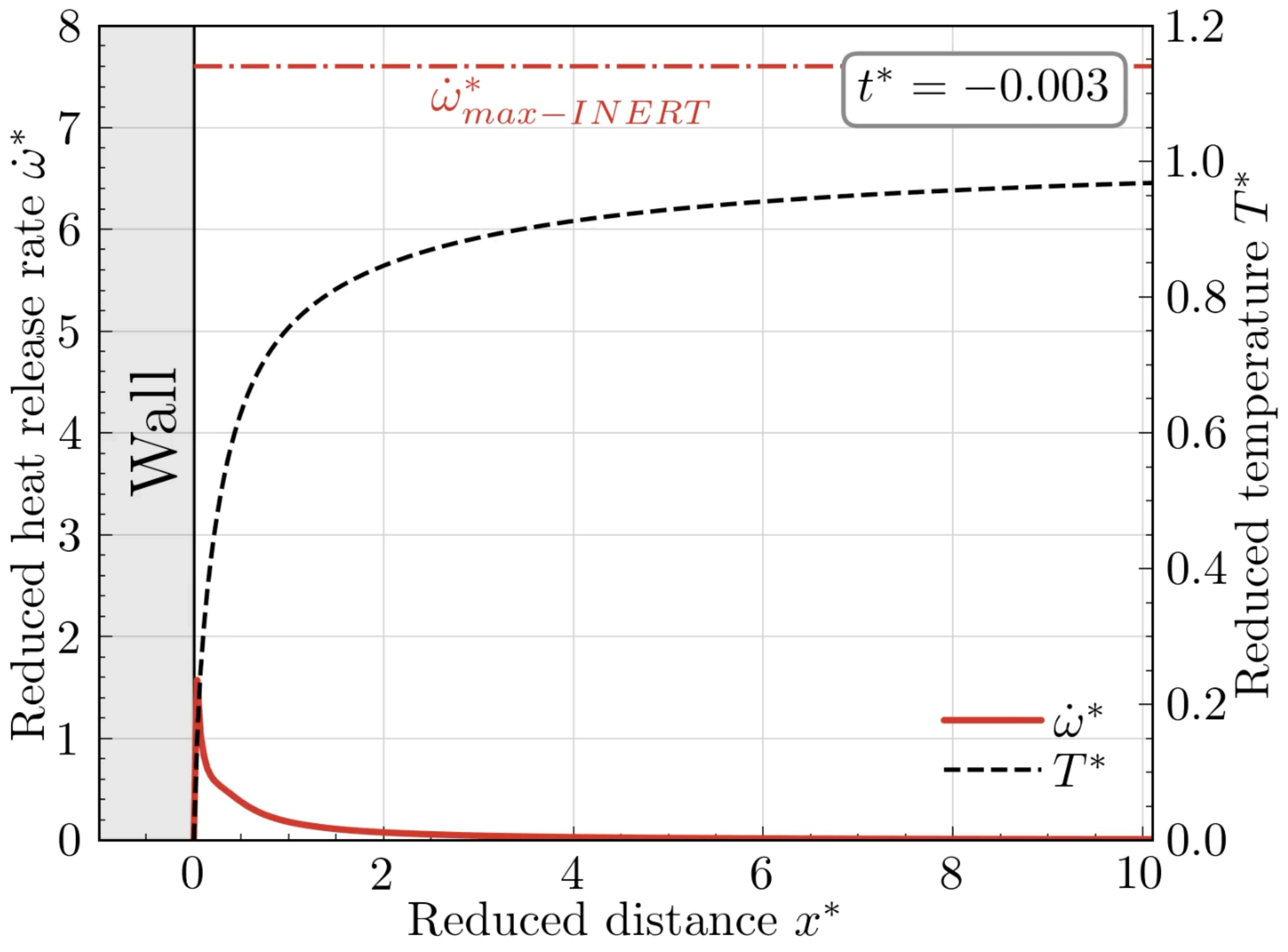
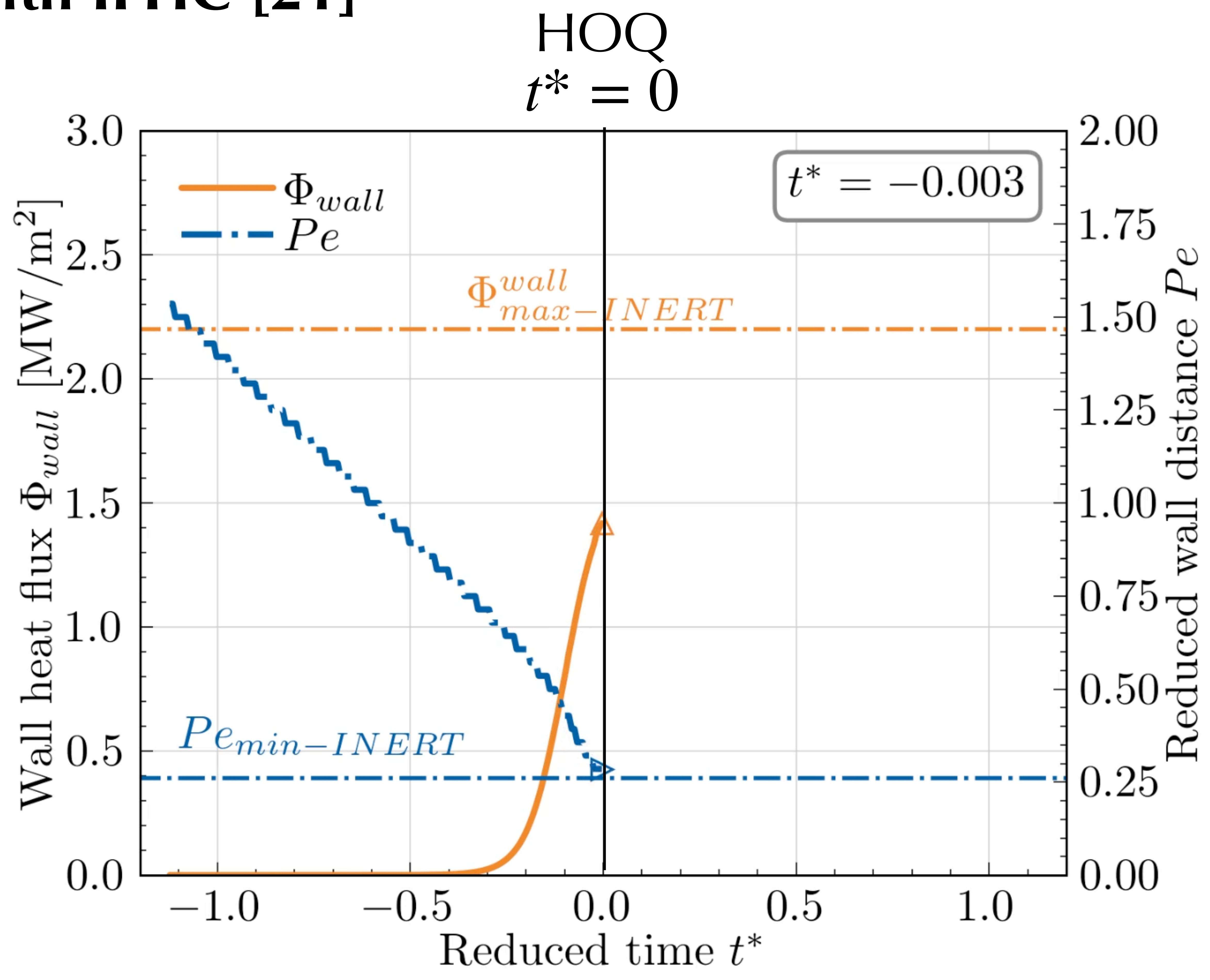


