DETAILED KINETIC MODELING OF SOOT FORMATION: COMPARISON OF THE RESULTS OF CALCULATIONS BY THE METHOD OF MOMENTS, SECTIONAL METHOD, AND DISCRETE GALERKIN METHOD

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Abstract: The results of a direct comparison of detailed kinetic modeling of the process of soot formation during pyrolysis and oxidation of various hydrocarbons by the numerical method of moments, sectional method and discrete Galerkin method with the results of experimental measurements in reflected shock waves are reported. The predictions of kinetic simulations of the soot yield, average particle size and total particle concentration by the method of moments and the discrete Galerkin method were found to be in close agreement with the available experimental results. For the first time, the unified kinetic mechanism of soot formation we developed was used to describe in detail the particle size distribution functions for all hydrocarbons tested in the present work. The obtained distribution functions exhibit a bimodal behavior. The first peak can be associated with soot nuclei and small soot particles, whereas the second peak corresponds to large particles, formed as a result of surface growth and coagulation.

Keywords: pyrolysis and oxidation of hydrocarbons; reflected shock waves; soot formation; detailed kinetic modeling

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References

1. Frenklach, M., and H. Wang. 1991. Size distribution and chemical composition of nascent soot formed in premixed ethylene flames. *P. Combust. Inst.* 23:1559–1566.
2. Frenklach, M., and H. Wang. 1994. Detailed mechanism and modeling of soot particle formation. *Soot formation in combustion: Mechanisms and models of soot formation*. Ed. H. Bockhorn. Springer ser. in chemical physics. Berlin: Springer-Verlag. 59:165–192.
3. Colket, M. B., and R. J. Hall. 1994. Successes and uncertainties in modeling soot formation in laminar, premixed flames. *Soot formation in combustion: Mechanisms and models of soot formation*. Ed. H. Bockhorn. Springer ser. in chemical physics. Berlin: Springer-Verlag. 59:442–468.
4. Mauss, F., T. Schafer, and H. Bockhorn. 1994. Inception and growth of soot particles in dependence on the surrounding gas phase. *Combust. Flame* 99(3-4):697–705.
5. Kazakov, A., H. Wang, and M. Frenklach. 1995. Detailed modeling of soot formation in laminar premixed ethylene flames at a presuure of 10 bar. *Combust. Flame* 100:111–120.
6. Wang, H., and M. Frenklach. 1997. Detailed kinetic modeling study of aromatics formation in laminar premixed acetylene and ethylene flames. *Combust. Flame* 110:173–221.
7. Kennedy, I. M. 1997. Models of soot formation and oxidation. *Prog. Energ. Combust.* 23:95–132.
8. Appel, J., H. Bockhorn, and M. Frenklach. 2000. Kinetic modeling of soot formation with detailed chemistry and physics: Laminar premixed flames of C_2 hydrocarbons. *Combust. Flame* 121(1-2):122–136.
9. Richter, H., and J. B. Howard. 2000. Formation of polycyclic aromatic hydrocarbons and their growth to soot — a review of chemical reaction pathways. *Prog. Energ. Combust.* 26:565–608.
10. Frenklach, M. 2002. Reaction mechanism of soot formation in flames. *Phys. Chem. Chem. Phys.* 4:2028–2037.
11. D'Anna, A., and A. Violi. 2005. Detailed modeling of the molecular growth process in aromatic and aliphatic premixed flames. *Energ. Fuel.* 19:79–86.
12. Richter, H., S. Granata, W. H. Green, and J. B. Howard. 2005. Detailed modeling of PAH and soot formation in laminar preliminary mixture benzene/oxygen/argon at low pressure flame. *P. Combust. Inst.* 30:1397–1405.
13. Schuetz, C. A., and M. Frenklach. 2002. Nucleation of soot: Molecular dynamics simulations of pyrene dimerization. *P. Combust. Inst.* 29:2307–2314.

