STUDY OF SOOT PRODUCTION IN ETHYLENE PYROLYSIS USING A SECTIONAL MODEL

24th ETH-Conference on Combustion Generated Nanoparticles
June 22-24, 2021 – Online Conference

Leonardo PACHANO
Damien AUBAGNAC-KARKAR

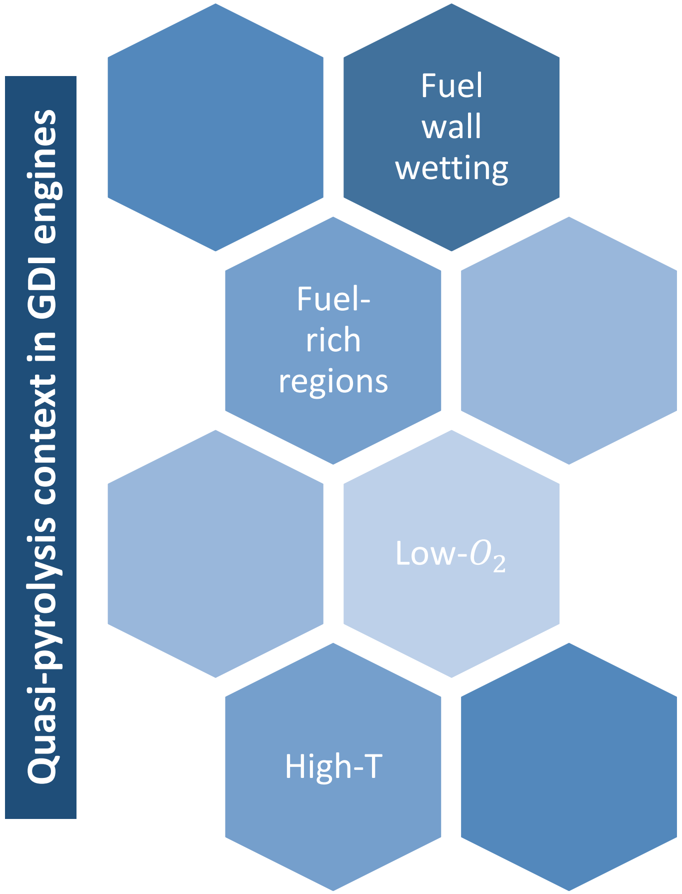
OUTLINE

- Background
- Objective and target conditions
- Soot modeling approach
- Results
- Summary

BACKGROUND

Nanoparticle formation from hydrocarbons oxidation and pyrolysis

- Of interest for the development of more efficient processes for nanoparticle synthesis
- Crucial for suppressing undesirable soot emissions that might impact the environment and human health



Video courtesy of Kamal Shway Shway
kamal.shway-shway@ifpen.fr

OUTLINE

- Background
- **Objective and target conditions**
- **Soot modeling approach**
- Results
- Summary

Modeling soot production in ethylene pyrolysis using a sectional approach

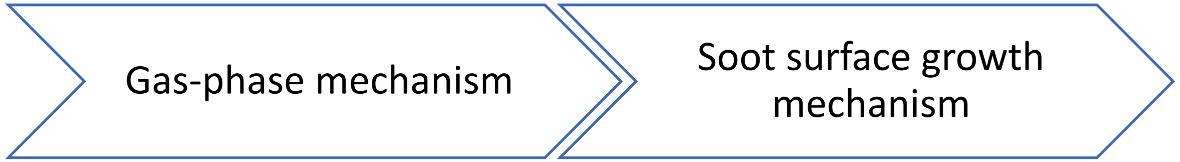
Long-term objective: Assessment in CFD pyrolyzing spray cases

- **PFR**: Soot concentration, PAH, ethylene and acetylene mole fraction at long residence times
- **Shock tube:** Time-resolved soot volume fraction and acetylene mole fraction

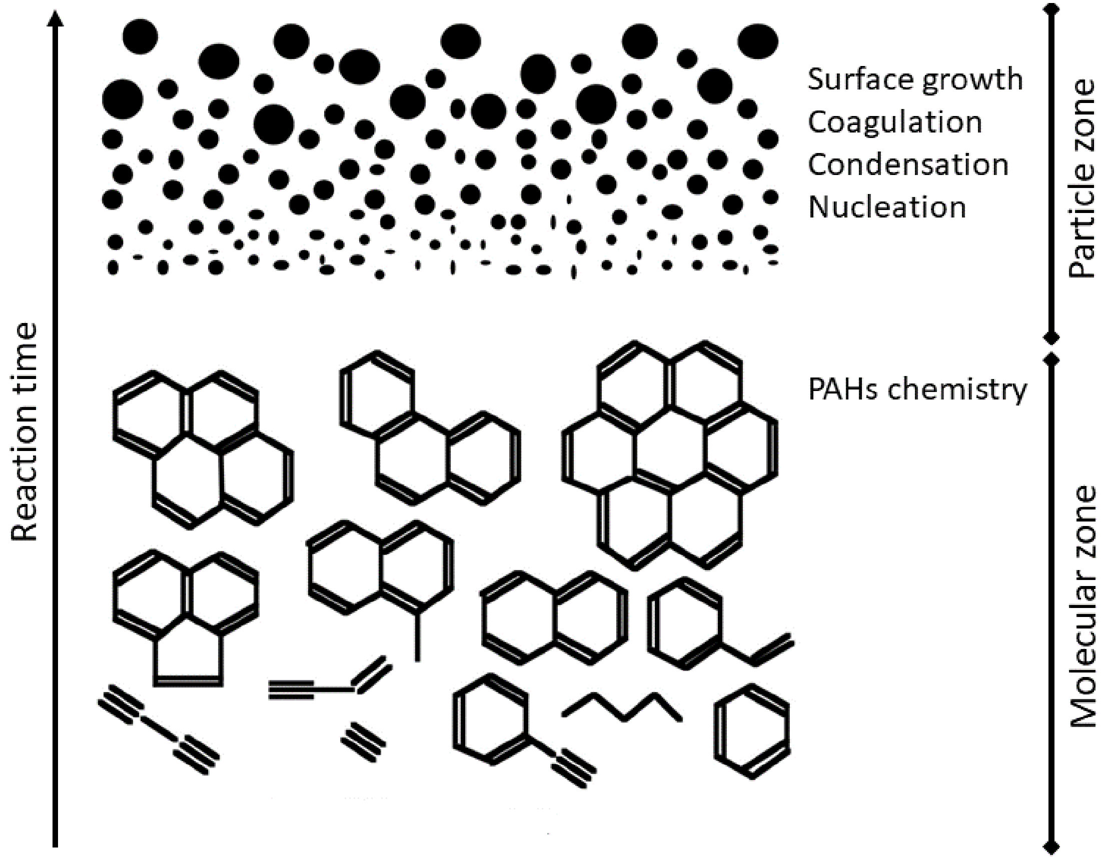
	PFR [1]	ST [2]
Experiment type	Plug flow reactor	Shock tube
X_{fuel}	0.03	0.0235
Diluent	Nitrogen	Argon
T $[K]$	1223-1423	2179
P $[Pa]$	$1.01x10^5$	$3.80x10^5$
Time $[ms]$	$\sim 10^3$	$\sim 10^1$

