

Achim Wick

## **Population Balance Modeling and Large-Eddy Simulation of Soot Formation and Oxidation**

# **Population Balance Modeling and Large-Eddy Simulation of Soot Formation and Oxidation**

## **Populationsbilanz-Modellierung und Large-Eddy Simulation von Rußbildung und Rußoxidation**

Von der Fakultät für Maschinenwesen der Rheinisch-Westfälischen  
Technischen Hochschule Aachen zur Erlangung des akademischen Grades  
eines Doktors der Ingenieurwissenschaften genehmigte Dissertation

vorgelegt von

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Tag der mündlichen Prüfung: 19.12.2019



Berichte aus der Strömungstechnik

**Achim Wick**

**Population Balance Modeling and  
Large-Eddy Simulation of  
Soot Formation and Oxidation**

Shaker Verlag  
Düren 2020

**Bibliographic information published by the Deutsche Nationalbibliothek**

The Deutsche Nationalbibliothek lists this publication in the Deutsche Nationalbibliografie; detailed bibliographic data are available in the Internet at <http://dnb.d-nb.de>.

Zugl.: D 82 (Diss. RWTH Aachen University, 2019)

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Printed in Germany.

ISBN 978-3-8440-7275-4

ISSN 0945-2230

Shaker Verlag GmbH • Am Langen Graben 15a • 52353 Düren

Phone: 0049/2421/99011-0 • Telefax: 0049/2421/99011-9

Internet: [www.shaker.de](http://www.shaker.de) • e-mail: [info@shaker.de](mailto:info@shaker.de)

## Acknowledgements

The research presented in this thesis was conducted at the Institute for Combustion Technology (ITV) of the RWTH Aachen University. Most of this work was funded by the German Research Foundation (DFG) and the German Research Association for Combustion Engines e.V. (FVV), which I gratefully acknowledge. I am also thankful for funding by the European Union within the Seventh Framework Programme (FP7/2007-2013).

During the course of this work, I had the pleasure to work in an inspiring environment and to collaborate with international researchers, and there are a lot of people I am beholden to. First and foremost, I would like to thank my adviser Prof. Heinz Pitsch for his continuous support and guidance and for his encouragement in developing and following my own research ideas. Further, I would like to thank Prof. Rodney Fox, from whom I learned a lot about numerical methods for aerosol dynamics. I would like to thank Prof. Karl Alexander Heufer for his interest in my work and for being the chair of the examination committee.

I would like to thank all colleagues at the ITV. In particular, I would like to express my gratitude to Dr. Antonio Attili, Lukas Berger, Dr. Stephan Kruse, Raymond Langer, and Abhinav Sharma for our fruitful collaboration and for sharing their expertise in computational and experimental methods to study soot formation. I would also like to thank all students who contributed to this work as student research assistants and with their Bachelor and Master theses.

Posthumously, I would like to thank Prof. Norbert Peters, whose enthusiasm for science was always a great inspiration for me. I would also like to thank my former colleagues Dr. Michael Gauding and Daniel Mayer, who introduced me to the world of turbulence and combustion research when I was an undergraduate student at the ITV.

Last but not least, I would like to thank my parents and grandparents for all their love and support in every aspect of my life.



## Publications

This thesis is mainly based on the following peer-reviewed publications; some material is updated, together with some new introduced results:

- A. Wick, M. Frenklach, and H. Pitsch (2020): Systematic assessment of the Method of Moments with Interpolative Closure and guidelines for its application to soot particle dynamics in laminar and turbulent flames. *Combustion and Flame*, 214, 450-463.
- A. Wick, T.-T. Nguyen, F. Laurent, R. O. Fox, and H. Pitsch (2017): Modeling soot oxidation with the Extended Quadrature Method of Moments. *Proceedings of the Combustion Institute*, 36, 789-979.
- A. Wick, A. Attili, F. Bisetti, and H. Pitsch (2020): DNS-driven analysis of the Flamelet/Progress Variable model assumptions regarding soot inception, growth, and oxidation in turbulent flames. *Combustion and Flame*, 214, 437-449.
- A. Wick, F. Priesack, and H. Pitsch (2017): Large-Eddy Simulation and detailed modeling of soot evolution in a model aero engine combustor. *Proceedings of the ASME Turbo Expo 2017*, GT2017-63293.





# Abstract

To meet the increasing global demand of usable energy and mobility, the combustion of fossil as well as renewable fuels in multifarious applications such as power plants, vehicles, airplanes, industrial furnaces, and household heating systems, is of utmost importance. Pivotal targets for the development of next-generation, high-fidelity combustion devices are the enhancement of the energy efficiency, applicability to alternative fuels and combustion strategies, and the reduction of pollutant emissions, in particular soot emissions due to their adverse impact on the environment as well as human health.

Computational Fluid Dynamics simulations have been established as an integral part of the development process of modern combustion applications. With increasing computing capacities and focus on pollutant emissions, the interest in Large-Eddy Simulations (LES) is growing. In particular, the accuracy of simulations of soot formation in turbulent flames largely benefits from the resolution of large-scale turbulent fluctuations in LES. However, a further exploitation of the predictive potential of LES requires the improvement of a variety of submodels that an integral soot model is composed of as well as appropriate coupling strategies for these model components. Furthermore, methodologies are needed for the systematic analysis of integrated models to identify critical modeling assumptions regarding the overall performance.