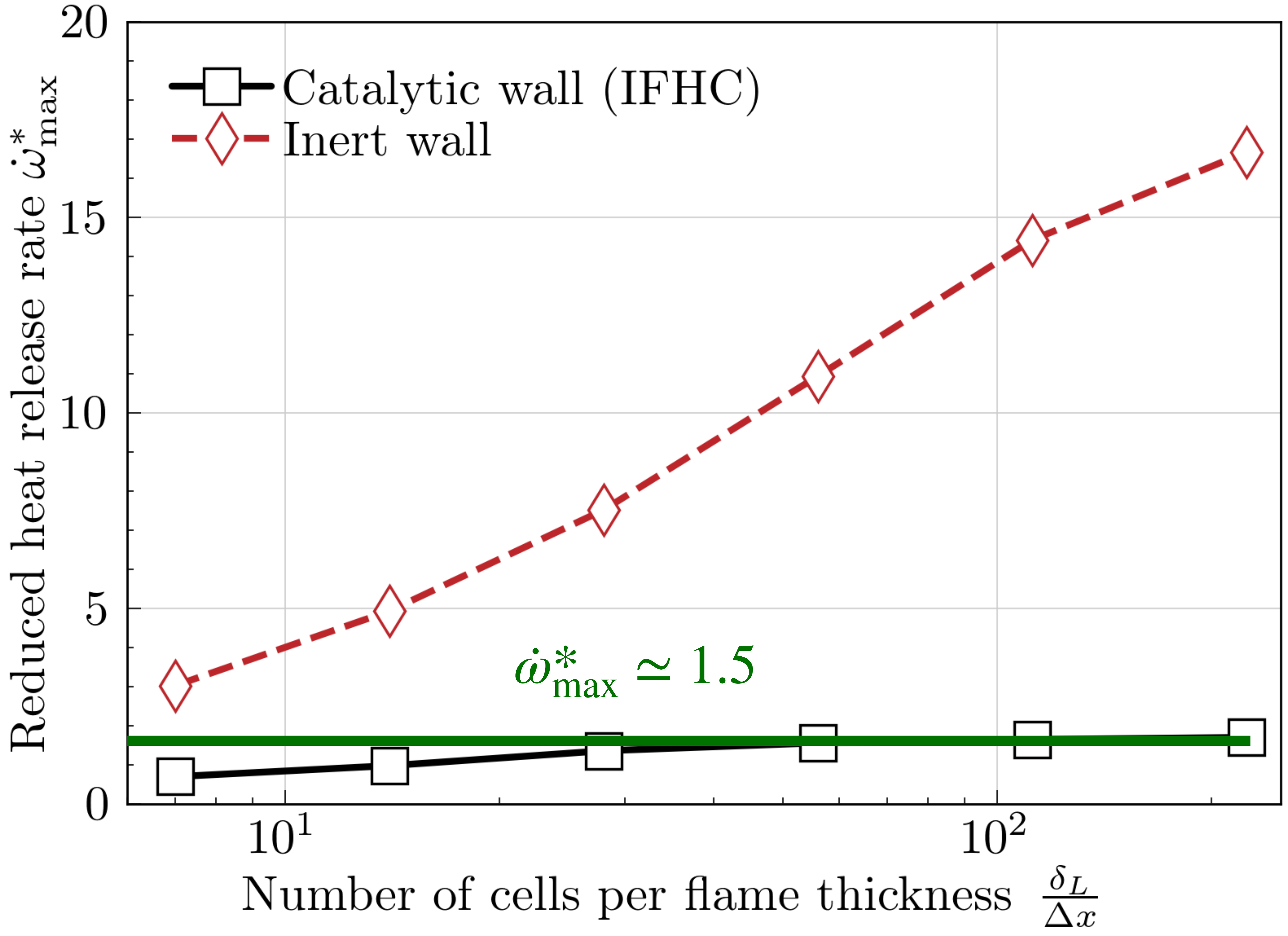


[21] De Nardi et al. (CNF, 2024)

IFHC model for H_2 FWI

iv Laminar FWI - catalytic wall using IFHC

▸ **Results for an isothermal wall with IFHC [21]**



- Effect of grid resolution with IFHC: $\delta_L = 7\Delta x - 224\Delta x$
- $\dot{\omega}^*$ peak value controlled with IFHC

→ Grid convergence retrieved at $\delta_L = 28\Delta x$

[21] De Nardi et al. (CNF, 2024)

Adaptation of TFLES

Thickened Flame model: TFLES

Key parameters

- Balance equation for the mass fraction of species k

$$\frac{\partial \rho Y_k}{\partial t} + \nabla \cdot \left(\rho \mathbf{u} Y_k\right) = \nabla \cdot \left(\rho D_k \nabla Y_k\right) + \dot{\omega}_k$$

- Balance equation for the mass fraction of species k in the **TFLES model**

$$\frac{\partial \rho Y_k}{\partial t} + \nabla \cdot \left(\rho \mathbf{u} Y_k\right) = \nabla \cdot \left(\rho D F E \nabla Y_k\right) + \frac{E}{F}\dot{\omega}_k$$

$F \equiv$ Thickening factor

- The larger F is, the less points we need in the flame front
- Thickened flame thermal thickness:

$$\delta_L^1 = F\delta_L^0$$

$E \equiv$ Efficiency function

- Damkohler number: $\mathrm{Da}^1 = \mathrm{Da}^0/F$
- Karlovitz number: $\mathrm{Ka}^1 = \mathrm{Ka}^0/F$
- Flame front wrinkles less in the presence of turbulence
- E compensate this loss