[1] Sánchez, Nazly E. Estudio de la formación de hidrocarburos aromáticos policíclicos (hap) en la pirólisis de acetileno y etileno. Diss. Universidad de Zaragoza, 2014.
[2] Utsav, K. C., Mohamed Beshir, and Aamir Farooq. "Simultaneous measurements of acetylene and soot during the pyrolysis of ethylene and benzene in a shock tube." Proceedings of the Combustion Institute 36.1 (2017): 833-840.

SOOT MODELING APPROACH



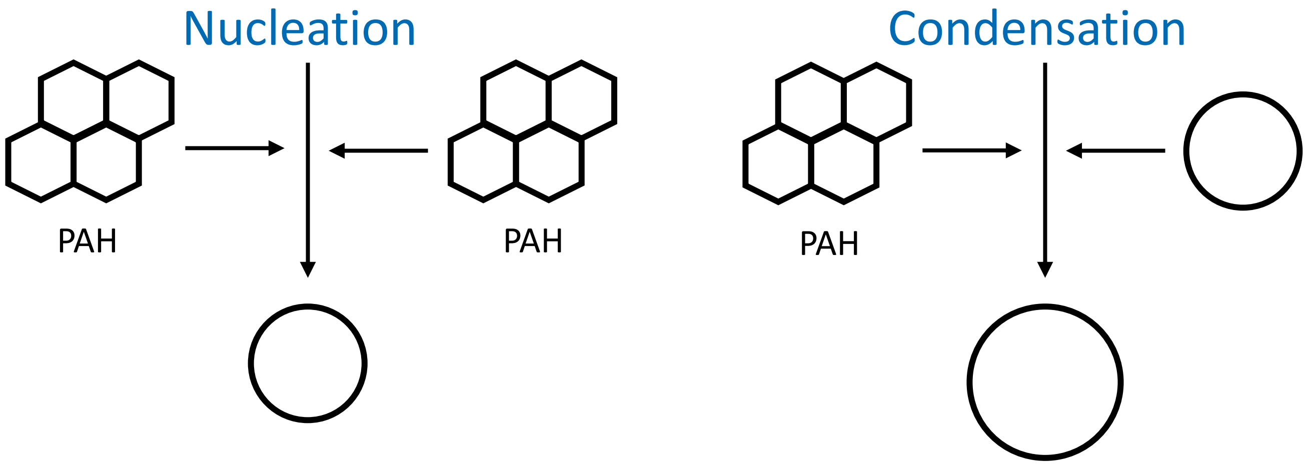
Pachano Prieto, L. M. (2020). CFD modeling of combustion and soot production in Diesel sprays (Doctoral dissertation).
Adapted from: J. Warnatz, U. Maas, and R. W. Dibble. Combustion. Vol. 4. Berlin: Springer, 2006



Zero-dimensional simulations using Cantera and a Sectional Soot Model (SSM) [1,2]

Modeled collisional phenomena and surface chemistry

Processes increasing soot mass



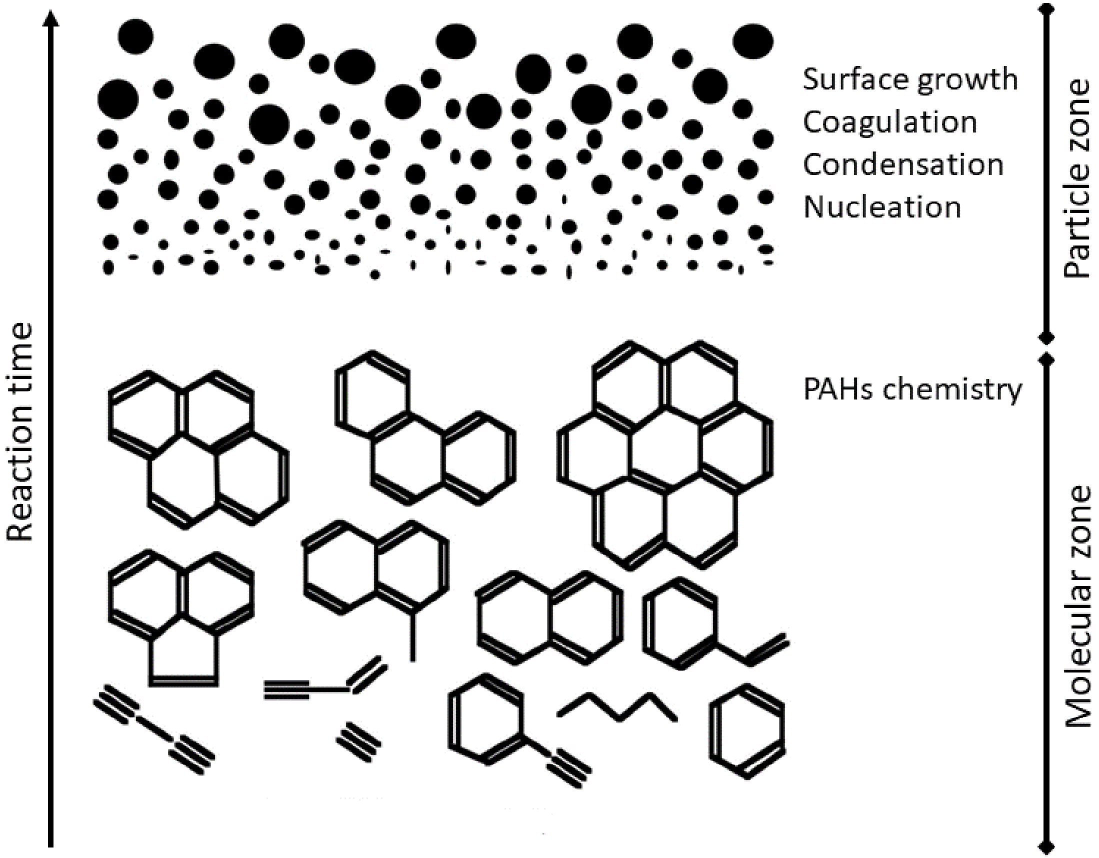
Reference numerical setup	
Gas phase coupling	Two-way
Nucleation model	Reversible with dampening factor
Condensation model	Reversible
Solver	Cantera 2.5.0a3

