

Computational Models for Turbulent Reacting Flows

This book presents the current state of the art in computational models for turbulent reacting flows, and analyzes carefully the strengths and weaknesses of the various techniques described. The focus is on formulation of practical models as opposed to numerical issues arising from their solution.

A theoretical framework based on the one-point, one-time joint probability density function (PDF) is developed. It is shown that all commonly employed models for turbulent reacting flows can be formulated in terms of the joint PDF of the chemical species and enthalpy. Models based on direct closures for the chemical source term as well as transported PDF methods, are covered in detail. An introduction to the theory of turbulence and turbulent scalar transport is provided for completeness.

The book is aimed at chemical, mechanical, and aerospace engineers in academia and industry, as well as developers of computational fluid dynamics codes for reacting flows.

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Preface

In setting out to write this book, my main objective was to provide a reasonably complete introduction to computational models for turbulent reacting flows for students, researchers, and industrial end-users new to the field. The focus of the book is thus on the *formulation* of models as opposed to the numerical issues arising from their solution. Models for turbulent reacting flows are now widely used in the context of computational fluid dynamics (CFD) for simulating chemical transport processes in many industries. However, although CFD codes for non-reacting flows and for flows where the chemistry is relatively insensitive to the fluid dynamics are now widely available, their extension to reacting flows is less well developed (at least in commercial CFD codes), and certainly less well understood by potential end-users. There is thus a need for an introductory text that covers all of the most widely used reacting flow models, and which attempts to compare their relative advantages and disadvantages for particular applications.

The primary intended audience of this book comprises graduate-level engineering students and CFD practitioners in industry. It is assumed that the reader is familiar with basic concepts from chemical-reaction-engineering (CRE) and transport phenomena. Some previous exposure to theory of turbulent flows would also be very helpful, but is not absolutely required to understand the concepts presented. Nevertheless, readers who are unfamiliar with turbulent flows are encouraged to review Part I of the recent text *Turbulent Flows* by Pope (2000) before attempting to tackle the material in this book. In order to facilitate this effort, I have used the same notation as Pope (2000) whenever possible. The principal differences in notation occur in the treatment of multiple reacting scalars. In general, vector/matrix notation is used to denote the collection of thermodynamic variables (e.g., concentrations, temperature) needed to describe a reacting flow. Some familiarity with basic linear algebra and elementary matrix operations is assumed.

The choice of models to include in this book was dictated mainly by their ability to treat the wide range of turbulent reacting flows that occur in technological applications of interest to chemical engineers. In particular, models that cannot treat ‘general’ chemical

kinetics have been excluded. For example, I do not discuss models developed for premixed turbulent combustion based on the ‘turbulent burning velocity’ or on the ‘level-set’ approach. This choice stems from my desire to extend the CRE approach for modeling reacting flows to be compatible with CFD codes. In this approach, the exact treatment of the chemical kinetics is the *sine qua non* of a good model. Thus, although most of the models discussed in this work can be used to treat non-premixed turbulent combustion, this will not be our primary focus. Indeed, in order to keep the formulation as simple as possible, all models are presented in the context of constant-density flows. In most cases, the extension to variable-density flows is straightforward, and can be easily undertaken after the reader has mastered the application of a particular model to constant-density cases.

In order to compare various reacting-flow models, it is necessary to present them all in the same conceptual framework. In this book, a statistical approach based on the one-point, one-time joint probability density function (PDF) has been chosen as the common theoretical framework. A similar approach can be taken to describe turbulent flows (Pope 2000). This choice was made due to the fact that nearly all CFD models currently in use for turbulent reacting flows can be expressed in terms of quantities derived from a joint PDF (e.g., low-order moments, conditional moments, conditional PDF, etc.). Ample introductory material on PDF methods is provided for readers unfamiliar with the subject area. Additional discussion on the application of PDF methods in turbulence can be found in Pope (2000). Some previous exposure to engineering statistics or elementary probability theory should suffice for understanding most of the material presented in this book.

The material presented in this book is divided into seven chapters and two appendices. Chapter 1 provides background information on turbulent reacting flows and on the two classical modeling approaches (chemical-reaction-engineering and fluid-mechanical) used to describe them. The chapter ends by pointing out the similarity between the two approaches when dealing with the effect of molecular mixing on chemical reactions, especially when formulated in a Lagrangian framework.

