A THEORETICAL STUDY OF PAHS GROWTH BY PHENYLACETYLENE ADDITION

Zepeng Li[a,b], Peng Liu[a], Peng Zhang[b], Hong He[b], Suk Ho Chung[a], William L. Roberts[a]

a King Abdullah University of Science and Technology (KAUST), Clean Combustion Research Center, Saudi Arabia
b Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, China

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King Abdullah University of Science and Technology

Background

❑ Phenylacetylene ($C_6H_5C_2H$, C_8H_6, or A1C2H)

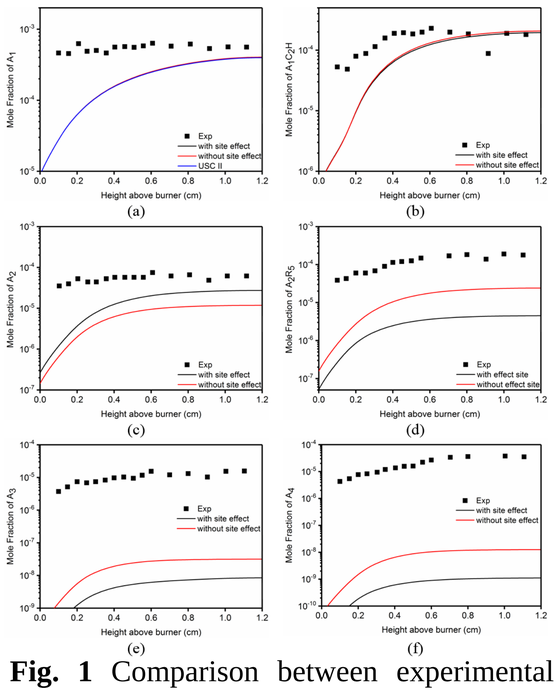


Fig. 1 Comparison between experimental and calculated mole fraction of benzene and various PAHs in C_2H_4 premixed flame [1]. (1 atm, $C_2H_4/O_2/N_2$ = 21.3/20.9/57.8).

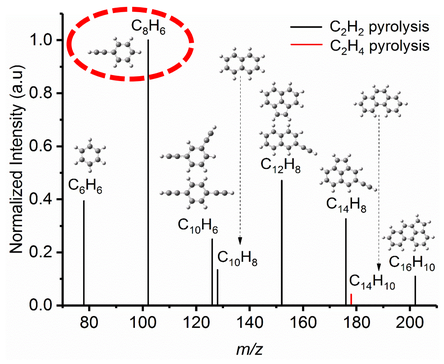


Fig. 2 The mass spectra of acetylene and ethylene pyrolysis products observed at 1485 K [2]

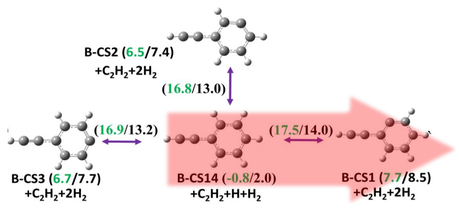


Fig. 3 Potential energy surface with energies calculated at CBS-QB3 (labeled as green) and 6-311+g(d,p) (labeled as black), respectively

Calculation details

- ❑ Potential energy surface of the reaction system
  - Optimize the structures and calculate energies at DFT B3LYP 6-311+G(d,p) level
  - Refine energies at CBS-QB3 level
- ❑ Kinetic analysis
  - Calculate reaction rate coefficients by TST theory
  - Check pressure dependence by RRKM theory
- ❑ Simulated using 0-D batch reaction in Chemkin

Yield distributions in 0-D batch reactor

In the whole $C_6H_5C_2H$-C_2H_2-H-Ar reaction system, the initial reactants are set the same as the experimental premixed ethylene flames in Castaldi's work [3]: 193 ppm, 100 ppm, 0.045 and 0.954707 for $C_6H_5C_2H$, H, and C_2H_2 and Ar, respectively.

Fig. 4 Comparison of HACA mechanism and this reaction network in the production of PAH with different sizes at elevated temperatures.