[1] Aubagnac-Karkar, D., Michel, J. B., Colin, O., Vervisch-Kljakic, P. E., & Darabiha, N. (2015). Sectional soot model coupled to tabulated chemistry for Diesel RANS simulations. Combustion and Flame, 162(8), 3081-3099.
[2] Aubagnac-Karkar, D., El Bakali, A., & Desgroux, P. (2018). Soot particles inception and PAH condensation modelling applied in a soot model utilizing a sectional method. Combustion and Flame, 189, 190-206.

SOOT MODELING APPROACH

Gas-phase mechanism → Soot surface growth mechanism

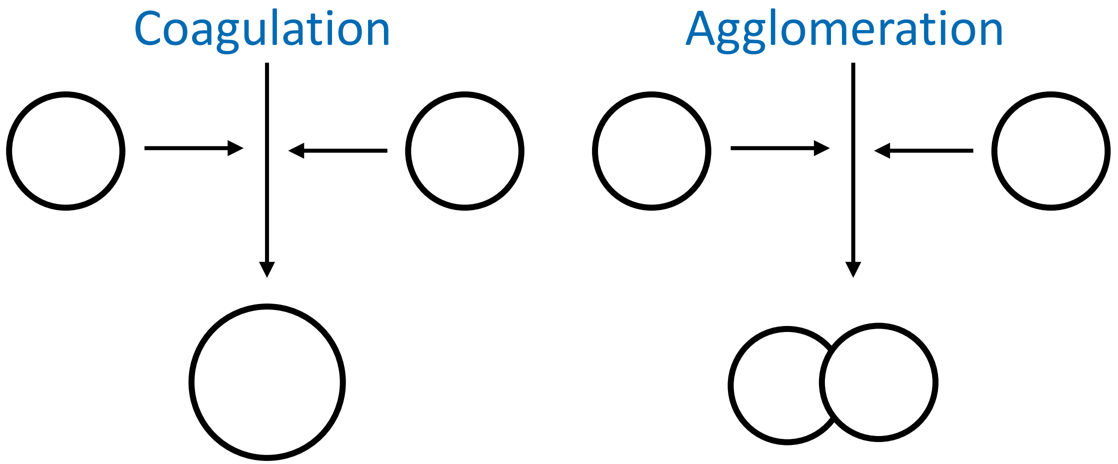
Pachano Prieto, L. M. (2020). CFD modeling of combustion and soot production in Diesel sprays (Doctoral dissertation).
Adapted from: J. Warnatz, U. Maas, and R. W. Dibble. Combustion. Vol. 4. Berlin: Springer, 2006



Zero-dimensional simulations using Cantera and a Sectional Soot Model (SSM) [1,2]

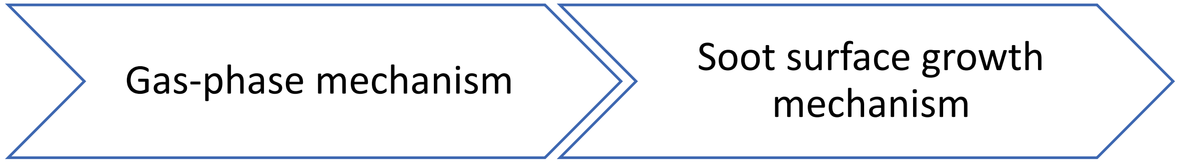
Modeled collisional phenomena and surface chemistry

Processes acting on the number of particles



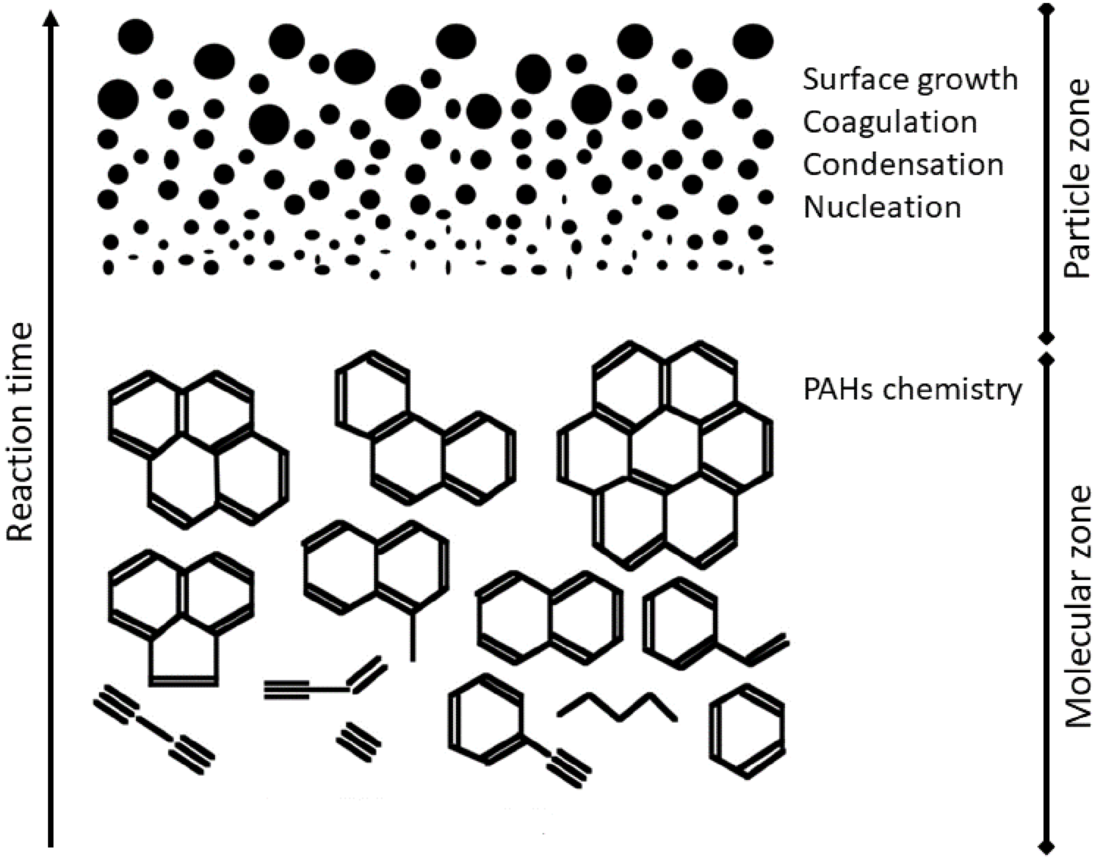
Reference numerical setup	
Gas phase coupling	Two-way
Nucleation model	Reversible with dampening factor
Condensation model	Reversible
Solver	Cantera 2.5.0a3