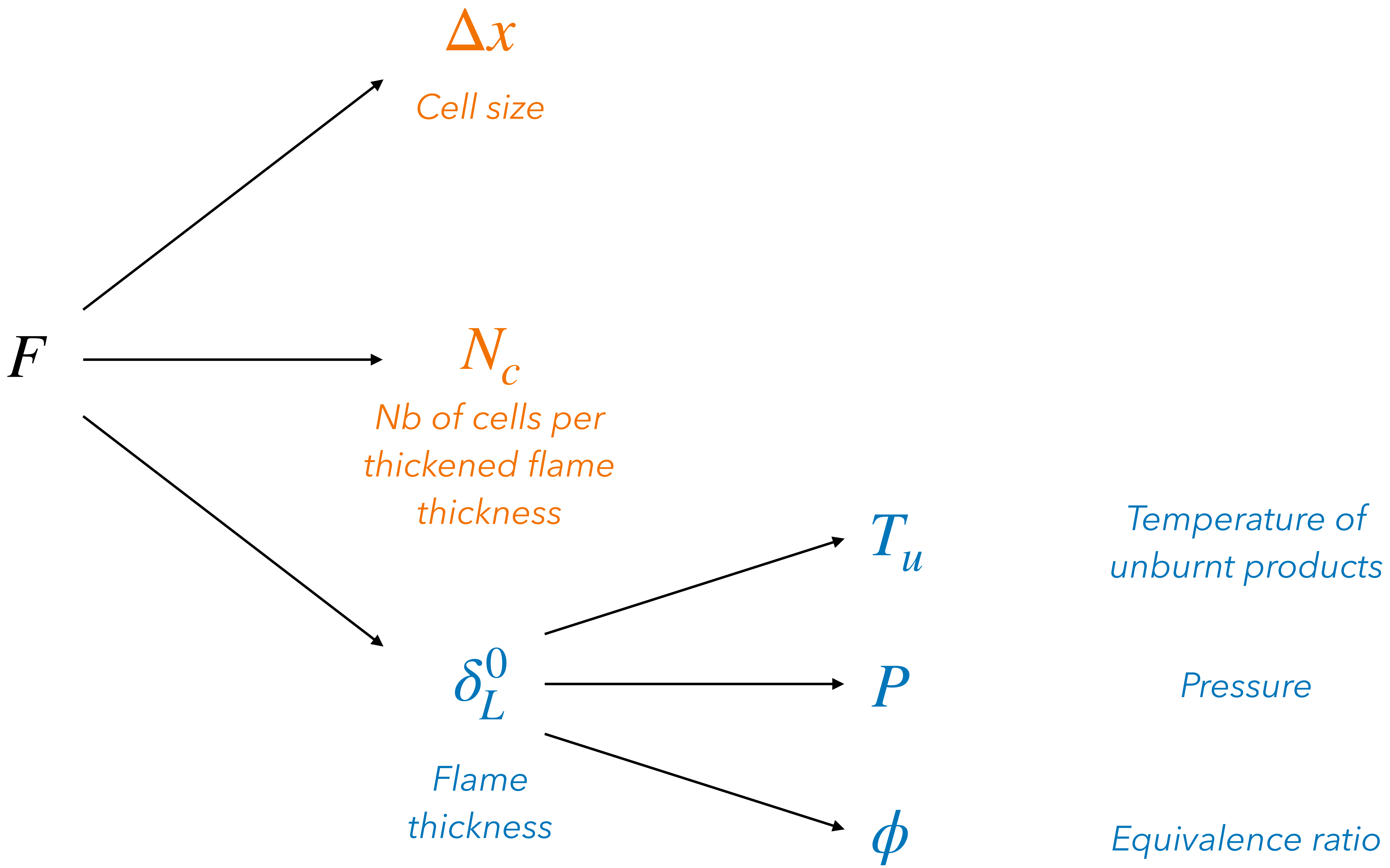
[1] Colin et al., Phys. Fluids, 2000

Thickened Flame model: TFLES

Key parameters: dependancy of the thickening factor

Physical parameters

Model parameters



F

Δx

Cell size

N_c

Nb of cells per thickened flame thickness

δ_L^0

Flame thickness

T_u

Temperature of unburnt products

P

Pressure

ϕ

Equivalence ratio

$$\boxed{F\delta_L^0 = N_c \Delta x}$$

Dynamic thickening (DTFLES)

Thickened Flame model: TFLES

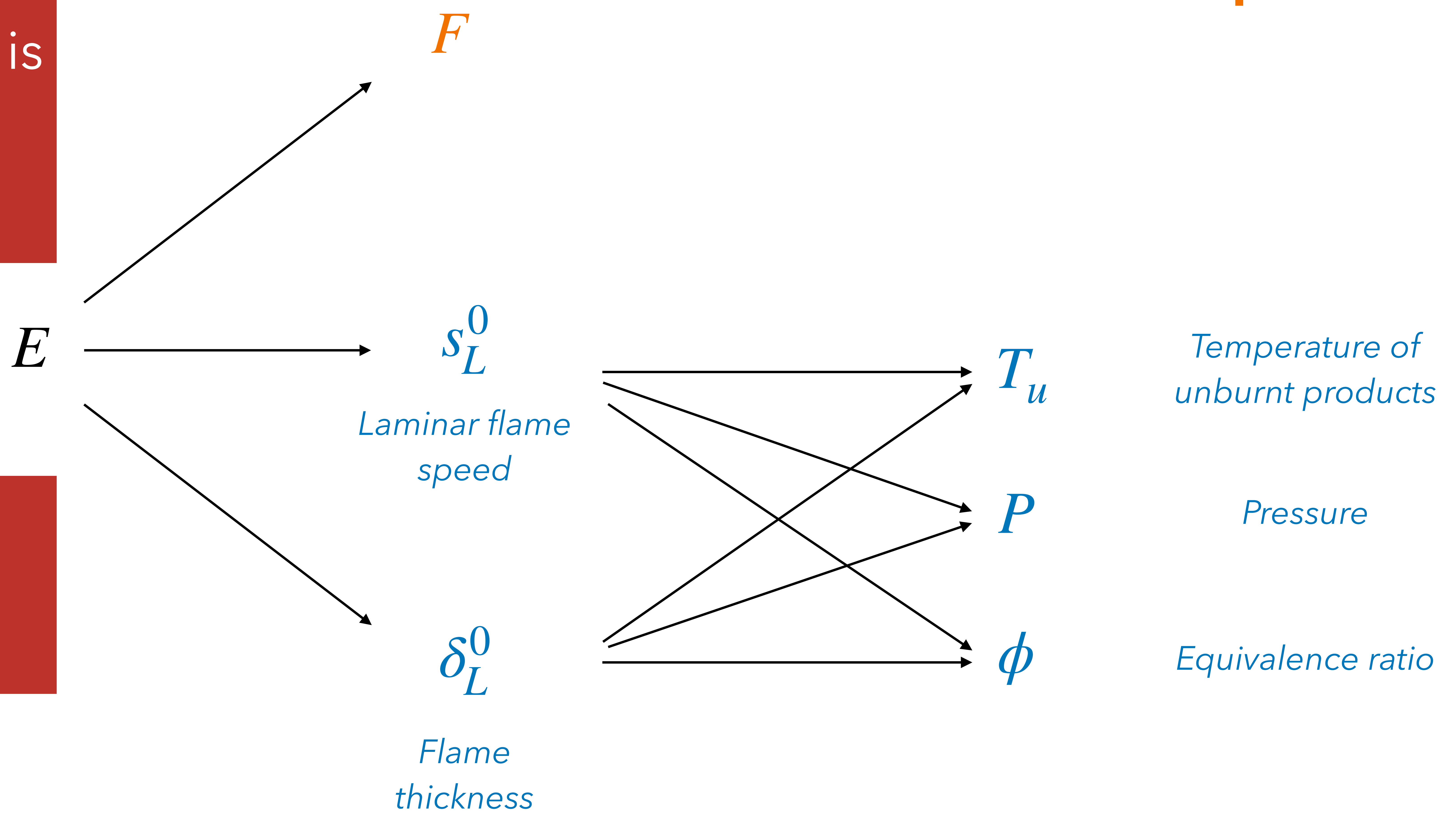
Key parameters: dependancy of the efficiency function

