COAGULATION OF COMBUSTION-GENERATED CARBONACEOUS NANOPARTICLES OF ETHYLENE AND ETHYLENE/ETHANOL FLAMES IN AN ATMOSPHERIC SIMULATION CHAMBER

V. Liguoro*, V. Vernocchi, G. De Falco*, F. Picca***, F. Sasso***, A. Sannino****, P. Minutolo*, A. D'Anna***, T. Isolabella*****, P. Prati*****, D. Massabò*****, M. Commodo***

vincenzo.liguoro@stems.cnr.it

* Istituto di Scienze e Tecnologie per l'Energia e la Mobilità Sostenibili, CNR, NA, Italy

** INFN, Sezione di Genova, via Dodecaneso 33, 16146 Genoa, Italy

*** DICMAPI, Università degli Studi di Napoli Federico II, Napoli, Italy

**** Dipartimento di Fisica "Ettore Pancini", Università di Napoli Federico II, NA, Italy

***** Dipartimento di Fisica – Università degli Studi di Genova and INFN, Genoa, Italy

Abstract

The coagulation of soot nanoparticles was experimentally and numerically investigated using an atmospheric simulation chamber operated in dark conditions to isolate coagulation from photochemical processes. The nanoparticles were produced in a highly controlled laboratory flame fueled with pure ethylene and ethylene/ethanol mixtures. The model used for the simulations is based on the Smoluchowski equation, in which the gas-kinetic constant is calculated in the transitional regime. The fractal theory has been incorporated into the model. The coagulation enhancement factor β was observed to be constant while the mobility fractal dimension d_{fm} was shown to decrease with the Ethanol addition.

Introduction

The combustion of biofuels can emit ultrafine particles (UFPs) with distinct size distributions, structures, and chemical compositions compared to conventional fossil fuels. Research indicates that while biofuels generally reduce total particulate matter emissions, they tend to produce a higher concentration of smaller particles [1], which may pose greater health risks. Additionally, the early-stage coagulation of soot nanoparticles, within the exhaust system or shortly after emission, can alter their size distribution and aggregation state, impacting their atmospheric lifetime, optical properties, and potential toxicity [2]. In this work, the coagulation of soot nanoparticles was experimentally and numerically investigated using an atmospheric simulation chamber, ChAMBRe [3], operated in dark conditions to isolate coagulation from photochemical processes. Details of the chamber are reported in [4]. The nanoparticles were produced in a highly controlled laboratory flame fueled with pure ethylene and ethylene/ethanol mixtures, used as proxies for petro-derived and bio-derived fuels, respectively. Particles are collected at a height above the burner *H*, which differs for the two fuels to ensure that the same particle distribution is injected into the chamber. The model used for the simulations is based on the Smoluchowski equation, in which the gas-kinetic constant is calculated in the transitional regime [5]. Following previous works [6], the fractal theory has been incorporated into the model. The coagulation enhancement factor β and the mobility fractal dimension d_{fm} have been optimized by the best fit of the experimental data with the simulation results.

Results and discussion

Three experiments were analyzed, one for the ethylene flame and two in which 20% carbon has been replaced with ethanol "EtOH20%", all with a C/O=0.67. Fig. 1 shows the results obtained by inserting the input data of Table 1 into the simulations.

