

Towards regime identification and appropriate chemistry tabulation for computation of autoigniting turbulent reacting flows

By M. Kostka, E. Knudsen AND H. Pitsch

1. Motivation and objective

Many technical devices such as internal combustion engines, gas turbines, furnaces and the like burn hydrocarbon fuels to generate heat and power. To increase efficiency and reduce residence time, flow rates are increased, which causes the flow field to become turbulent, with the added benefit of enhanced mixing.

Following the trend to carry out the bulk of prototype testing computationally, engineers need accurate and reliable, yet computationally feasible, models. However, the accurate prediction of turbulent combustion is a complex problem. Even relatively simple fuels, such as hydrogen or methane, involve dozens of species and hundreds of elementary reactions during the conversion of fuel and oxidizer to products. Therefore, approaches have been developed that describe chemistry in terms of a reduced set of scalars and map the thermochemical quantities based on those.

These mappings often stem from the solution to an asymptotically valid problem in the limit of diffusion flame burning or premixed flame propagation. However, at the operating point of a technical device it is rarely the case that a single asymptotic limit describes all of the chemical behavior in the flow.

Knudsen & Pitsch (2009) have recently proposed a transformation of the transport equation of a suitably defined progress variable, where different budget terms are examined and groups of terms are identified to represent distinct asymptotic limits of combustion physics. Introducing this method, Knudsen & Pitsch (2009) focus on the distinction between non-premixed burning and steady premixed flame propagation.

The present brief outlines the application of the transformation to include information about unsteady effects, which are hoped to assist in the prediction of inherently unsteady combustion scenarios such as autoigniting mixtures.

The vitiated coflow burner investigated by Cabra *et al.* (2005) is chosen as a test case for this approach. This case's behavior is largely governed by autoignition and should provide an accurate assessment of the performance of the proposed methodology.

2. Transformation

Flamelet models for turbulent combustion transform scalar transport equations from physical space to a coordinate system which is attached to isosurfaces of mixture fraction (Peters 1984, 1986, 2000). After neglecting the two directions tangential to the isosurface and retaining the normal direction the familiar reaction-diffusion equation is obtained,

$$\rho \frac{\partial \phi_i}{\partial t} - \rho \frac{\chi}{2} \frac{\partial^2 \phi_i}{\partial Z^2} = \dot{\omega}_i. \quad (2.1)$$

This equation describes combustion in the limits of diffusion burning where, in the steady case in which $\frac{\partial \phi_i}{\partial t} = 0$, diffusion in the direction of the mixture fraction gradient balances the chemical source term.

The application of a similar transformation that uses two independent coordinates Z and Λ makes it possible to recover the asymptotic equations for diffusion burning, premixed propagation and unsteady ignition all from one common equation.

The parameter Λ is called the flamelet index and is chosen such that it describes deviations from a flamelet solution. It therefore is a quantity that describes the progress of reaction, from an unburned state to chemical equilibrium. Yet, Λ has to be statistically independent of the mixture fraction. For this reason, following Ihme *et al.* (2005), Pitsch & Ihme (2005) and Ihme & Pitsch (2005), it can be taken to be the value of any quantity uniquely parametrizing the reaction progress at a fixed point in mixture fraction space. Thus, for the remainder of this work it is taken to be

$$\Lambda = C(Z_{st}, T_{st}^*), \quad (2.2)$$

where the subscript st represents the stoichiometric point in mixture fraction space and T_{st}^* denotes one specific solution to the flamelet equations.

In the case of methane combustion it was found that $C = Y_{CO} + Y_{CO_2} + Y_{H_2O} + Y_{H_2}$ is a viable choice (Ihme 2007) for the progress variable. This is the definition of progress variable that will be used throughout this work.