Recovering the flame properties in the flame front is **key** to the modeling of thickened flames

We need:

(T_u, P, ϕ)

Typical model:
Charlette efficiency function



Physical parameters

Model parameters

E

F

s_L^0

Laminar flame speed

δ_L^0

Flame thickness

T_u *Temperature of unburnt products*

P *Pressure*

ϕ *Equivalence ratio*

---

[1] F. Charlette et al. A power-law wrinkling model for LES of premixed turbulent combustion: Part I-non-dynamic formulation and initial tests. *Combustion and Flame*, 131:159–180, 2002.

Thickened Flame model: TFLES

Equivalence ratio based on Bilger mixture fraction

Passive scalar constructed from elemental mass fractions (Z_p).

Y_F and Y_{Ox} vary across the flame due to decomposition; Z_C, Z_H, and Z_O do not.

$$Z_p = \sum_{k=1}^{N} \frac{a_{p,k} W_p}{W_k} Y_k$$

W_k → Mol. weight of species k

Y_k → Mass fraction of species k

W_p → Mol. weight of element p

$a_{p,k}$ → No. of p element atoms in species k

$$\beta = \sum_{p=1}^{P} \gamma_p Z_p$$

Linear combination so that $\beta_{st} = 0$.

$$\xi = \frac{\beta - \beta_{Oxid}}{\beta_{Fuel} - \beta_{Oxid}}$$

Normalization between the two bounding cases.

These operations require the definition of the oxidizer composition and the fuel composition.

$$\Phi = \frac{\xi}{(1 - \xi)} * \frac{1 - \xi_{st}}{\xi_{st}}$$

Eq. ratio from mixture fraction.

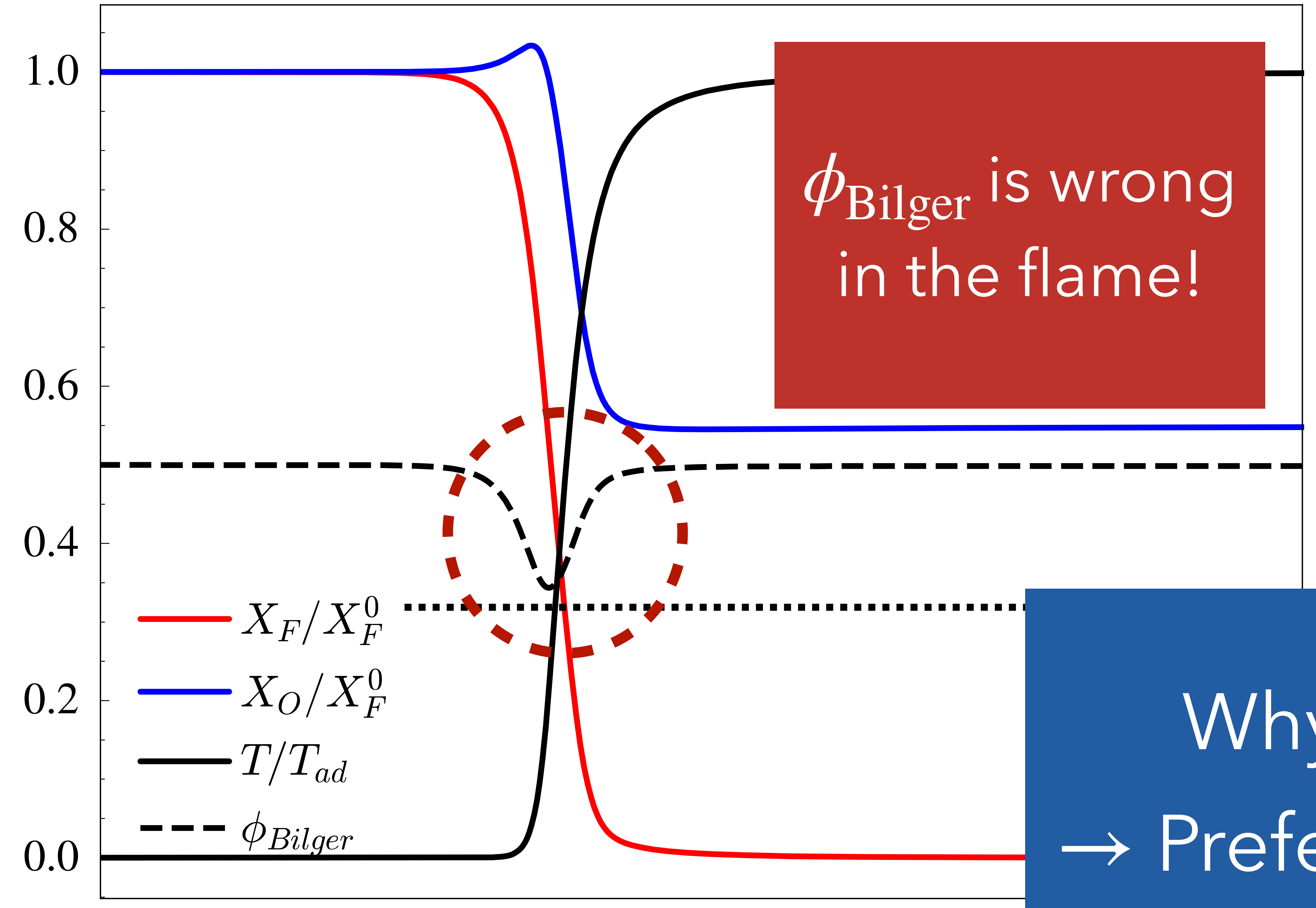
Classical formulation to retrieve the equivalence ratio

