

Chapter 1

Introduction

1.1 Motivation, aims and audience

1.1.1 *Relevance*

Perhaps the most prominent of the applications of turbulent reacting flows is *turbulent combustion*. Turbulent flames exist in very simple devices such as a blow torch, in domestic boilers, car engines, industrial burners for process heating, and in aircraft or stationary gas turbines. The range of power in these examples ranges from a fraction of a kW to hundreds of MW, i.e. a range of about six orders of magnitude. Increasing the power is of course achieved partly by increasing the size of the combustor and the pressure. However, this alone cannot account for this large range. Increasing overall the rate of the chemical reaction is made possible by the feature of turbulence to decrease the smallest lengthscale of the motion as the Reynolds number increases, and this affects the rate at which fuel and air mix and hence react. To understand this particular phenomenon, i.e. how the turbulent motions affect the rate of chemical reactions, constitutes one of the main objectives of *turbulent reacting flow theory* and is the main focus of this book.

Other applications in the *chemical engineering* industry include chemicals production, pharmaceuticals, and particle synthesis. Problems here include scale-up from laboratory or pilot scale to realistic sizes, which is assisted by Computational Fluid Dynamics (CFD) codes, but only if these incorporate appropriate models for turbulence-chemistry interactions. In the *environment*, we encounter turbulent dispersion of pollutants from various sources into clean air. Most of these pollutants undergo chemical transformations when they react with the air or with other pollutants. There is evidence that the uptake of oxygen by living organisms in water is a

function of the turbulence. To calculate the evolution of these phenomena, and hence be in a position to assess the air or water quality, one needs to understand turbulent reacting flows.

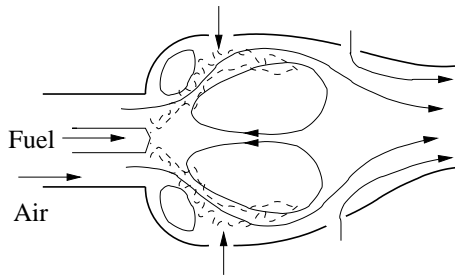
The main classification of turbulent reacting flows in this book follows usual practice and is based on whether the reactants are mixed or not before they reach the main reaction zone. Therefore, we have *non-premixed* and *premixed* flows and some examples are given in Fig. 1.1. Typical flows with non-premixed reactants are found in boilers, gas turbines, *HCl* production, and a power station plume in the atmosphere. For premixed flows, we may mention spark-ignition engines and some stationary gas turbines for combustion applications, and polymerisation fronts from the chemical engineering sector.

Note from Fig. 1.1a, which shows schematically the streamlines in a typical gas turbine burner, the very complicated fluid mechanical patterns that can be encountered in technological applications. Indeed, the success of a simulation of turbulent reacting flows depends to a large extent on the degree to which we can simulate properly the turbulent flow itself and the mixing it causes. Therefore, a study of turbulence and mixing is a prerequisite for an understanding of turbulent reacting flows.

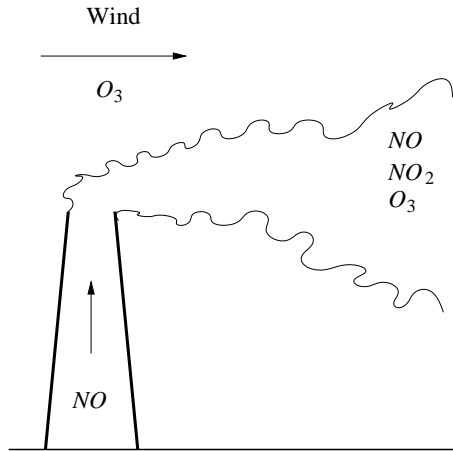
1.1.2 *Aims and structure*

The main aim of this book is to introduce researchers in the early stages of their career or those who are newcomers to the field to the main physical aspects of turbulent flows with chemical reactions and to the theories used for describing such flows. The material is expected to be useful to experimentalists and modellers alike, as it tries to build physical intuition.

We will start with the main concepts and the terminology used in the description of turbulent reacting flows, followed by a brief review of *turbulence*, including its basic features and quantities of interest. Because of its unique importance, *mixing* is discussed in a separate chapter. We then begin our main work on *turbulent flows with non-premixed reactants* and we will present the main theoretical descriptions, followed by a chapter with similar structure on *turbulent flows with premixed reactants*. Finally, the particular requirements that numerical and experimental methods for turbulent reacting flows should have are briefly discussed in the final two chapters of the book.