[1] Aubagnac-Karkar, D., Michel, J. B., Colin, O., Vervisch-Kljakic, P. E., & Darabiha, N. (2015). Sectional soot model coupled to tabulated chemistry for Diesel RANS simulations. Combustion and Flame, 162(8), 3081-3099.
[2] Aubagnac-Karkar, D., El Bakali, A., & Desgroux, P. (2018). Soot particles inception and PAH condensation modelling applied in a soot model utilizing a sectional method. Combustion and Flame, 189, 190-206.



SOOT MODELING APPROACH

Pachano Prieto, L. M. (2020). CFD modeling of combustion and soot production in Diesel sprays (Doctoral dissertation).
Adapted from: J. Warnatz, U. Maas, and R. W. Dibble. Combustion. Vol. 4. Berlin: Springer, 2006

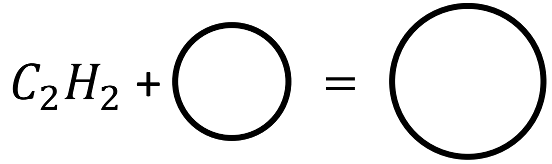


Zero-dimensional simulations using Cantera and a Sectional Soot Model (SSM) [1,2]

Modeled collisional phenomena and surface chemistry

Processes increasing soot mass

Surface growth



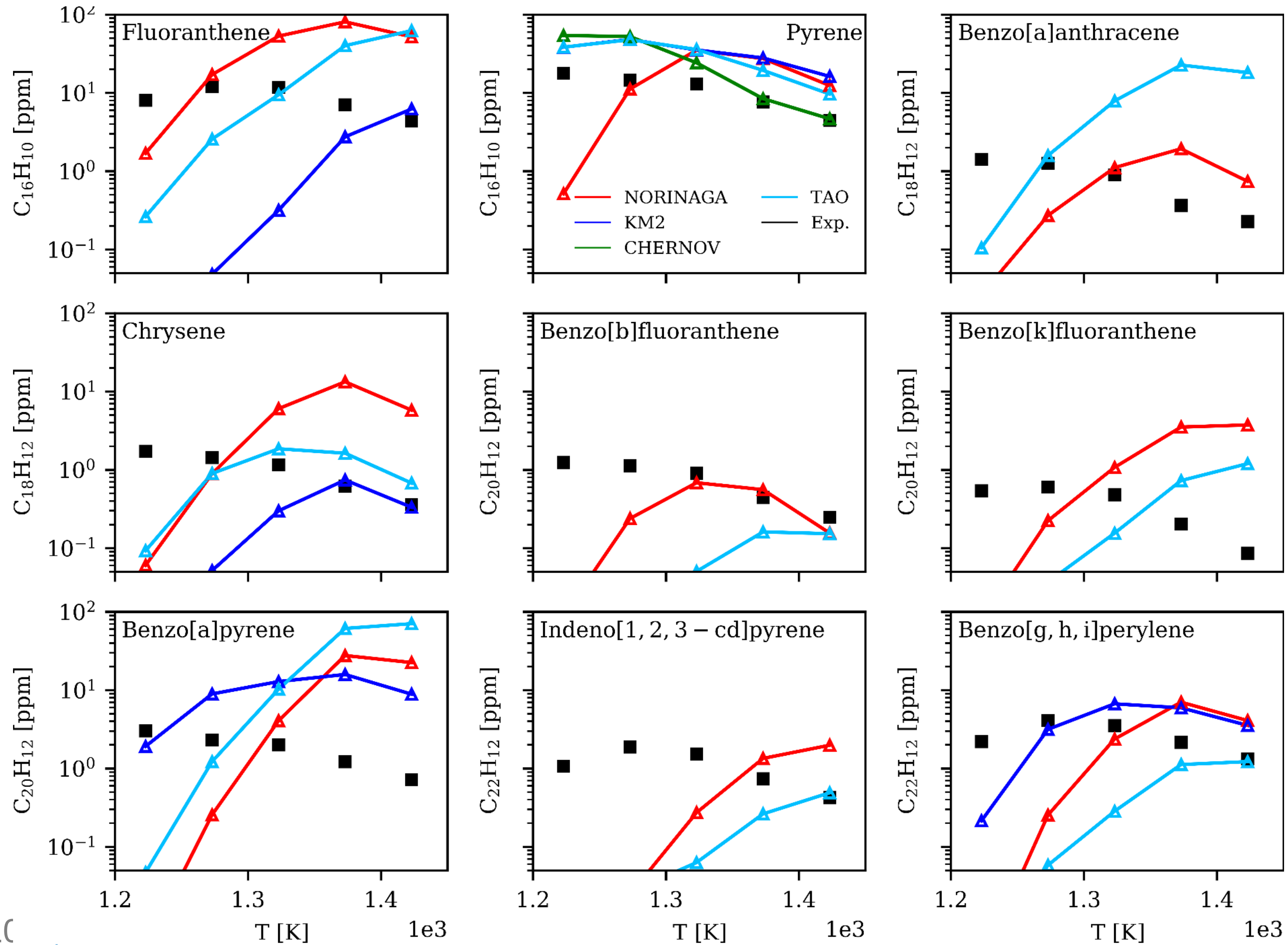
Reference numerical setup	
Gas phase coupling	Two-way
Nucleation model	Reversible with dampening factor
Condensation model	Reversible
Solver	Cantera 2.5.0a3

[1] Aubagnac-Karkar, D., Michel, J. B., Colin, O., Vervisch-Kljakic, P. E., & Darabiha, N. (2015). Sectional soot model coupled to tabulated chemistry for Diesel RANS simulations. Combustion and Flame, 162(8), 3081-3099.
[2] Aubagnac-Karkar, D., El Bakali, A., & Desgroux, P. (2018). Soot particles inception and PAH condensation modelling applied in a soot model utilizing a sectional method. Combustion and Flame, 189, 190-206.