TNC. Chapter 3 Section 3.6.2

Thickened Flame model: TFLES

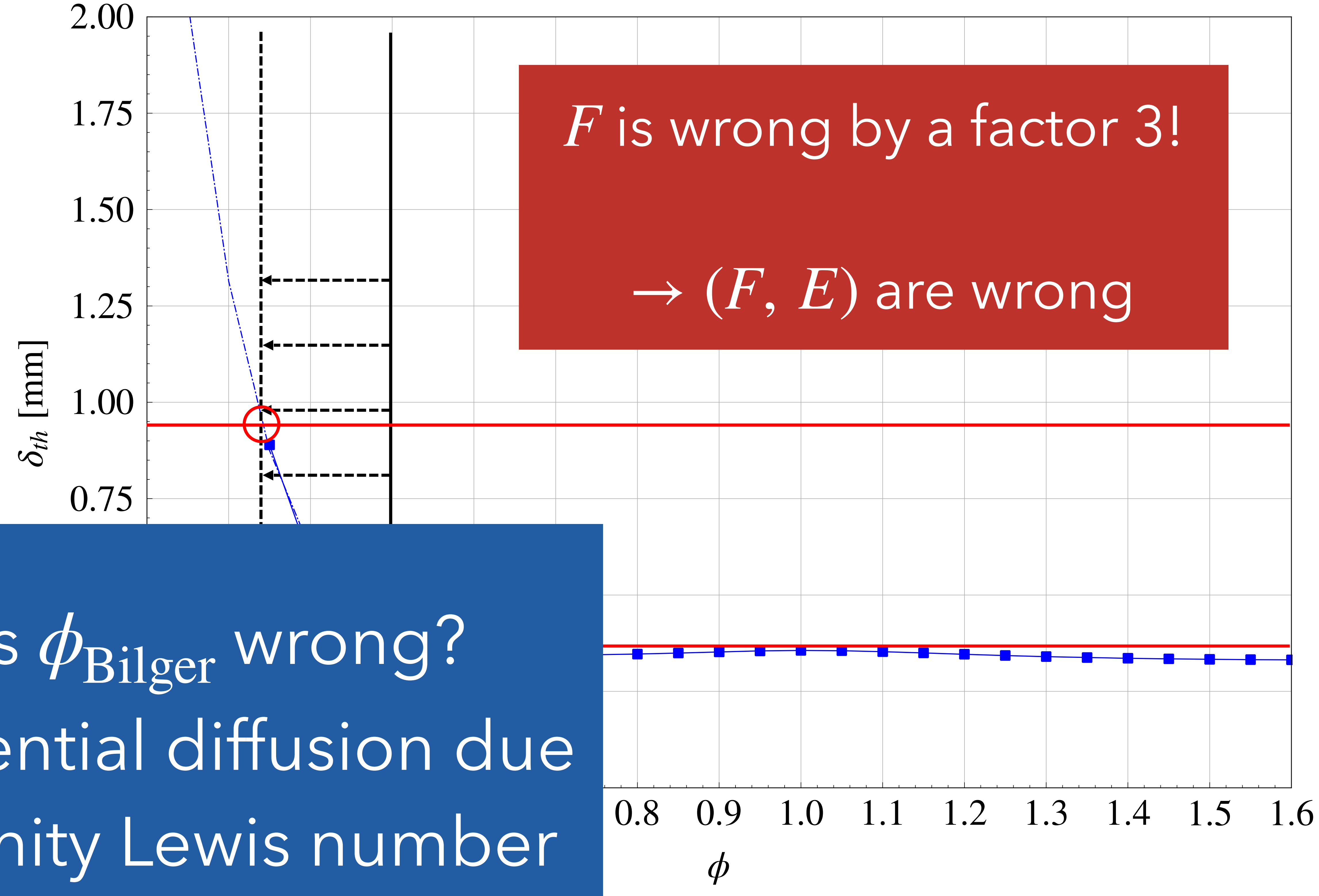
Equivalence ratio based on Bilger mixture fraction

Laminar flame thickness



ϕ_{Bilger} is wrong in the flame!

Lean H$_2$-Air Flame ($\phi = 0.5$, $T_u = 300$ K,



F is wrong by a factor 3!

→ $(F,\ E)$ are wrong

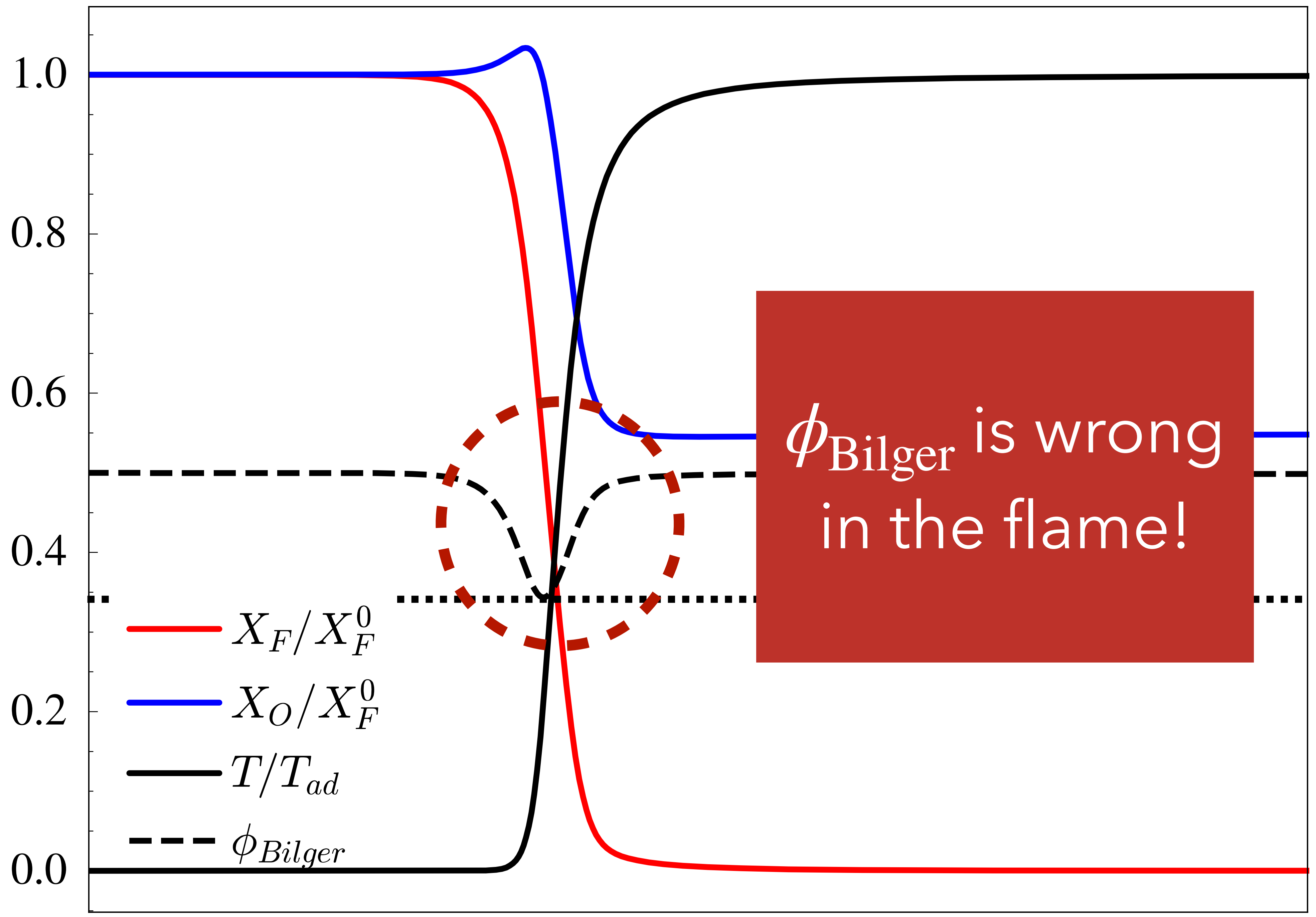
Why is ϕ_{Bilger} wrong?
→ Preferential diffusion due to non-unity Lewis number