Chapter 2 reviews the statistical theory of turbulent flows. The emphasis, however, is on collecting in one place all of the necessary concepts and formulae needed in subsequent chapters. The discussion of these concepts is necessarily brief, and the reader is referred to Pope (2000) for further details. It is, nonetheless, essential that the reader become familiar with the basic scaling arguments and length/time scales needed to describe high-Reynolds-number turbulent flows. Likewise, the transport equations for important one-point statistics in inhomogeneous turbulent flows are derived in Chapter 2 for future reference.

Chapter 3 reviews the statistical description of scalar mixing in turbulent flows. The emphasis is again on collecting together the relevant length and time scales needed to describe turbulent transport at high Reynolds/Schmidt numbers. Following Pope (2000), a model scalar energy spectrum is constructed for stationary, isotropic scalar fields. Finally, the transport equations for important one-point scalar statistics in inhomogeneous turbulent mixing are derived in Chapter 3.

In order to model turbulent reacting flows accurately, an accurate model for turbulent transport is required. In Chapter 4 I provide a short introduction to selected computational models for *non-reacting* turbulent flows. Here again, the goal is to familiarize the reader with the various options, and to collect the most important models in one place for future reference. For an in-depth discussion of the physical basis of the models, the reader is referred to Pope (2000). Likewise, practical advice on choosing a particular turbulence model can be found in Wilcox (1993).

With regards to reacting flows, the essential material is presented in Chapters 5 and 6. Chapter 5 focuses on reacting flow models that can be expressed in terms of Eulerian (as opposed to Lagrangian) transport equations. Such equations can be solved numerically using standard finite-volume techniques, and thus can be easily added to existing CFD codes for turbulent flows. Chapter 6, on the other hand, focuses on *transported PDF* or *full PDF* methods. These methods typically employ a Lagrangian modeling perspective and ‘non-traditional’ CFD methods (i.e., Monte-Carlo simulations). Because most readers will not be familiar with the numerical methods needed to solve transported PDF models, an introduction to the subject is provided in Chapter 7.

Chapter 5 begins with an overview of chemical kinetics and the chemical-source-term closure problem in turbulent reacting flows. Based on my experience, closure methods based on the moments of the scalars are of very limited applicability. Thus, the emphasis in Chapter 5 is on presumed PDF methods and related closures based on conditioning on the mixture fraction. The latter is a non-reacting scalar that describes mixing between non-premixed inlet streams. A general definition of the mixture-fraction vector is derived in Chapter 5. Likewise, it is shown that by using a so-called ‘mixture-fraction’ transformation it is possible to describe a turbulent reacting flow by a reduced set of scalars involving the mixture-fraction vector and a ‘reaction-progress’ vector. Assuming that the mixture-fraction PDF is known, we introduce closures for the reaction-progress vector based on chemical equilibrium, ‘simple’ chemistry, laminar diffusion flamelets, and conditional moment closures. Closures based on presuming a form for the PDF of the reacting scalars are also considered in Chapter 5.

Chapter 6 presents a relatively complete introduction to transported PDF methods for turbulent reacting flow. For these flows, the principal attraction of transported PDF methods is the fact that the highly non-linear chemical source term is treated without closure. Instead, the modeling challenges are shifted to the molecular mixing model, which describes the combined effects of turbulent mixing (i.e., the scalar length-scale distribution) and molecular diffusion on the joint scalar PDF. Because the transported PDF treatment of turbulence is extensively discussed in Pope (2000), I focus in Chapter 6 on modeling issues associated with molecular mixing. The remaining sections in Chapter 6 deal with Lagrangian PDF methods, issues related to estimation of statistics based on ‘particle’ samples, and with tabulation methods for efficiently evaluating the chemical source term.

Chapter 7 deviates from the rest of the book in that it describes computational *methods* for ‘solving’ the transported PDF transport equation. Although Lagrangian PDF codes are

generally preferable to Eulerian PDF codes, I introduce both methods and describe their relative advantages and disadvantages. Because transported PDF codes are less developed than standard CFD methods, readers wishing to utilize these methods should consult the literature for recent advances.