OUTLINE

PFR RESULTS

Gas-phase mechanism → Soot surface growth mechanism



- PAH mole fraction results without soot modeling
- Choice of gas-phase mechanism inconclusive based on PAH results

Mechanism	Ref.
NORINAGA	[1]
KM2	[2]
CHERNOV	[3]
TAO	[4]

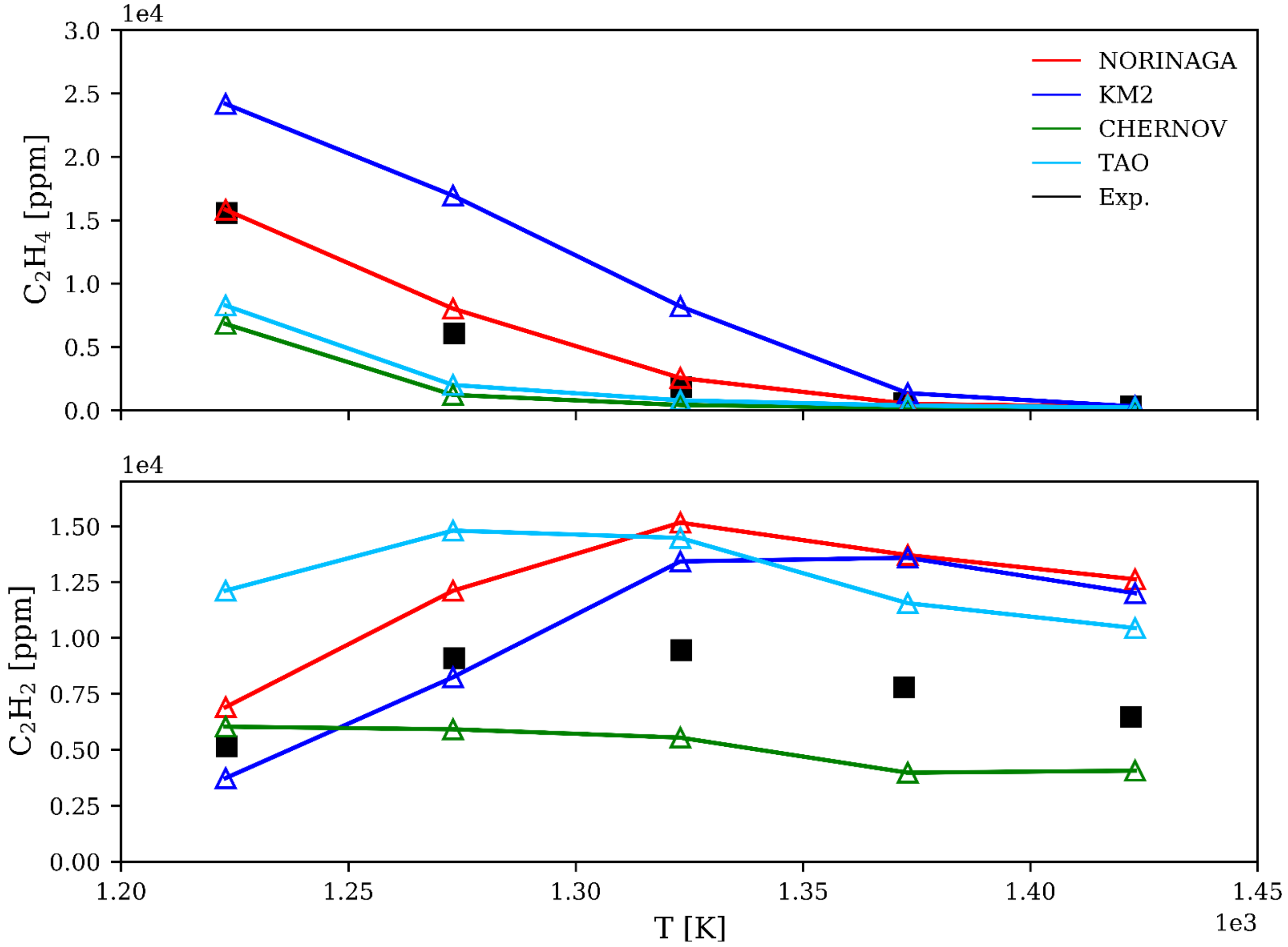
[1] Norinaga, Koyo, and Olaf Deutschmann. Industrial & engineering chemistry research 46.11 (2007): 3547-3557.
[2] Wang, Yu, Abhijeet Raj, and Suk Ho Chung. Combustion and flame 160.9 (2013): 1667-1676.
[3] Chernov, Victor, et al. Combustion and Flame 161.2 (2014): 592-601.
[4] Tao, Hairong, et al. Fuel 255 (2019): 115796.

Experimental results from:
Sánchez, Nazly E. Estudio de la formación de hidrocarburos aromáticos policíclicos (hap) en la pirólisis de acetileno y etileno. Diss. Universidad de Zaragoza, 2014.