[2] F. Charlette et al. A power-law wrinkling model for LES of premixed turbulent combustion: Part I-non-dynamic formulation and initial tests. *Combustion and Flame*, 131:159–180, 2002.

Thickened Flame model: TFLES

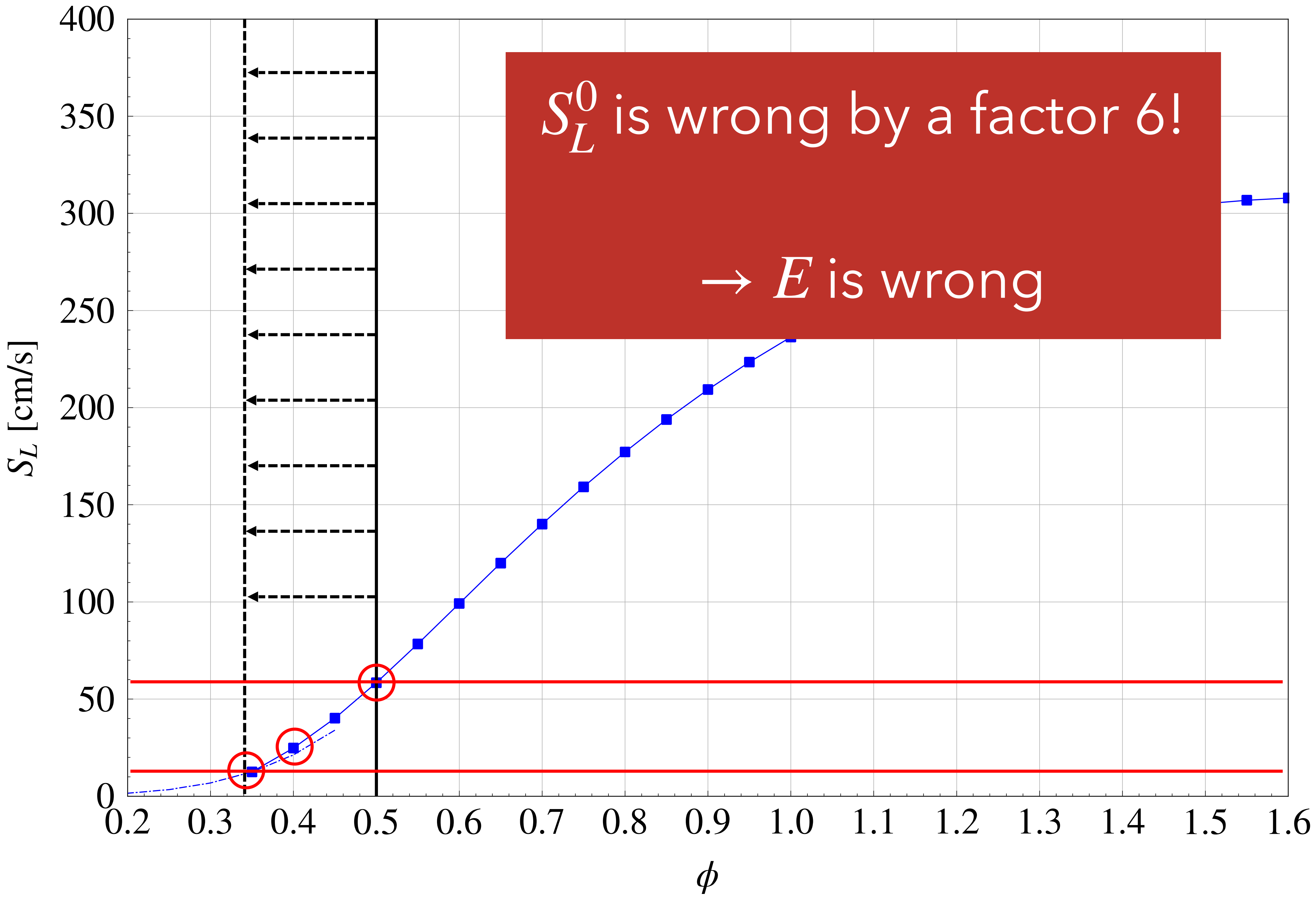
Equivalence ratio based on Bilger mixture fraction

Laminar flame speed



ϕ_{Bilger} is wrong in the flame!

Lean H_2-Air Flame ($\phi = 0.5$, $T_u = 300$ K, $P = 1$ bar)



S_L^0 is wrong by a factor 6!

→ E is wrong

[2] F. Charlette et al. A power-law wrinkling model for LES of premixed turbulent combustion: Part I-non-dynamic formulation and initial tests. *Combustion and Flame*, 131:159–180, 2002.