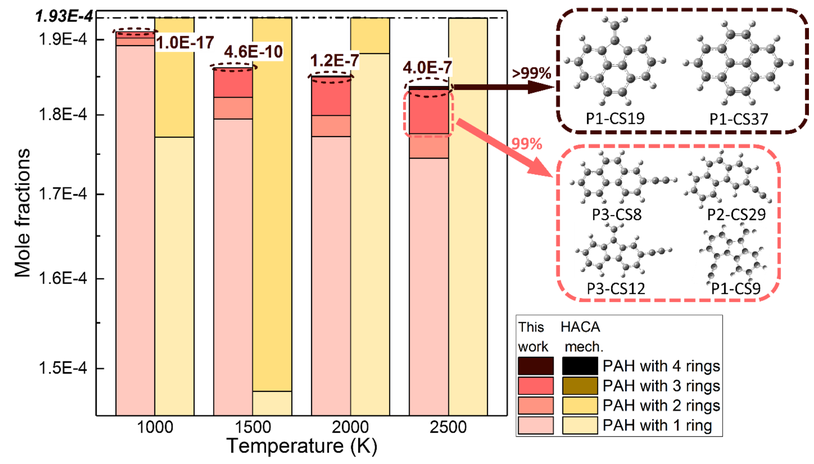
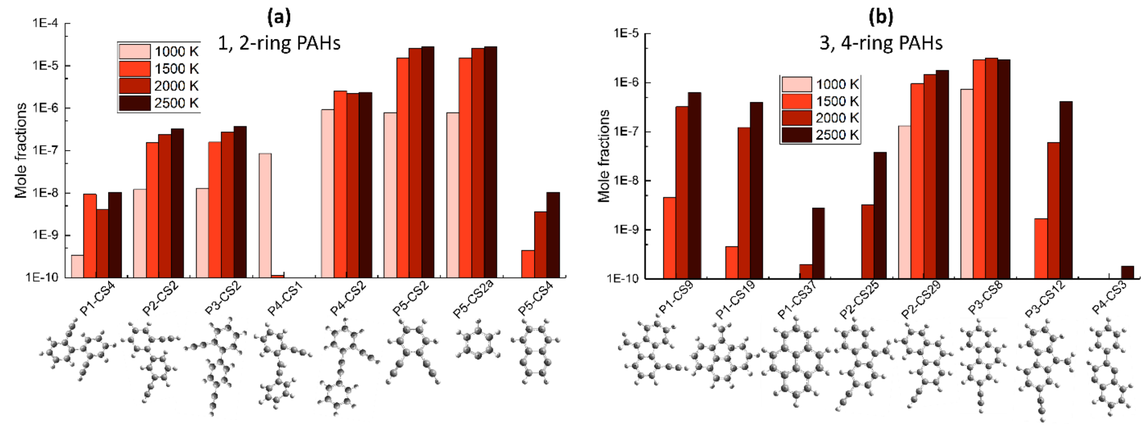
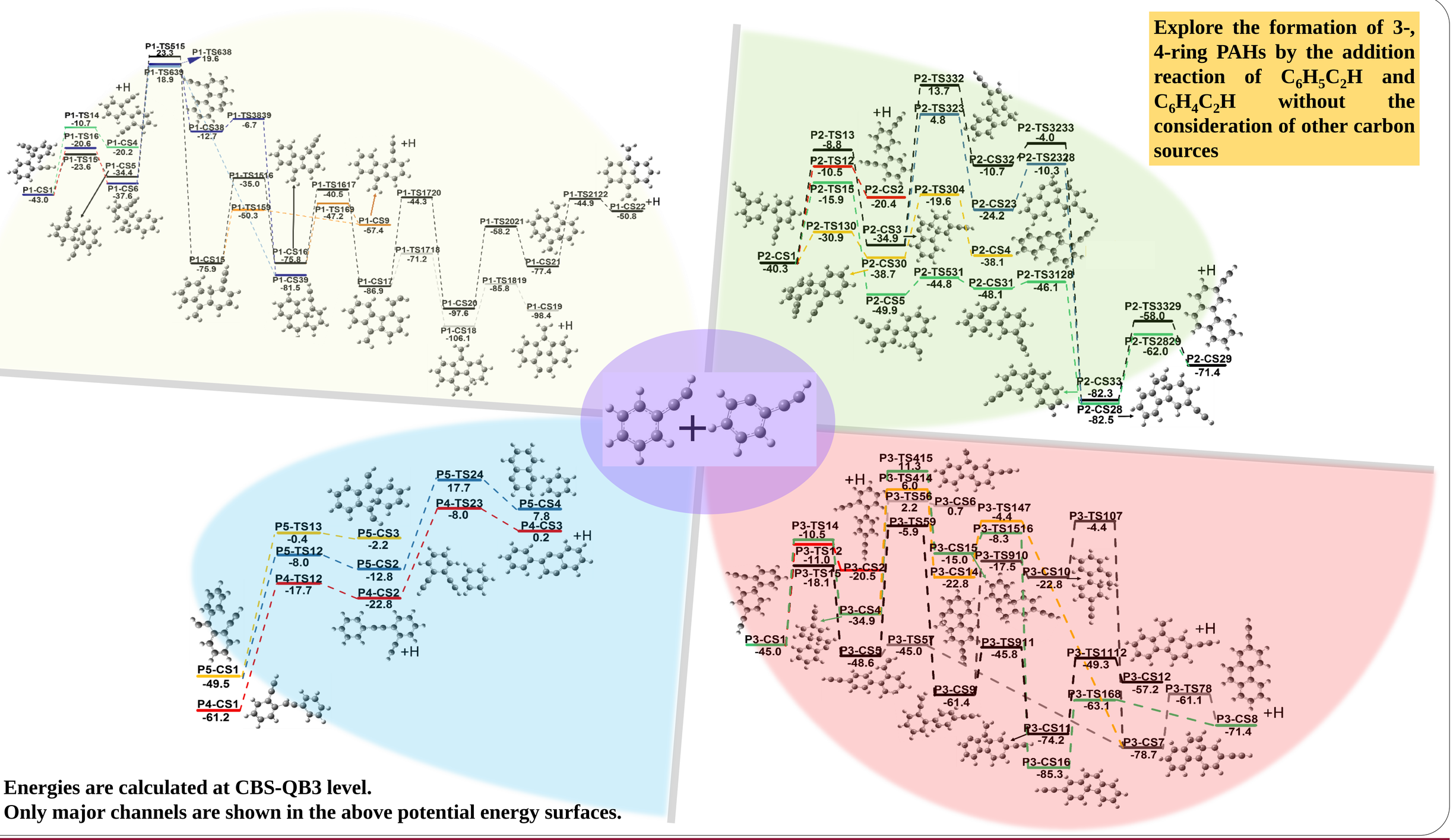