(a) Gas turbine



(b) Plume

Fig. 1.1 Schematics of applications of turbulent reacting flows. (a) Gas turbine combustor. Depending on the exact location of the fuel injection, the flame may be of non-premixed or premixed character. (b) Power station plume emitting NO in the atmosphere that contains ozone.

1.1.3 Bibliography

This book does not contain a full presentation of every model available and often does not go deeply enough for a complete understanding of the theory. It is aimed as an introduction to the topic and gives to the interested reader some guidance to the large bibliography on turbulent reacting

flows. Here, we suggest some important papers, research monographs and textbooks with some comments as to their usefulness for various parts of the present book, so that the reader may develop a studying strategy more effectively from the beginning.

Turbulence

Many presentations of turbulence, from various points of view and to different depths are available. The following are appropriate for most of the background needed for turbulent reacting flows:

- H. Tennekes and J.L. Lumley (1972) - *A First Course in Turbulence* [139]: one of the first texts and still very good at providing physical intuition;
- S.B. Pope (2000) - *Turbulent Flows* [114] and P.A. Davidson (2004) - *Turbulence. An Introduction for Scientists and Engineers* [46]: both more recent and covering a wide range of topics;
- P.A. Durbin and B.A. Petterson Reif (2001) - *Statistical Theory and Modelling for Turbulent Flows* [49]: concise and very useful for turbulence modelling.

Mixing

This is a topic that is covered at various depths in turbulence books, such as the above, and review articles. We also recommend:

- G.T. Csanady (1973) - *Turbulent Diffusion in the Environment* [43]: an excellent, didactic presentation of molecular mixing, random walks, and mixing by turbulent dispersion;
- V. Kuznetsov and V.A. Sabel'nikov (1990) - *Turbulence and Combustion* [83] and Fox (2003) - *Computational Models for Turbulent Reacting Flows* [54] focus on small-scale aspects of mixing and the PDF method;
- A Special Issue of *Flow Turbulence and Combustion*, vol. 72, (2004) contains very useful reviews (e.g. Refs. [17, 132]). See also the article by Warhaft (2000) [145].

Turbulent reacting flows

There are many review papers and books that describe the main problems and techniques used in describing turbulent reacting flows. We recommend the following, as a start:

- The books by Peters (2000) - *Turbulent Combustion* [107] and

Poinsot and Veynante (2005) - *Theoretical and Numerical Combustion* [109] are complete expositions and are very important entries in the literature.

- The book edited by P.A. Libby and F.A. Williams (1980) - *Turbulent Reacting Flows* is, in our view, a seminal contribution. It contains an excellent introduction to the basic concepts and no study of turbulent reacting flows is complete without reading some chapters of this book, so as to understand at least the historical development of the subject. We strongly recommend [14] for non-premixed flows; [23] for premixed flows; [103] for the PDF method. The sequel appeared in 1994 [85] and contains some updated reviews of the topics included in the first edition.
- R.O. Fox (2003) - *Computational Models for Turbulent Reacting Flows* [54] contains complete mathematical expositions of many models and also discusses models from the chemical engineering community.
- Various review articles have appeared over the years, some on general aspects of reacting flows and others on particular methods. We will use them later at various points in this book. The review by Veynante and Vervisch [143] is a good recent starting point.

Combustion

Most combustion textbooks contain chapters on turbulent combustion that can provide additional material for the reader of this book. The student of turbulent combustion must have a solid background on laminar flames. The reader may wish to consult the following:

- D.B. Spalding (1979) - *Combustion and Mass Transfer* [131];
- F.A. Williams (1985) - *Combustion Theory* [148];
- S.R. Turns (2000) - *An Introduction to Combustion* [141];
- J. Warnatz *et al.* (2005) - *Combustion* [147].

1.2 Governing equations

1.2.1 Instantaneous equations and basic concepts

The content of this section is based on Refs. [107, 109], but it appears in a virtually identical manner in most textbooks and review articles. The equations governing flows with chemical reactions are the continuity, the

Navier-Stokes, the energy and the species conservation equations. A solution to these equations provides in principle all the information we seek from a reacting flow.