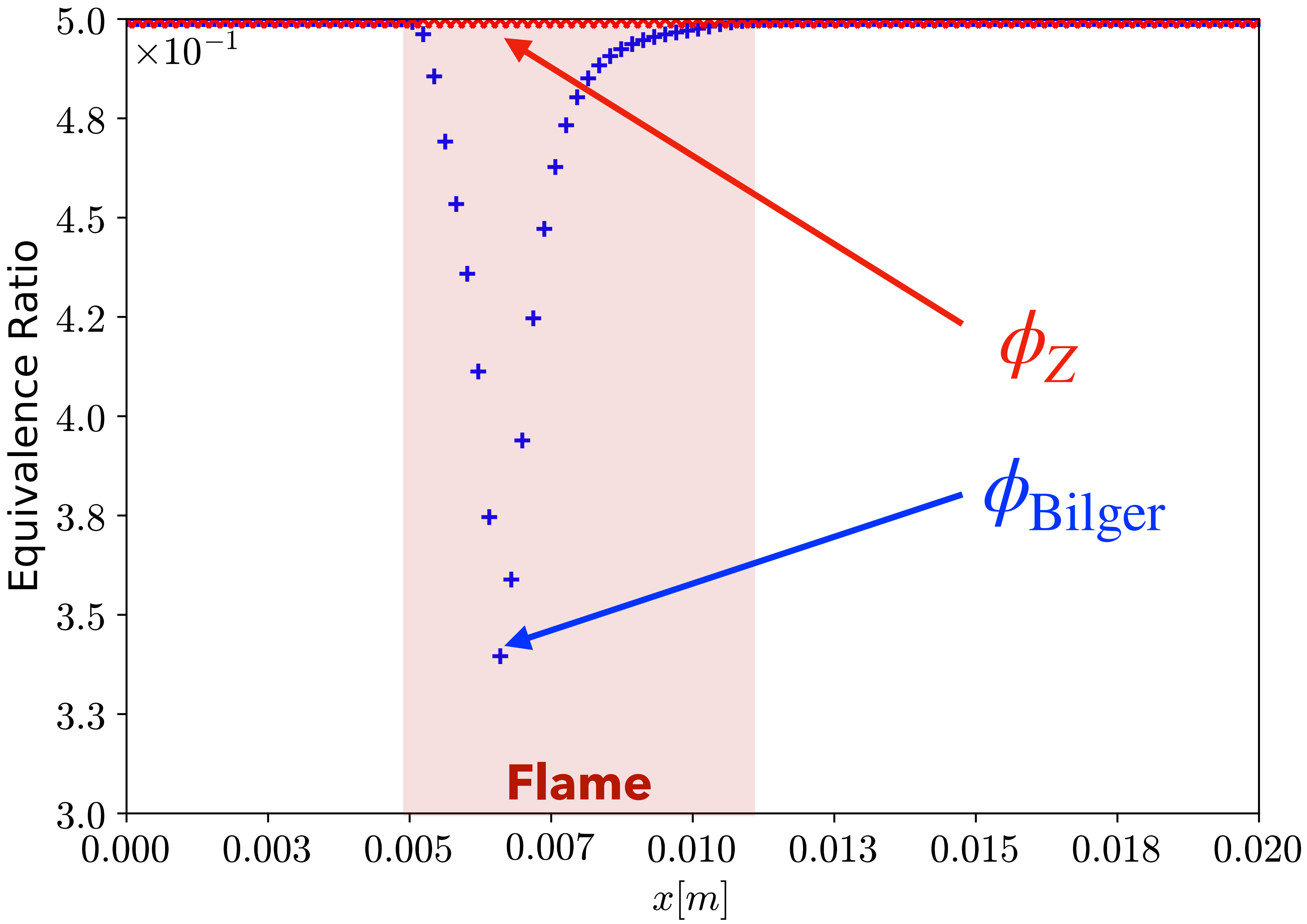
Thickened Flame model: TFLES

Solution: equivalence ratio based on a passive scalar

- Solving an additional transport equation for a passive scalar:

$$\frac{\partial \rho Z}{\partial t} + \frac{\partial}{\partial x_i}\left(\rho u_i Z\right) = \frac{\partial}{\partial x_i}\left(\rho D_Z \frac{\partial Z}{\partial x_i}\right)$$

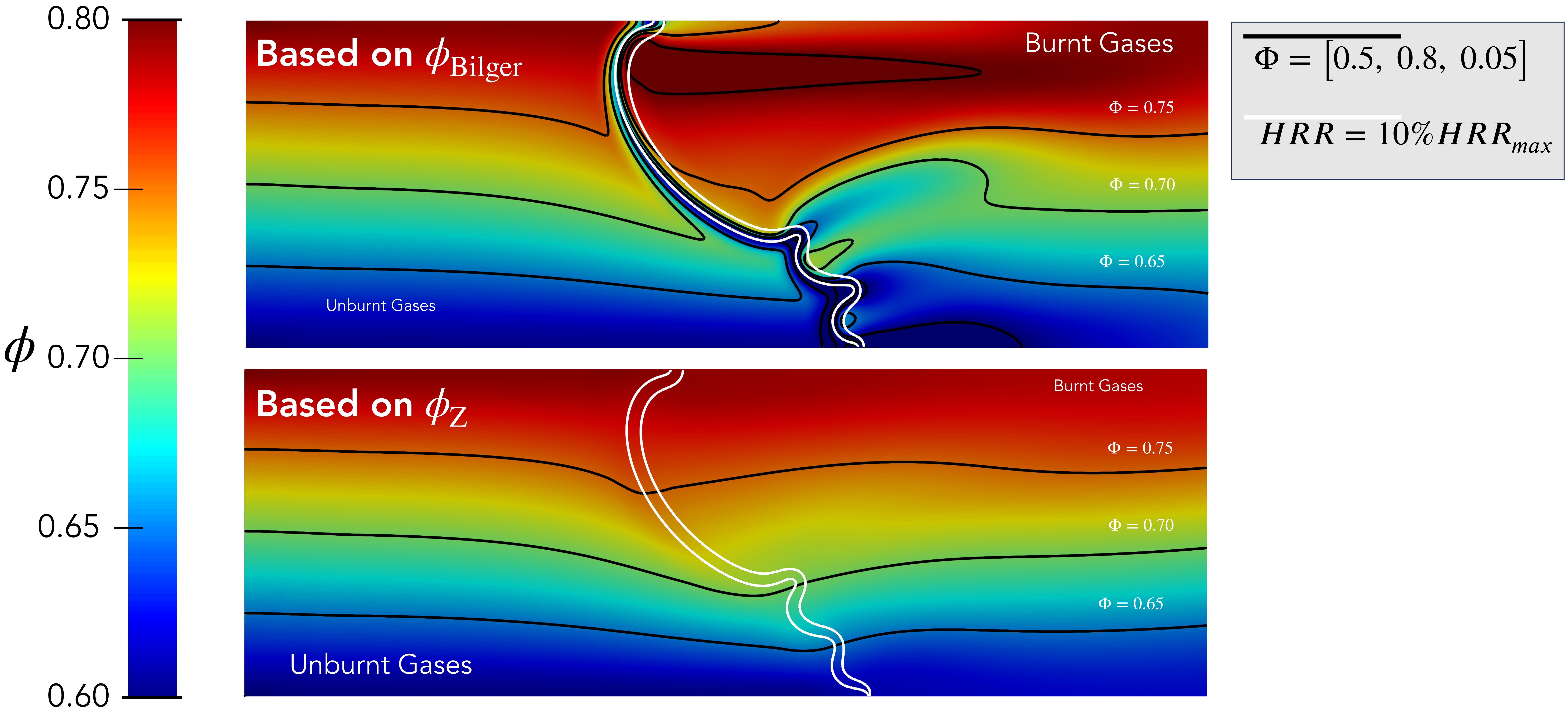
- Z is used (instead of Z_{Bilger}) to recover the equivalence ratio