The present thesis aims at contributing to both the systematic analysis of integrated LES models for soot evolution and the development of submodels of such integrated modeling suites. The first part of the thesis focuses on the assessment and development of moment methods for the solution of population balance equations governing the soot particle dynamics. A combined a-priori and a-posteriori analysis of different interpolation-based moment methods led to recommendations for an optimal choice of interpolation functions and orders to achieve highly accurate soot predictions for a variety of laminar and turbulent flame conditions. In addition, an advanced quadrature-based moment method, which also provides a reconstruction of an approximation of the soot Number Density Function (NDF), was coupled with a method to describe the disappearance of particles during soot oxidation in a mathematically rigorous manner. Furthermore, a multivariate physico-chemical soot model accounting for the diversity of the chemical composition of soot

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particles was formulated and validated for the evolution of the NDF in a laminar flame.

Next, the propagation of errors related to flamelet assumptions into soot quantities was investigated taking advantage of large-scale Direct Numerical Simulation (DNS) data of a sooting turbulent jet flame. Various terms coupling flamelet-based combustion models with soot models were first analyzed a-priori. Then, a partial a-posteriori analysis was performed, where the soot evolution along Lagrangian trajectories was computed using the flow field and selected thermodynamic properties extracted from the DNS. The resulting error decomposition and quantification identified the modeling of polycyclic aromatic hydrocarbon-based soot growth processes to be associated with uncertainties of leading order. Eventually, LES of a model aircraft combustor were performed employing a flamelet combustion model and a state-of-the-art soot model based on a bivariate description of the soot particles.

# Zusammenfassung

Um den weltweit steigenden Bedarf an nutzbarer Energie und Mobilität zu decken, ist die Verbrennung von fossilen sowie erneuerbaren Brennstoffen in vielfältigen Anwendungen wie Kraftwerken, Fahrzeugen, Flugzeugen, Industrieöfen und Haushaltsheizungen von großer Bedeutung. Hauptziele für die Entwicklung von Verbrennungsanlagen der nächsten Generation sind die Verbesserung der Energieeffizienz, die Kompatibilität mit alternativen Kraftstoffen und Verbrennungsstrategien sowie die Reduzierung von Schadstoffemissionen, insbesondere von Rußemissionen, da diese mit starken negativen Einflüssen auf die Umwelt und die menschliche Gesundheit einhergehen.

Strömungssimulationen haben sich als integraler Bestandteil des Entwicklungsprozesses moderner Verbrennungsanwendungen etabliert. Mit zunehmender Rechenleistung und Fokussierung auf Schadstoffemissionen wächst das Interesse an Large-Eddy Simulationen (LES). Insbesondere die Genauigkeit der Vorhersage von Rußbildung in turbulenten Flammen profitiert erheblich von der Auflösung großskaliger turbulenter Fluktuationen in LES. Eine weitere Verbesserung der Prädiktivität von LES in Bezug auf Rußbildung erfordert die Verbesserung einer Vielzahl von Modellkomponenten, aus denen ein integrales Rußmodell besteht, sowie geeignete Kopplungsstrategien dieser Modellkomponenten. Darüber hinaus werden Methoden für die systematische Analyse integrierter Modelle benötigt, um kritische Modellierungsannahmen bezüglich der Genauigkeit des Gesamtmodells zu identifizieren.

Gegenstand dieser Arbeit ist sowohl die systematische Analyse integrierter Rußmodelle für LES als auch die Entwicklung von Modellkomponenten solcher integrierter Modelle. Fokus des ersten Teils der Arbeit bildet die Analyse und Entwicklung von Momentenmethoden zur Lösung von die Rußpartikeldynamik beschreibenden Populationsbilanzgleichungen. Durch eine kombinierte a-priori und a-posteriori Analyse verschiedener interpolationsbasierter Momentenmethoden konnten Empfehlungen für eine optimale Wahl von Interpolationsfunktionen und -ordnungen abgeleitet werden, welche hochgenaue Vorhersagen der Rußbildung in laminaren und turbulenten Flammen ermöglichen. Darüber hinaus wurde eine neuartige quadraturbasierte Momentenmethode, welche auch eine Rekonstruktion einer Approximation der Rußpartikelverteilungsfunktion ermöglicht, mit einem numerischen Verfahren zur Beschreibung des Verschwindens von Partikeln während der Rußoxidation gekoppelt. Weiter-

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hin wurde ein multivariates physikalisch-chemisches Rußmodell formuliert, welches die starke Diversität der chemischen Zusammensetzung von Rußpartikeln berücksichtigt. Dieses Modell wurde für die zeitliche Entwicklung der Partikelverteilungsfunktion in einer laminaren Flamme validiert.

Im weiteren Verlauf der Arbeit wurde der Einfluss von auf Flamelet-Annahmen beruhenden Modellfehlern auf die Berechnung von Rußgrößen mittels systematischer Analyse von Direkten Numerischen Simulationen (DNS) einer rußenden turbulenten Freistrahlflamme untersucht. Zunächst wurden verschiedene Größen, welche Flamelet-basierte Verbrennungsmodelle mit Rußmodellen koppeln, a-priori analysiert. Anschließend wurde eine partielle a-posteriori-Analyse durchgeführt. Hierbei wurde die Rußbildung entlang Lagrangescher Trajektorien nachgerechnet, wobei das Strömungsfeld sowie ausgewählte thermodynamische Größen aus der DNS extrahiert wurden. Mittels der aus dieser Analyse resultierenden Fehlerzerlegung und -quantifizierung konnte die Modellierung von auf polyzyklischen aromatischen Kohlenwasserstoffen basierenden Rußwachstumsprozessen als Fehlerquelle führender Ordnung identifiziert werden. Weiterhin wurden LES einer Modellbrennkammer eines Flugtriebwerks unter Verwendung eines Flamelet-Verbrennungsmodells sowie eines auf einer bivariaten Beschreibung der Rußpartikel basierenden Rußmodells durchgeführt.

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