Conservation of mass

$$\frac{\partial \rho}{\partial t} + \frac{\partial(\rho u_k)}{\partial x_k} = 0 \quad (1.1)$$

Navier-Stokes

$$\frac{\partial(\rho u_i)}{\partial t} + \frac{\partial(\rho u_k u_i)}{\partial x_k} = -\frac{\partial p}{\partial x_i} + \rho g_i + \frac{\partial \tau_{ik}}{\partial x_k} \quad (1.2)$$

with the shear stress related to strain rate by:

$$\tau_{ij} = \mu \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) - \frac{2}{3} \mu \frac{\partial u_k}{\partial x_k} \delta_{ij} \quad (1.3)$$

Conservation of mass for species α

$$\frac{\partial(\rho Y_\alpha)}{\partial t} + \frac{\partial(\rho u_k Y_\alpha)}{\partial x_k} = \frac{\partial}{\partial x_k} \left(\rho D \frac{\partial Y_\alpha}{\partial x_k} \right) + w_\alpha \quad (1.4)$$

where we have assumed Fick's law for diffusion.

Conservation of energy

The energy equation can be given in terms of enthalpy, internal energy, or temperature and the form to use is often a matter of taste or convenience [107, 109], although internal energy and enthalpy are easier to work with in turbulent combustion. In terms of enthalpy,

$$\frac{\partial(\rho h)}{\partial t} + \frac{\partial(\rho u_k h)}{\partial x_k} = \frac{\partial p}{\partial t} + \frac{\partial}{\partial x_k} \left(\lambda \frac{\partial T}{\partial x_k} \right) + \frac{\partial}{\partial x_k} \left(\rho D \sum_{\alpha=1}^N h_\alpha \frac{\partial Y_\alpha}{\partial x_k} \right) \quad (1.5)$$

while in terms of temperature,

$$\begin{aligned} c_{p,m} \frac{\partial(\rho T)}{\partial t} + c_{p,m} \frac{\partial(\rho u_k T)}{\partial x_k} &= \frac{\partial p}{\partial t} + \frac{\partial}{\partial x_k} \left(\lambda \frac{\partial T}{\partial x_k} \right) \\ &\quad - \sum_{\alpha=1}^N h_\alpha w_\alpha + \rho D \frac{\partial T}{\partial x_k} \sum_{\alpha=1}^N c_{p,\alpha} \frac{\partial Y_\alpha}{\partial x_k} \end{aligned} \quad (1.6)$$

The main quantities appearing in these equations are the velocity u_i , the density ρ , the pressure p . g_i is the body force, τ_{ij} the viscous stress, Y_α is the mass fraction of species α , D its diffusion coefficient (here taken equal for all species), w_α the chemical source term, λ is the conductivity, T the temperature, and h_α is the absolute enthalpy, i.e. the sensible enthalpy

plus the standard enthalpy of formation (e.g. [141], Ch. 1). The mixture specific heat capacity is given by $c_{p,m} = \sum_{\alpha=1}^N Y_{\alpha} c_{p,\alpha}$, where N is the total number of species comprising the mixture and the mixture enthalpy by $h = \sum_{\alpha=1}^N Y_{\alpha} h_{\alpha}$. We have assumed a low speed flow so that viscous heating and acoustic interactions are negligible and we have neglected radiation.

Equation of state

For most problems of practical interest, the pressure, density and temperature are related through the ideal gas equation of state with R the gas constant for the mixture ($R = R_u/M_m$; R_u the universal gas constant and M_m the mixture molar mass):

$$p = \rho RT \quad (1.7)$$

Non-dimensional numbers

Various non-dimensional numbers can be formed from the parameters that appear in the above equations. The main ones are the Reynolds, Prandtl and Schmidt numbers, defined by:

$$\text{Re} = \frac{\rho U L}{\mu} \quad (1.8)$$

$$\text{Pr} = \frac{\mu c_p}{\lambda} = \frac{\nu}{\alpha}, \quad \nu = \frac{\mu}{\rho}, \quad \alpha = \frac{\lambda}{\rho c_p} \quad (1.9)$$

$$\text{Sc} = \frac{\mu}{\rho D} = \frac{\nu}{D} \quad (1.10)$$

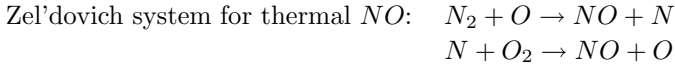
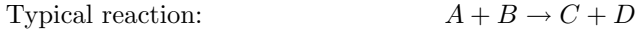
with α the thermal diffusivity and ν the kinematic viscosity. The quantity U is a scale characterizing the velocity and L is a typical lengthscale of the flow. Finally, the ratio between the thermal diffusivity and the mass diffusivity is called the Lewis number

$$\text{Le} = \frac{\lambda}{\rho c_p D} \quad (1.11)$$

and is used often in combustion.