PFR RESULTS

Gas-phase mechanism

Soot surface growth mechanism

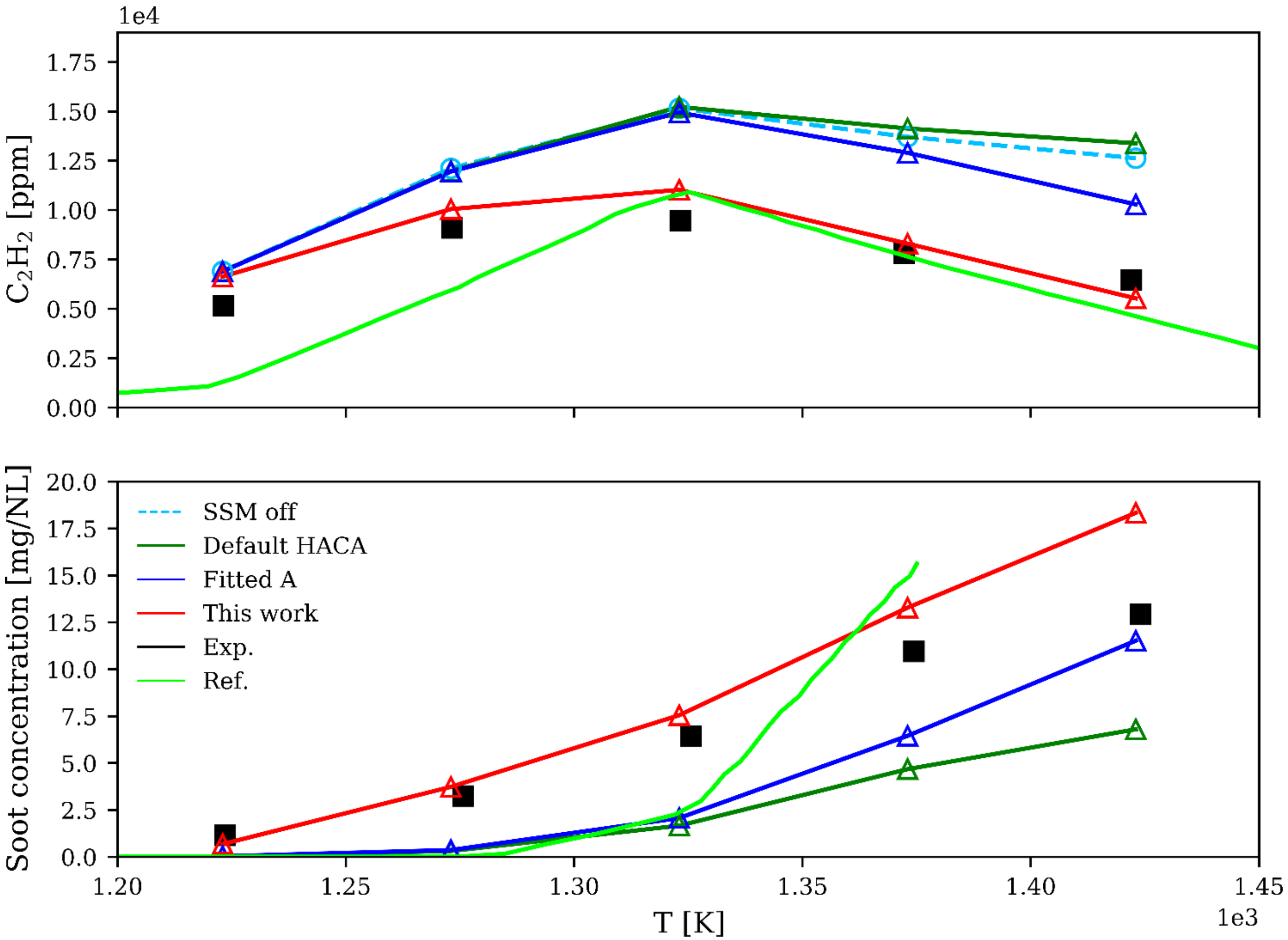


- Fuel and acetylene mole fraction results
- NORINAGA is the best compromise despite differences in PAH results

Experimental results from:
Sánchez, Nazly E. Estudio de la formación de hidrocarburos aromáticos policíclicos (hap) en la pirólisis de acetileno y etileno. Diss. Universidad de Zaragoza, 2014.

PFR RESULTS

Gas-phase mechanism → Soot surface growth mechanism



- Reference results without soot modeling
  - C_2H_2 overprediction in line with no soot modeling
- Setup 1: Starting point
  - Pyrene and all bigger PAHs used as precursors
  - Surface growth follows HACA mechanism [1]
- Setup 2 : Fitting C_2H_2 addition pre-exponential factor (A)
- Setup 3: Enhanced surface growth through C_2H, CH_3 and C_3H_3 [2]

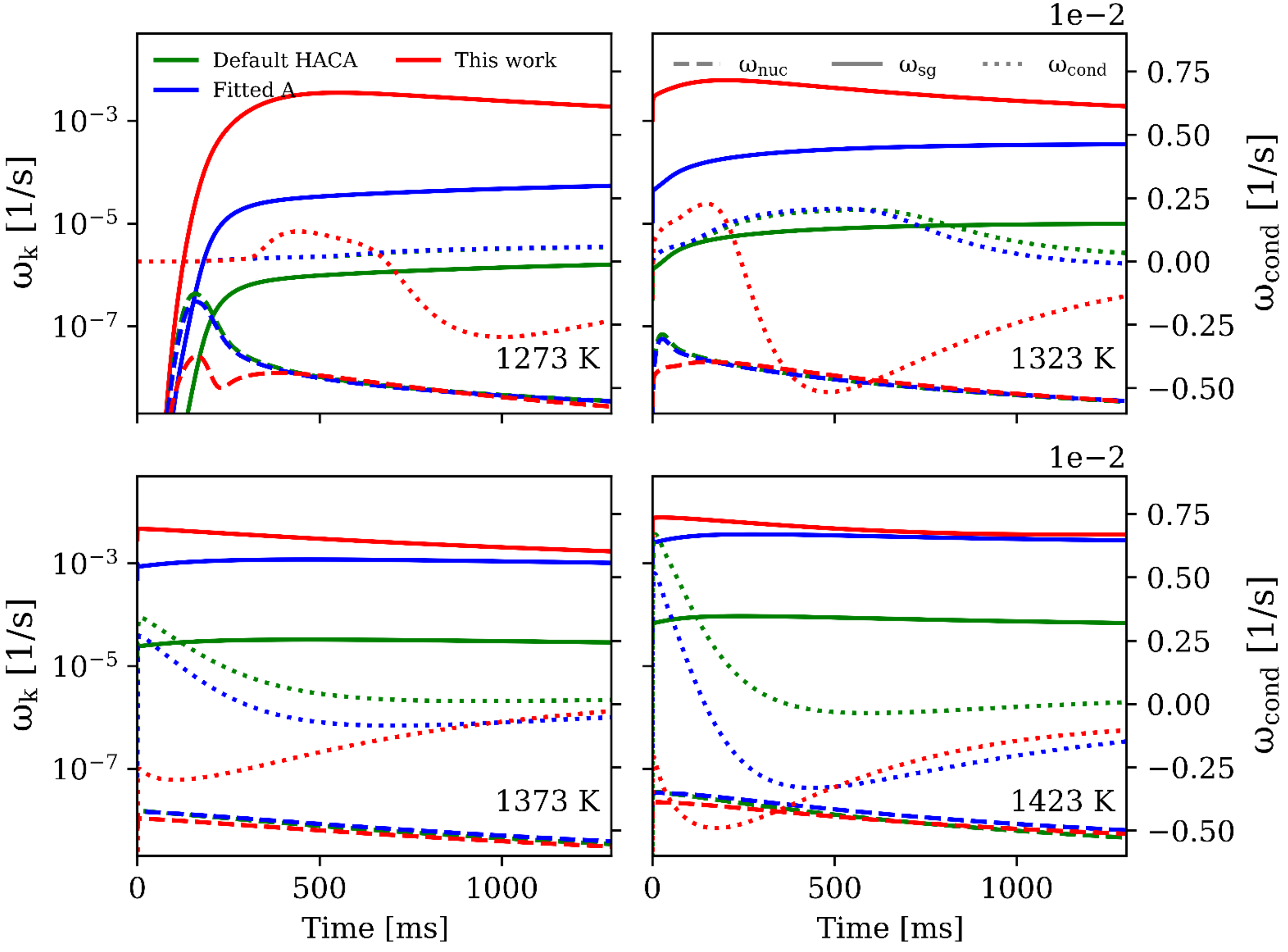
	Setup 1	Setup 2	Setup 3
A	80	2500	80
Surface growth mechanism	$C_{soot} + H \rightleftarrows C_{soot}^* + H_2$ $C_{soot}^* + H \rightarrow C_{soot}$ $C_{soot}^* + C_2H_2 \rightarrow C_{soot} + H$		HACA + $C_{soot} + C_2H \rightarrow C_{soot}^* + C_2H_2$ $C_{soot} + CH_3 \rightarrow C_{soot}^* + CH_4$ $C_{soot} + C_3H_3 \rightarrow C_{soot}^* + AC_3H_4$