Now, using simple transformation rules and some assumption about the curvature of (Z, Λ) space, the transport equation for a generic scalar ϕ_i

$$\rho \frac{\partial \phi_i}{\partial t} + \nabla \rho \mathbf{u} \cdot \phi_i = \nabla \cdot (\rho \mathcal{D} \nabla \phi_i) + \dot{\omega}_i \quad (2.3)$$

is transformed into

$$\begin{aligned} & \rho \frac{\partial \phi_i}{\partial \tau} + \frac{\partial \phi_i}{\partial \Lambda} \left[\rho \frac{\partial \Lambda}{\partial t} + \rho \mathbf{u} \cdot \nabla \Lambda - \nabla \cdot (\rho \mathcal{D} \nabla \Lambda) \right] \\ &= \rho \frac{\chi_Z}{2} \frac{\partial^2 \phi_i}{\partial Z^2} + \rho \frac{\chi_\Lambda}{2} \frac{\partial^2 \phi_i}{\partial \Lambda^2} + \rho \chi_{Z,\Lambda} \frac{\partial^2 \phi_i}{\partial Z \partial \Lambda} + \dot{\omega}_i. \end{aligned} \quad (2.4)$$

Setting $\phi_i = C$ provides an equation for the progress variable in terms of two statistically independent parameters, Z and Λ ,

$$\begin{aligned} & \rho \frac{\partial C}{\partial \tau} + \frac{\partial C}{\partial \Lambda} \left[\rho \frac{\partial \Lambda}{\partial t} + \rho \mathbf{u} \cdot \nabla \Lambda - \nabla \cdot (\rho \mathcal{D} \nabla \Lambda) \right] \\ &= \rho \frac{\chi_Z}{2} \frac{\partial^2 C}{\partial Z^2} + \rho \frac{\chi_\Lambda}{2} \frac{\partial^2 C}{\partial \Lambda^2} + \rho \chi_{Z,\Lambda} \frac{\partial^2 C}{\partial Z \partial \Lambda} + \dot{\omega}_C. \end{aligned} \quad (2.5)$$

Equation (2.5) contains three distinct limits of combustion physics: diffusion burning (or non-premixed combustion), premixed flame propagation and purely unsteady, 0-dimensional scenarios such as autoignition.

The 1-D steady premixed flame equations can be recovered from equation (2.5) by considering a scenario where $Z = Z_{st}$. Under this condition $\Lambda = C$, and the burning velocity s_L is expected to appear as a parameter. Adding zero to equation (2.5) as $(\rho s_L \mathbf{n} \cdot \nabla \Lambda) - (\rho s_L \mathbf{n} \cdot \nabla \Lambda)$, and recognizing that the cross dissipation term and the pseudo-temporal term are zero under steady premixed conditions, makes it possible to group terms of the resulting equation based on association with one of the three asymptotic limits. For details regarding the simplifying assumptions in this step the reader is referred to the original publication of Knudsen & Pitsch (2009).

The final form of the transformed equation is

$$\begin{aligned} & \left(\frac{\partial C}{\partial \Lambda} \left[\rho \frac{\partial \Lambda}{\partial t} + (\rho \mathbf{u} - \rho_u s_{L,u} \mathbf{n}) \cdot \nabla \Lambda \right] \right)_1 \\ & + \left(\frac{\partial C}{\partial \Lambda} [\rho_u s_{L,u} |\nabla \Lambda| - \nabla \cdot (\rho \mathcal{D} \nabla \Lambda)] - \rho \frac{\chi_\Lambda}{2} \frac{\partial^2 C}{\partial \Lambda^2} \right)_2 \\ & + \left(-\rho \frac{\chi_Z}{2} \frac{\partial^2 C}{\partial Z^2} \right)_3 = \dot{\omega}_C. \end{aligned} \quad (2.6)$$

Here, group 1 describes unsteady physics, group 2 describes steady premixed physics and group 3 is associated with steady non-premixed combustion.

Equation (2.6) suggests a means of locally evaluating the most appropriate combustion regime, such that a well-informed decision can be made regarding the type of tabulated chemistry. For example, in cases where steady physics describe the local conditions well, a ratio of the grouped terms in equation (2.6),

$$\Theta_{1,23} = \frac{\frac{\partial C}{\partial \Lambda} \left[\rho \frac{\partial \Lambda}{\partial t} + (\rho \mathbf{u} - \rho_u s_{L,u} \mathbf{n}) \cdot \nabla \Lambda \right]}{\max \left(\frac{\partial C}{\partial \Lambda} [\rho_u s_{L,u} |\nabla \Lambda| - \nabla \cdot (\rho \mathcal{D} \nabla \Lambda)] - \rho \frac{\chi_\Lambda}{2} \frac{\partial^2 C}{\partial \Lambda^2}, -\rho \frac{\chi_Z}{2} \frac{\partial^2 C}{\partial Z^2} \right)}, \quad (2.7)$$