Chemical source term and the Damköhler number

The chemical source term that appears in Eqs. (1.4) and (1.6) depends on the particular system we are studying. Some examples are given below:



Usually, reactions proceed through a series of elementary reactions such as those above and this series is then called a *chemical mechanism*. More formally, in a system where N species participate in a mechanism with K reactions, each elementary reaction can be written as:

$$\sum_{i=1}^N \nu'_{ij} M_i = \sum_{i=1}^N \nu''_{ij} M_i \quad (1.12)$$

where M_i is the chemical symbol for i , ν'_{ij} and ν''_{ij} the stoichiometric coefficients, and k_j the rate constant for the j -th reaction. The reaction rate constant is given by:

$$k_j = A_j T^{n_j} \exp\left(-\frac{T_{act,j}}{T}\right) \quad (1.13)$$

The first term on the r.h.s. of Eq. (1.13) is called the *pre-exponential factor* and the second brings in a weak temperature dependence. The exponential term includes the greatest non-linearity of the system and is the main reason why turbulent reacting flows in combustion, which introduce temperature fluctuations, are so difficult to model. For the purposes of illustration of the main idea, we will consider here that the reaction is simply $A_1 + A_2 \rightarrow A_3$ and that S kg of A_2 react with 1 kg of A_1 to give $(1 + S)$ kg of A_3 , which gives a production rate

$$w_1 = -A\rho Y_1 Y_2 \exp\left(-\frac{T_{act}}{T}\right) \quad (1.14)$$

Despite the fact that in reality chemical reactions proceed according to a complex mechanism, the model of Eq. (1.14) is very useful and we will use it to demonstrate what happens in a turbulent reacting flow in the next sub-section.

The ratio between a physical timescale, τ_{phys} , and a chemical timescale, τ_{chem} , is a very important non-dimensional number in reacting flows and is called the *Damköhler number*,

$$\text{Da} = \frac{\tau_{phys}}{\tau_{chem}} \quad (1.15)$$

Various definitions exist depending on the exact choice of these timescales. Often, $\tau_{phys} = L/U$ would be used for the physical timescale with U and L corresponding to turbulence scales, e.g. the characteristic magnitude of the velocity fluctuations and their correlation lengthscale, respectively. An alternative physical timescale is one associated with molecular diffusion and then it is taken as the inverse of the *scalar dissipation rate*, which will be discussed extensively in this book in later chapters due to its unique prominence in turbulent reacting flows.

In most chemical mechanisms in combustion, chemical engineering or the atmosphere, some elementary reactions are very fast, while other are very slow, which causes the problem of *stiffness*, which will be dealt with in Chapter 6. The species can be affected by both fast and slow reactions and hence their chemical timescale can be very tricky to define. For illustration purposes, say we have only the reaction $A + B \rightarrow C$ with a reaction rate constant k . Assume that B has very large concentration relative to A and that both reactants are dilute and their initial concentrations are $[A]_0$ and $[B]_0$. Assume a flow with uniformly distributed reactants and constant temperature and pressure. Then, the governing equation (Eq. 1.4) becomes $d[A]/dt = -k'[A]$ with $k' = k[B]_0$, the solution of which is an exponential decay with time constant $(k[B]_0)^{-1}$. This time constant can serve as a definition of chemical timescale.

If $\text{Da} \ll 1$, the chemistry is very slow compared to fluid mechanical timescales and some simplifications are possible. If $\text{Da} \gg 1$, the chemistry is very fast compared to mixing and a different type of approach is needed. The limits of very fast and very slow chemistry are quite commonly employed in practical calculations mainly because of the simplified nature of the models they allow. However, real chemistries have a range of timescales and the various species of engineering interest may follow very different rates. For example, the combustion itself may be fast, but the pollutant formation slow. Therefore theories for turbulent reacting flows that can treat a wide range of Damköhler numbers are required. Such theories are discussed in Chapters 4 and 5.

1.2.2 Reynolds averaging

Once we have established the main systems of equations governing the instantaneous velocities and scalar concentrations, we may attempt to construct equations for the averaged quantities by a straightforward Reynolds averaging procedure. Hence, if $u_i = \bar{u}_i + u'_i$ and $Y = \bar{Y} + Y'$, where primed quantities are the fluctuations and the overbar denotes the averaged value, the equation for the mean value of the mass fraction Y becomes:

$$\frac{\partial \bar{Y}}{\partial t} + \frac{\partial(\bar{u}_k \bar{Y})}{\partial x_k} = \frac{\partial}{\partial x_k} \left(D \frac{\partial \bar{Y}}{\partial x_k} \right) - \frac{\partial(\overline{u'_i Y'})}{\partial x_i} + \frac{\bar{w}}{\rho} \quad (1.16)$$

In Eq. (1.16), we have assumed constant ρ for ease of presentation. Solution of Eq. (1.16) would give the mean species mass fractions, which are the quantities of main interest from an engineering point of view.