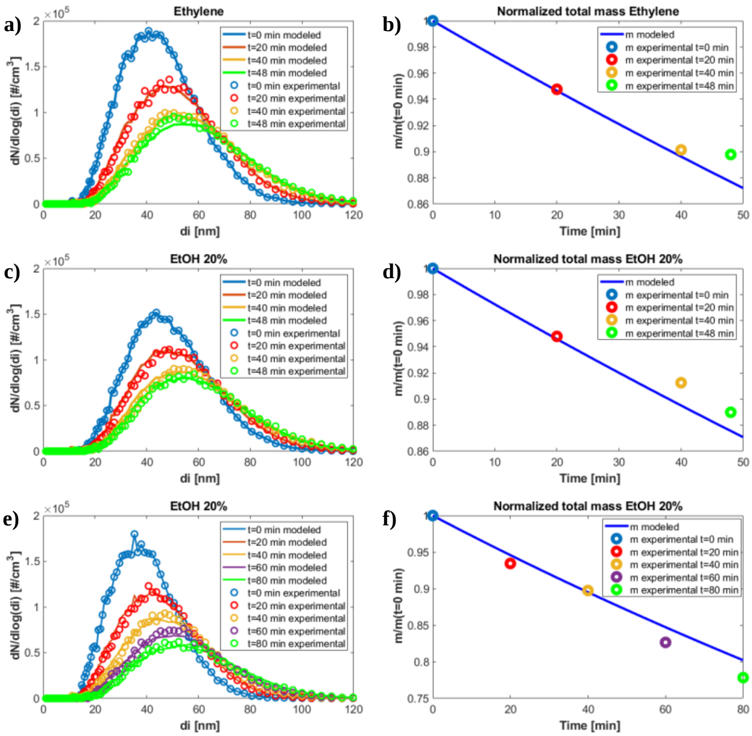

Figure 1 Comparison of experimental (dots) and simulated data (line) for particles from pure ethylene (panel c) and d)) and EtOH 20% 0.67 flame (panel a), b), e) and f)). In panels a), c), and e), the particles' distribution functions in the ChAMBRe at different times are shown, while in panels b), d), and f), the normalized total mass versus time is shown.

The temperature was set at 300 K and the pressure at 1.01 bar. The primary particles' mean diameter, d_0, is 4 nm, while their density ρ_0 and the pre-exponential factor are fixed respectively at 1.5 g/cm^3 and 1.0. The simulations exhibit good agreement with experimental data. In Fig 1, panels a), c) and e) report the experimental and modeled particles' distribution functions at different times in order to check the reliability of simulations, while panels b), d) and f) show the normalized total mass versus time, which is essential for evaluating d_{fm}.There is a reduction of mass due to the diffusion and settling losses in the chamber and to the air sampling from SMPS.

Table 1. Optimized parameters for experiments and simulations.

Experiment	C/O	H, mm	β	d_{fm}	d_0, nm	k_g	ρ_0, g/cm^3
EtOH 0%	0.67	11.5	2.2	2.19	4	1.0	1.5
EtOH 20% exp 1	0.67	17	2.2	2.16	4	1.0	1.5
EtOH 20% exp 2	0.67	17	2.2	2.18	4	1.0	1.5

Conclusions

Based on the simulation results, we can conclude that adding ethanol to the flame does not affect the value of β, which remains constant at 2.2. However, there is a reduction in the mobility fractal dimension d_{fm}. Consequently, the coagulation velocity is the same in both types of flame, while the variation of d_{fm} implies a difference in the particles' fractal properties. EtOH 20% flame generates more stretched agglomerates than the pure ethylene flame.

Acknowledgments

This work was supported by the European Union under the Next generation EU program project number 2022CH87SA.

References

[1] Lapuerta, M., Armas, O., Rodríguez-Fernández, J., "Effect of biodiesel fuels on diesel engine emissions", *Prog. Energy Combust. Sci.* 34:198-223 (2008).
[2] Kittelson, D. B., Watts, W. F., Johnson, J. P., "On-road and laboratory evaluation of combustion aerosols—Part1: Summary of diesel engine results", *J. Aerosol Sci.* 37: 913–930 (2006)
[3] https://actris.it/infn-genova-nf-chambre/
[4] Massabò, D., Danelli, S.G., Brotto, P., Comite, A., Costa, C., Di Cesare, A., Doussin, J.F., Ferraro, F., Formenti, P., Gatta, E., Negretti, L., Oliva, M., Parodi, F., Vezzulli, L., Prati, P. "ChAMBRe: a new atmospheric simulation chamber for aerosol modelling and bio-aerosol research", *Atmos. Meas. Tech.* 11:5885–5900 (2018)
[5] Fuks, N.A., *The mechanics of aerosols*, AH CCCP, Institute of Scientific Information, 1955.
[6] Maricq, M.M., "Coagulation dynamics of fractal-like soot aggregates", J. *Aerosol Sci.*, 38: 141-156 (2007)