[1] Appel, J., Bockhorn, H., & Frenklach, M. (2000). Kinetic modeling of soot formation with detailed chemistry and physics: laminar premixed flames of C2 hydrocarbons. Combustion and flame, 121(1-2), 122-136.
[2] Hwang, J. Y., & Chung, S. H. (2001). Growth of soot particles in counterflow diffusion flames of ethylene. Combustion and flame, 125(1-2), 752-762.
Experimental and modeling reference (Ref.) results from:
Sánchez, Nazly E. Estudio de la formación de hidrocarburos aromáticos policíclicos (hap) en la pirólisis de acetileno y etileno. Diss. Universidad de Zaragoza, 2014.

PFR RESULTS

Gas-phase mechanism

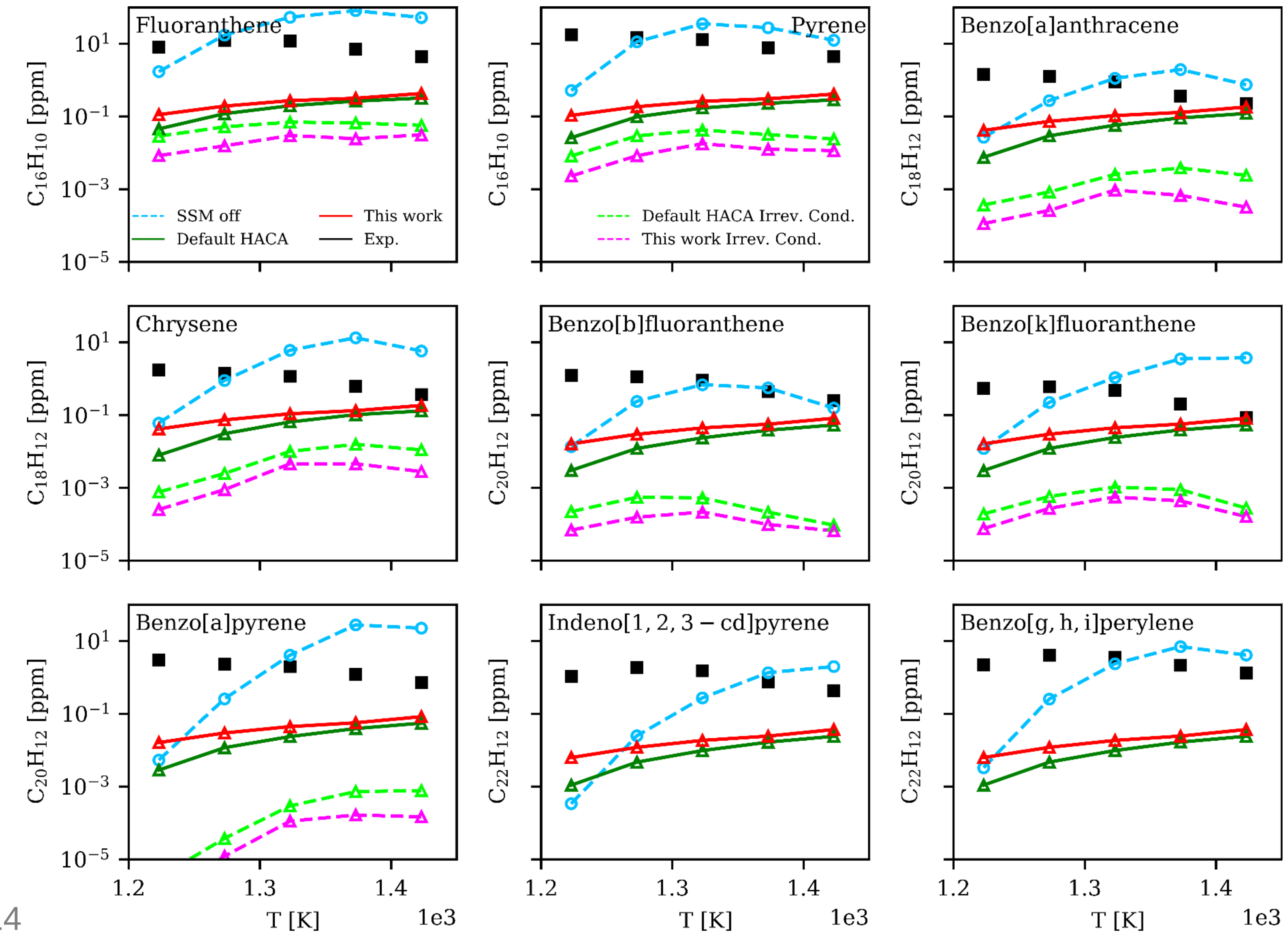
Soot surface growth mechanism



- Soot source terms for nucleation (ω_{nuc}), surface growth (ω_{sg}) and condensation (ω_{cond})
- Minimum ω_{sg} level required to induce a difference in the amount of soot produced
- Negligible effect on nucleation
- Increased relevance of condensation reversibility with the increase of surface growth