It is evident that the Reynolds averaging procedure brings in a new term, $\overline{u'_i Y'}$, which is the average flux of Y along the i -th direction due to the turbulent velocity fluctuation u'_i . It represents *turbulent diffusion* of Y and requires modelling, which is discussed in Chapter 2. Averaging with density fluctuations can be dealt with by performing Favre or density-weighted averaging; most texts in turbulent combustion describe this well (e.g. [107, 109, 14, 23]) and will be described in more detail in Chapter 5. The main problem is the last term, which is discussed next.

1.2.3 The mean reaction rate

Understanding the nature of the main difficulty with turbulent reacting flows can be achieved by examination of the mean reaction rate \bar{w} , which appears in Eq. (1.16). If $w = A\rho Y_1 Y_2 \exp(-T_{act}/T)$ (Eq. 1.14), then in the general case with density fluctuations,

$$w = \bar{w} + w' = A(\bar{\rho} + \rho')(\bar{Y}_1 + Y'_1)(\bar{Y}_2 + Y'_2) \exp\left(-\frac{T_{act}}{\bar{T} + T'}\right) \quad (1.17)$$

If $T' = 0$, then it is reasonable to assume that also $\rho' = 0$ (for dilute systems) and only a product between the species mass fraction fluctuations appears:

$$\bar{w} = A\bar{\rho} \left(\bar{Y}_1 \bar{Y}_2 + \overline{Y'_1 Y'_2} \right) \exp\left(-\frac{T_{act}}{\bar{T}}\right) \quad (1.18)$$

Hence, the mean reaction rate is not given by a straightforward application of the reaction rate expression in Eq. (1.14) evaluated at the averaged mass fractions and temperature. We may wish to view w as a deterministic

function of the random variables Y_1 , Y_2 , ρ and T . The above illustration is a manifestation of a more general result for non-linear functions of random variables: in general, *the mean of the function is not equal to the function of the mean*.

Therefore, to calculate the mean reaction rate, one needs a model for $\overline{Y_1'Y_2'}$. Efforts to predict this correlation will be described in later chapters and have met some success for isothermal flows in the environment or in chemical engineering. In turbulent combustion, where $T' \neq 0$ and $\rho' \neq 0$, additional correlations appear in Eq. (1.18) involving the temperature and the density. To illustrate the difficulty involved in the averaging of the exponential by the presence of the temperature fluctuations, Bilger [14] presents a series expansion about \overline{T} . It is shown that this converges only for very small temperature fluctuations and hence such an expansion is unusable. Theories for turbulent combustion have invented various methods of by-passing the problem of directly evaluating the average value of the non-linear exponential term and it is for this reason that there is a very active research community dealing with these problems and a vast literature. Finding our way through this literature is one of our objectives.

1.3 Summary

The time, ensemble, or volume-averaged equations for the mass fractions of species and temperature in a turbulent reacting flow include higher moments. The turbulent flux (and Reynolds stress) closure is the topic of turbulence modelling. Closure for the averaged chemical source term is the remit of turbulent reacting flows theories. For a second-order reaction, the mean reaction rate includes the term $\overline{Y_1'Y_2'}$, which must be modelled. For isothermal problems, modelling of this correlation may be possible, but this is not the case for turbulent combustion. This book presents some of the theories that have been put in place to address these issues.

1.4 Exercises

- (1) Calculate the percentage change of the reaction rate w if the temperature T changes from 1000 K to 1050 K for $T_{act} = 20000$ K, a typical activation temperature for combustion.
- (2) This problem is directly relevant to Large Eddy Simulations of atmo-

spheric reacting flows and illustrates the modelling difficulties in turbulent flows with thin reaction zones. It also illustrates how averaging may give the wrong value of the reaction rate. A certain volume V contains O_3 and NO . Assume that the reaction is $NO + O_3 \rightarrow NO_2 + O_2$ and the reaction rate constant is k . One half of V has C_{NO} and no O_3 and the other half has C_{O_3} and no NO , i.e. the species do not overlap anywhere in V . What is the true volume-averaged reaction rate? What is the reaction rate calculated from the volume-averaged NO and O_3 ?

- (3) Assuming small T'/\bar{T} , expand the exponential in Eq. (1.17) and then perform the averaging keeping up to second-order terms. For simplicity, assume negligible fluctuations of the species mass fractions and the density.