The material covered in the appendices is provided as a supplement for readers interested in more detail than could be provided in the main text. Appendix A discusses the derivation of the spectral relaxation (SR) model starting from the scalar spectral transport equation. The SR model is introduced in Chapter 4 as a non-equilibrium model for the scalar dissipation rate. The material in Appendix A is an attempt to connect the model to a more fundamental description based on two-point spectral transport. This connection can be exploited to extract model parameters from direct-numerical simulation data of homogeneous turbulent scalar mixing (Fox and Yeung 1999).

Appendix B discusses a new method (DQMOM) for solving the Eulerian transported PDF transport equation without resorting to Monte-Carlo simulations. This offers the advantage of solving for the joint composition PDF introduced in Chapter 6 using standard finite-volume CFD codes, without resorting to the chemical-source-term closures presented in Chapter 5. Preliminary results found using DQMOM are quite encouraging, but further research will be needed to understand fully the range of applicability of the method.

I am extremely grateful to the many teachers, colleagues and graduate students who have helped me understand and develop the material presented in this work. In particular, I would like to thank Prof. John C. Matthews of Kansas State University who, through his rigorous teaching style, attention to detail, and passion for the subject of transport phenomena, first planted the seed in the author that has subsequently grown into the book that you have before you. I would also like to thank my own students in the graduate courses that I have offered on this subject who have provided valuable feedback about the text. I want especially to thank Kuochen Tsai and P. K. Yeung, with whom I have enjoyed close collaborations over the past several years, and Jim Hill at Iowa State for his encouragement to undertake the writing of this book. I would also like to acknowledge the important contributions of Daniele Marchisio in the development of the DQMOM method described in Appendix B.

For his early support and encouragement to develop CFD models for chemical-reaction-engineering applications, I am deeply indebted to my post-doctoral advisor, Jacques Villermaux. His untimely death in 1997 was a great loss to his friends and family, as well as to the profession.

I am also deeply indebted to Stephen Pope in many different ways, starting from his early encouragement in 1991 to consider PDF methods as a natural modeling framework for describing micromixing in chemical reactors. However, I am particularly grateful that his text on turbulent flows appeared before this work (relieving me of the arduous task of covering this subject in detail!), and for his generosity in sharing early versions of his text, as well as his $\LaTeX$ macro files and precious advice on preparing the manuscript.

Beginning with a Graduate Fellowship, my research in turbulent reacting flows has been almost continuously funded by research grants from the US National Science Foundation. This long-term support has made it possible for me to pursue novel research ideas outside the traditional modeling approach used by chemical reaction engineers. In hindsight, the application of CFD to chemical reactor design and analysis appears to be a rather natural idea. Indeed, all major chemical producers now use CFD tools routinely to solve day-to-day engineering problems. However, as recently as the 1990s the gap between chemical reaction engineering and fluid mechanics was large, and only through a sustained effort to understand both fields in great detail was it possible to bridge this gap. While much research remains to be done to develop a complete set of CFD tools for chemical reaction engineering (most notably in the area of *multiphase* turbulent reacting flows), one is certainly justified in pointing to computational models for turbulent reacting flows as a highly successful example of fundamental academic research that has led to technological advances in real-world applications. Financial assistance provided by my industrial collaborators: Air Products, BASF, BASSELL, Dow Chemical, DuPont, and Fluent, is deeply appreciated.

I also want to apologize to my colleagues in advance for not mentioning many of their excellent contributions to the field of turbulent reacting flows that have appeared over the last 50 years. It was my original intention to include a section in Chapter 1 on the history of turbulent-reacting-flow research. However, after collecting the enormous number of articles that have appeared in the literature to date, I soon realized that the task would require more time and space than I had at my disposal in order to do it justice. Nonetheless, thanks to the efforts of Jim Herriott at Iowa State, I have managed to include an extensive Reference section that will hopefully serve as a useful starting point for readers wishing to delve into the history of particular subjects in greater detail.

Finally, I dedicate this book to my wife, Roberte. Her encouragement and constant support during the long period of this project and over the years have been invaluable.