14. Violi, A., A. Kubota, T. N. Truong, W. J. Pitz, C. K. Westbrook, and A. F. Sarofim. 2002. A fully integrated kinetic Monte Carlo/molecular dynamics approach for the simulation of soot precursor growth. *P. Combust. Inst.* 29:2343–2349.
15. Violi, A., A. F. Sarofim, and G. A. Voth. 2004. Kinetic Monte Carlo-molecular dynamics approach to model soot inception. *Combust. Sci. Technol.* 176:991–1005.
16. Gray, H. A., and G. R. Cass. 1998. Source contributions to atmospheric fine particle concentrations. *Atmos. Environ.* 32:3805–3825.
17. Haywood, J., and V. Ramaswamy. 1998. Global sensitivity studies of the direct radiative forcing due to anthropogenic sulfate and black carbon aerosols. *J. Geophys. Res.* 103:6043–6058.
18. Menon, S., J. Hansen, L. Nazarenko, and Y. Luo. 2002. Climate effects of black carbon aerosols in China and India. *Science* 297:2250–2253.
19. D'Alessio, A., A. D'Anna, A. D'Orsi, P. Minutolo, R. Barbella, and A. Ciajolo. 1992. Precursor formation and soot inception in premixed ethylene flames. *P. Combust. Inst.* 24:973–980.
20. Zhao, B., Z. Yang, M. V. Johnston, H. Wang, A. S. Wexler, M. Balthasar, and M. Kraft. 2003. Measurement and numerical simulation of soot particle distribution functions in a laminar premixed ethylene–oxygen–argon flame. *Combust. Flame* 133:173–188.
21. Sgro, L. A., A. De Filippo, G. Lanzuolo, and A. D'Alessio. 2007. Characterization of nanoparticles of organic carbon (NOC) produced in rich premixed flames by differential mobility analysis. *P. Combust. Inst.* 31:631–638.
22. Minutolo, P., G. Gambi, A. D'Alessio, and S. Carlucci. 1999. Spectroscopic characterisation of carbonaceous nanoparticles in premixed flames. *Atmos. Environ.* 33:2725–2732.
23. Novakov, T., and J. E. Penner. 1993. Large contribution of organic aerosols to cloud-condensation-nuclei concentrations. *Nature* 365:823–826.
24. Rivera-Carpio, C. A., C. E. Corrigan, T. Novakov, J. E. Penner, C. F. Rogers, and J. C. Chow. 1996. Derivation of contributions of sulfate and carbonaceous aerosols to cloud condensation nuclei from mass size distributions. *J. Geophys. Res.* 101:19483–19493.
25. Oberdorster, G., Z. Sharp, V. Atudorei, A. Elder, R. Gelein, W. Kreyling, and C. Cox. 2004. Translocation of inhaled ultrafine particles to the brain. *Inhal. Toxicol.* 16:437–445.
26. Agafonov, G. L., I. V. Bilera, P. A. Vlasov, Yu. A. Kolbanovskii, V. N. Smirnov, and A. M. Tereza. 2015. Soot formation during the pyrolysis and oxidation of acetylene and ethylene in shock waves. *Kinet. Catal.* 56(1):12–30.
27. Wang, H., X. You, A. V. Joshi, S. G. Davis, A. Laskin, F. Egolfopoulos, and C. K. Law. May, 2007. USC Mech Version II. High-temperature combustion reaction model of $H_2/CO/C_1–C_4$ compounds. Available at: http://ignis.usc.edu/USC_-Mech_II.htm (accessed November 29, 2019).
28. Agafonov, G. L., P. A. Vlasov, and V. N. Smirnov. 2015. Soot formation in the pyrolysis of benzene, methylbenzene, and ethylbenzene in shock waves. *Kinet. Catal.* 52(3):358–370.
29. Agafonov, G. L., V. N. Smirnov, and P. A. Vlasov. 2011. Shock tube and modeling study of soot formation during the pyrolysis and oxidation of a number of aliphatic and aromatic hydrocarbons. *P. Combust. Inst.* 33:625–632.
30. Skjøth-Rasmussen, M. S., P. Glarborg, M. Østberg, J. T. Johannessen, H. Livbjerg, A. D. Jensen, and T. S. Christensen. 2004. Formation of polycyclic aromatic hydrocarbons and soot in fuel-rich oxidation of methane in a laminar flow reactor. *Combust. Flame* 136:91–128.
31. Agafonov, G. L., I. V. Bilera., P. A. Vlasov, I. V. Zhil'tsova, Yu. A. Kolbanovskii, V. N. Smirnov, and A. M. Tereza. 2016. Unified kinetic model of soot formation in the pyrolysis and oxidation of aliphatic and aromatic hydrocarbons in shock waves. *Kinet. Catal.* 57:557–572.
32. Agafonov, G. L., I. Naydenova, P. A. Vlasov, and J. Warnatz. 2007. Detailed kinetic modeling of soot formation in shock tube pyrolysis and oxidation of toluene and n-heptane. *P. Combust. Inst.* 31:575–583.
33. Harris, S. J., and A. M. Weiner. 1983. Surface growth of soot particles in premixed ethylene/air flames. *Combust. Sci. Technol.* 31:155–167.
34. Frenklach, M. 1996. On surface growth mechanism of soot particles. *P. Combust. Inst.* 26:2285–2293.
35. Deuflhard, P., and M. Wulkow. 1989. Computational treatment of polyreaction kinetics by orthogonal polynomials of a discrete variable. *Impact Comput. Sci. Eng.* 1:269–301.
36. Ackermann, J., and M. Wulkow. 1990. MACRON — a program package for macromolecular kinetics. Berlin: Konrad-Zuse-Zentrum. Preprint SC 90-14. 36 p.
37. Frenklach, M., and S. J. Harris. 1987. Aerosol dynamics modeling using the method of moments. *J. Colloid Interf. Sci.* 118:252–261.
38. Smoluchowski, M. 1917. Versuch einer mathematischen Theorie derivative Koagulationskinetik kolloidaler Loesungen. *Z. Phys. Chem.* 92:129–168.
39. Gelbard, F., and J. H. Seinfeld. 1980. Simulation of multicomponent aerosol dynamics. *J. Colloid Interf. Sci.* 78(2):485–501.
40. Marchal, P., R. David, J. P. Klein, and J. Villermaux. 1988. Crystallization and precipitation engineering I: An effective method for solving the population balance for crystallization with agglomeration. *Chem. Eng. Sci.* 43(1):59–67.
41. Landgrebe, J. D., and S. E. Pratsinis. 1990. A discrete-sectional model for powder production by gas-phase chemical reaction and aerosol coagulation in the free-molecular regime. *J. Colloid Interf. Sci.* 139(1):63–86.
42. Hounslow, M. J., R. L. Ryall, and V. R. Marshall. 1988. A discretized population balance for nucleation, growth, and aggregation. *AICHE J.* 34(11):1821–1832.
43. Litster, J. D., D. J. Smit, and M. J. Hounslow. 1995. Adjustable discretized population balance for growth and aggregation. *AICHE J.* 41(3):591–603.