Fig. 5 Simulated mole fractions of the major products in the $C_6H_5C_2H$-C_2H_2-H-Ar system in a 0-D batch reactor via the $C_6H_5C_2H$ reaction network.



Potential energy surfaces of the reaction system

Explore the formation of 3-, 4-ring PAHs by the addition reaction of $C_6H_5C_2H$ and $C_6H_4C_2H$ without the consideration of other carbon sources



❖ **Energies are calculated at CBS-QB3 level.**
❖ **Only major channels are shown in the above potential energy surfaces.**

References

[1] M.J. Castaldi, N.M. Marinov, C.F. Melius, J. Huang, S.M. Senkan, W.J. Pit, C.K. Westbrook, Experimental and modeling investigation of aromatic and polycyclic aromatic hydrocarbon formation in a premixed ethylene flame, P. Combust. Inst. 26 (1996) 693-702.
[2] Liu, Peng, et al. "The site effect on PAHs formation in HACA-based mass growth process." Combustion and Flame 199 (2019): 54-68.
[3] M.J. Castaldi, Marinov, N. M., Melius, C. F., Huang, J., Senkan, S. M., Pit, W. J., Westbrook, C. K. , Experimental and modeling investigation of aromatic and polycyclic aromatic hydrocarbon formation in a premixed ethylene flame, Symposium (International) on Combustion 26 (1996) 673-702