Thickened Flame model: TFLES

Solution: equivalence ratio based on a passive scalar

2D stratified flame



$\Phi = [0.5,\ 0.8,\ 0.05]$

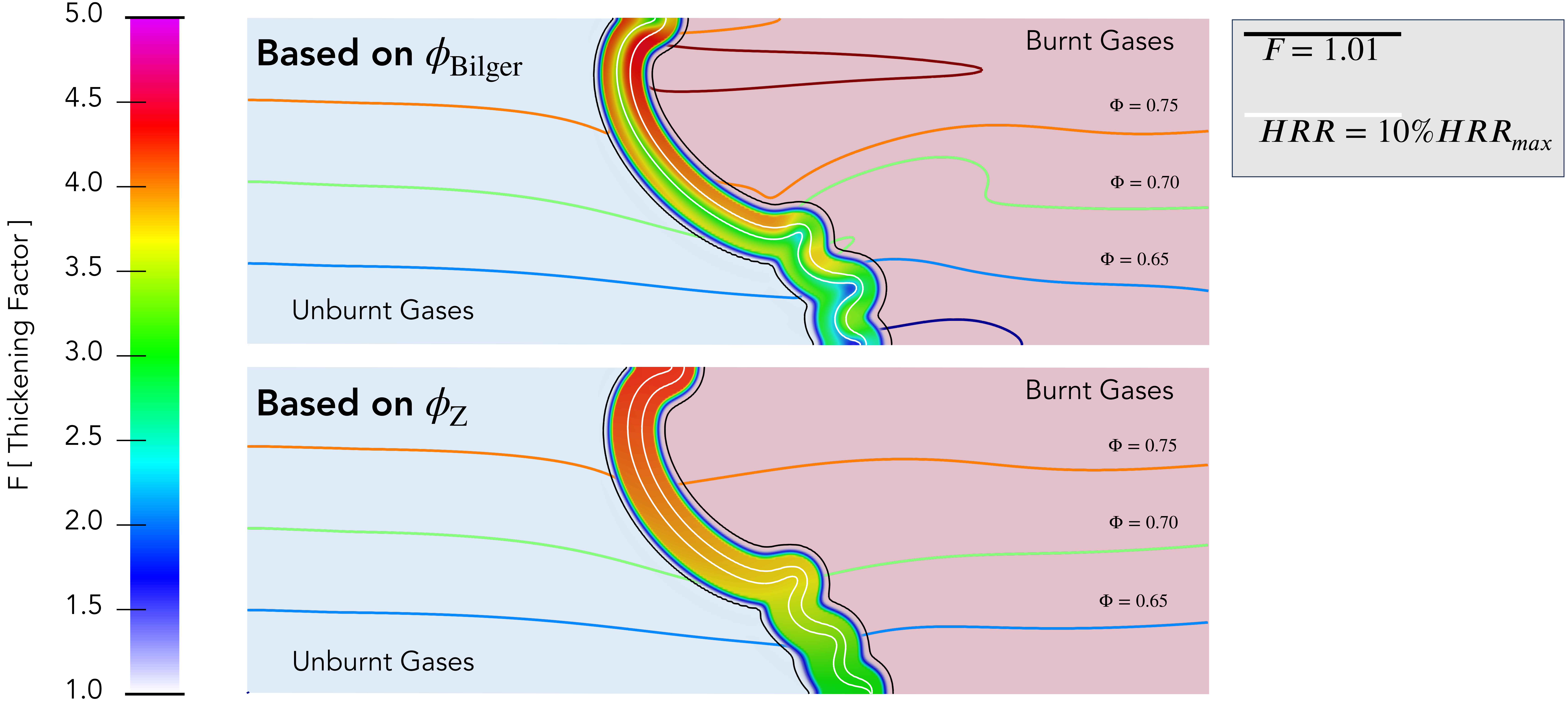
$HRR = 10\% HRR_{max}$

H. Vargas

Thickened Flame model: TFLES

Solution: equivalence ratio based on a passive scalar

2D stratified flame



Summary

What we discussed today

Chemistry

- ◉ Contrary to conventional fuel Detailed Chemistry is affordable for H_2

Transport properties

- Usual viscosity laws are wrong in pure H_2
  - ◉ Specific correction for H_2/air: Wilke's Law
    - ○ Variable Prandtl and Schmidt numbers to recover species and heat diffusion properties
- Mixture-averaged transport works for every mixtures → correct $\mu,\ D_k,\ \lambda$

Non-adiabatic walls

- Inert non-adiabatic walls are ill-posed
- Infinitely Fast Heterogenous Chemistry fixes this issue

Thickening Flame model

- Thickening factor F and efficiency function E are key parameters
- $F,\ E$ depend on model and physical parameters
  - ◉ $T_u,\ P,\ \phi$
- Bilger's mixture fraction is inaccurate in flame front
  - ◉ Need to add an additional transport equation
  - ◉ Accurate values of ϕ can then be recovered in the flame front

What we do not have time to discuss today

Other modeling aspects

Stretch response

- Consumption speed of a H_2 flames strongly affected by stretch (strain & curvature)
  - Classical TFLES does not properly model this
  - A model for the stretch response has been created for hydrocarbons [1] and H_2 [2]

Thermodiffusive (TD) instabilities

- Lean laminar H_2 flames are thermodiffusively unstable
  - A simple model (Aniello []) based on correlation of Berger [] is available in AVBP
  - WIP: Coupling TD models to corrected stretch response model gives promising results on laminar spherical flames [2]

Flame/turbulence interaction

- Charlette efficiency function used in most TFLES simulations
  - Based on the fractal hypothesis
  - Are H2/Air flame fractal?
    - If so, do they have similar fractal properties than other flames?
- What happens when lean flame become turbulent?
- Do turbulence kill the instabilities or are they still present at small scales?
- Are TD effects negligible compared to turbulent wrinkling ?
- Do they couple ?

Next talk!

Thank you for your attention

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