PFR RESULTS

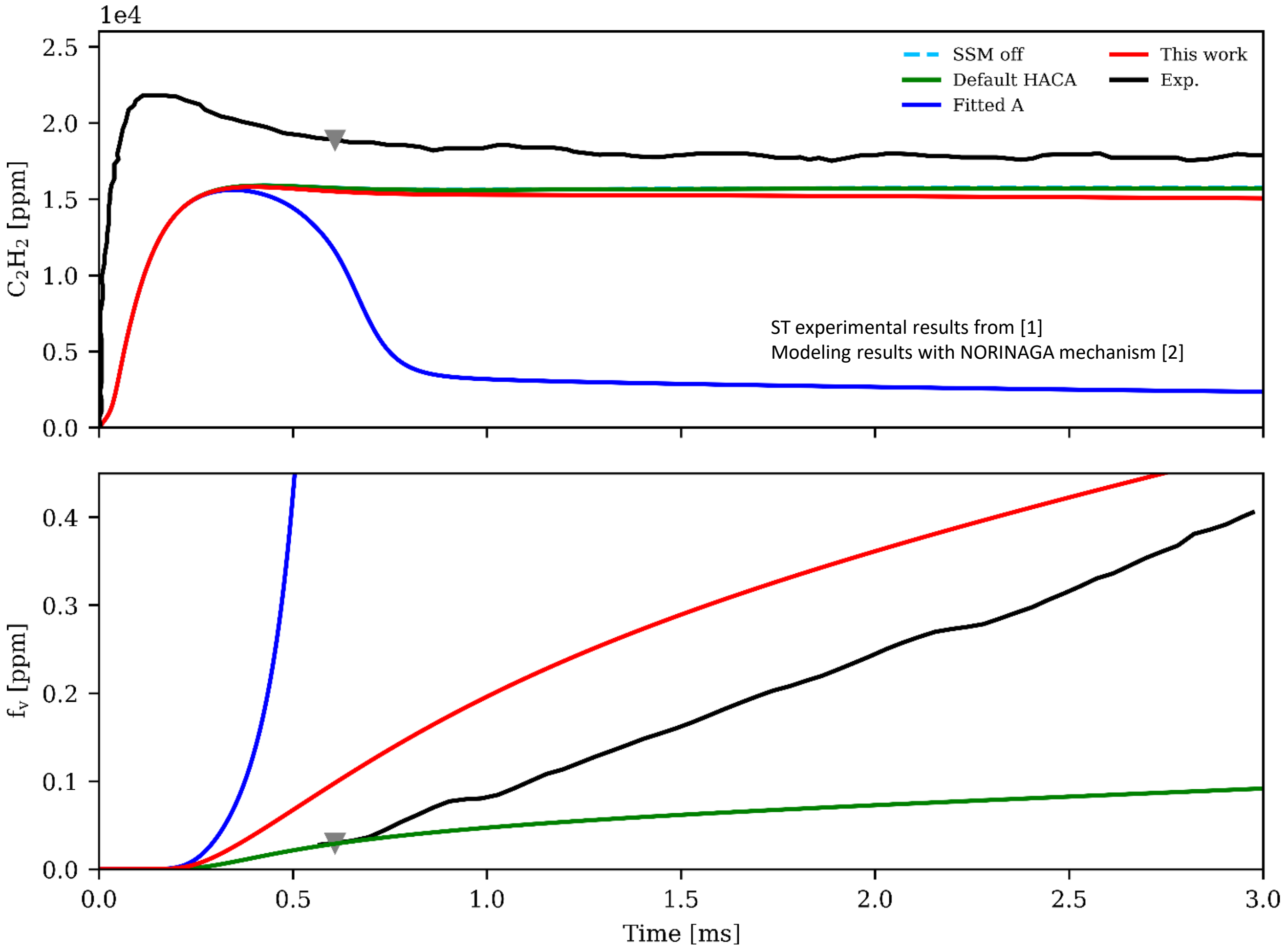
Gas-phase mechanism → Soot surface growth mechanism



- General lower PAH concentration when soot modelling is accounted for (as expected)
- Larger discrepancies with experimental data without condensation reversibility

ST RESULTS

Gas-phase mechanism → Soot surface growth mechanism



- Reference results without soot modeling
  - Slower initial increase in C_2H_2 mole fraction profile
- Setup 1: Soor volume fraction (f_v) under-prediction in line with PFR results
- Setup 2: Fitted A for PFR case over-estimates C_2H_2 addition
- Setup 3: Reasonable results despite under-prediction of soot onset

	Setup 1	Setup 2	Setup 3
A	80	2500	80
Surface growth mechanism	$C_{soot} + H \rightleftarrows C_{soot}{}^* + H_2$ $C_{soot}{}^* + H \rightarrow C_{soot}$ $C_{soot}{}^* + C_2H_2 \rightarrow C_{soot} + H$		HACA + $C_{soot} + C_2H \rightarrow C_{soot}{}^* + C_2H_2$ $C_{soot} + CH_3 \rightarrow C_{soot}{}^* + CH_4$ $C_{soot} + C_3H_3 \rightarrow C_{soot}{}^* + AC_3H_4$

Experimental results from:
Utsav, K. C., Mohamed Beshir, and Aamir Farooq. Proceedings of the Combustion Institute 36.1 (2017): 833-840.

IFP Energies nouvelles

OUTLINE

- Background
- Objective and target conditions
- Soot modeling approach
- Results
- **Summary**

SUMMARY

- PAH results were not conclusive for the choice of the gas-phase mechanisms → Assessment complemented by fuel and acetylene concentration predictions

- Acetylene and soot predictions emphasize the importance of capturing the carbon balance between soot and the gas-phase → Soot surface growth mechanism is crucial to that end

- Additional steps added to the HACA mechanism [1,2] proved to enhance results both in the PFR ($1.01x10^5$ Pa, $1223-1423\ K$) and ST ($3.80x10^5$ Pa, $2179\ K$) cases

- Condensation reversibility improve PAH agreement with experimental data although large discrepancies still remain

[1] Appel, J., Bockhorn, H., & Frenklach, M. (2000). Kinetic modeling of soot formation with detailed chemistry and physics: laminar premixed flames of C2 hydrocarbons. Combustion and flame, 121(1-2), 122-136.
[2] Hwang, J. Y., & Chung, S. H. (2001). Growth of soot particles in counterflow diffusion flames of ethylene. Combustion and flame, 125(1-2), 752-762.

Innovating for energy

Find us on:

www.ifpenergiesnouvelles.com

@IFPENinnovation