44. Wynn, E. J. W. 1996. Improved accuracy and convergence of discretized population balance of Litster *et al*. *AICHE J*. 42(7):2084–2086.
45. Kumar, S., and D. Ramkrishna. 1996. On the solution of population balance equations by discretization — III. Nucleation, growth and aggregation of particles. *Chem. Eng. Sci*. 52(24):4659–4679.
46. Kellerer, H., A. Müller, H.-J. Bauer, and S. Wittig. 1996. Soot formation in a shock tube under elevated pressure conditions. *Combust. Sci. Technol*. 113-114:67–80.
47. Mikheeva (Popova), E. Yu. 2013. Eksperimental'nye issledovaniya teplovykh effektov i protsessa formirovaniya sazhevykh chastits pri udarno-volnovom pirolize uglevodorodov [Experimental studies of the heat effects and soot formation process during shock-tube pyrolysis of hydrocarbons]. Moscow. PhD Diss.
48. Starke, R., B. Kock, and P. Roth. 2003. Nano-particle sizing by laser-induced-incandescence (LII) in a shock wave reactor. *Shock Waves* 12:351–360.
49. Warnatz, J., U. Maas, and R. W. Dibble. 2006. *Combustion: Physical and chemical fundamentals, modeling and simulation, experiments, pollutant formation*. 4th ed. Berlin–Heidelberg: Springer-Verlag. 378 p.
50. Schulz, C., B. F. Kock, M. Hofmann, H. Michelsen, S. Will, B. Bougie, R. Suntz, and G. Smallwood. 2006. Laser-induced incandescence: Recent trends and current questions. *Appl. Phys. B Lasers O*. 83:333–354.

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