should fulfill $\Theta_{1,23} \ll 1$ and would indicate that closures based on steady physics assumptions such as steady flamelets or laminar flames are appropriate. Once the distinction between the unsteady and steady limit has been made, the ratio

$$\Theta_{2,3} = \frac{\frac{\partial C}{\partial \Lambda} [\rho_u s_{L,u} |\nabla \Lambda| - \nabla \cdot (\rho \mathcal{D} \nabla \Lambda)] - \rho \frac{\chi_\Lambda}{2} \frac{\partial^2 C}{\partial \Lambda^2}}{-\rho \frac{\chi_Z}{2} \frac{\partial^2 C}{\partial Z^2}} \quad (2.8)$$

can be used as an indicator of whether non-premixed or premixed combustion dominates the local flow field. In the initial application of the regime indexing procedure, Knudsen & Pitsch (2009) assumed *a priori* that unsteady effects were negligible for their test cases and evaluated groups 2 and 3 to decide whether to locally use a chemistry tabulation based on steady non-premixed flamelets or on laminar premixed flame solutions. They showed that this distinction was capable of accurately predicting the physics of a turbulent swirled flame.

3. Unsteady regime

Values of $\Theta_{1,23} \ll 1$ indicate a balance between either of the groups 2 or 3 and the source term $\dot{\omega}_C$. This means that the assumption of steady physics should be justified. However, if this condition does not hold, ignition, extinction or other unsteady phenomena are important and should be treated with care. In regions where this parameter is large, a chemistry tabulation based on the solution to steady-state equations should not be used. Instead, solutions to the 0-D, perfectly stirred reactor equations, for example, could be used to determine the state of the chemical system including chemical source terms.

The aim of the present work is to include the evaluation of the ratio $\Theta_{1,23}$ into the procedure. Thus, values of $\Theta_{1,23} > 1$ indicate that locally in the flow field, unsteady effects dominate the balance of the transport terms in equation (2.6) with the chemical source term $\dot{\omega}_C$.

Unsteadiness per se, however, cannot directly be associated with autoignition. The grouping of terms in equation (2.6) identified terms associated with the steady-state

limits of non-premixed and premixed combustion, respectively. Thus, the unsteady term might contain information about processes that are unsteady but deviate only little from the steady state of that particular burning mode. In order to try and separate the autoignition regime from this general unsteadiness, an additional criterion is used.

Consider the reduced description of a hydrocarbon fuel. If this description contains the kinetics of radical production and consumption, there exists a point, more specifically a temperature, at which the chain branching elementary reaction becomes faster than the chain breaking one. This means that above this point, combustion can sustain itself, because the production of radicals necessary for chemical conversion exceeds the consumption of those species. Below this point, self-sustained burning is not possible. Thus, it seems like a condition which can be exploited to separate regions of unsteady flame dynamics from autoigniting ones. However, the crossover temperature is a quantity only clearly defined in the context of reduced kinetics.

For the purpose of identifying a quasi-crossover point in the context of comprehensive chemistry, we plan to use a criterion based on the progress variable source term as given by perfectly stirred reactor (PSR) calculations. Since the crossover temperature coincides with the inner-layer temperature for laminar premixed flames (Peters 2000), which is where the heat release rate peaks, we translate this into the value of the progress variable at which its source term peaks. This value shall then be taken as an indicator of when self-sustained combustion is possible and a flame is established and, conversely, when autoignition is still governing the chemistry.

4. Chemistry tabulation for the autoignition regime

4.1. Perfectly stirred reactor calculations

Asymptotic mappings for the regimes of non-premixed and premixed combustion based on steady non-premixed flamelet solutions or laminar premixed flame solutions have proven quite successful. Yet, due to the steady nature of these asymptotic limits they are not suitable for predicting a mixture undergoing autoignition. For this reason we conduct PSR calculations, spanning the range of equivalence ratios defined by the limits of mixture fraction for a particular problem. The equation governing a PSR is

$$\rho \frac{\partial \phi_i}{\partial t} = \omega_i, \quad (4.1)$$

where $\phi_i = \{Y_i, h\}$ and the initial conditions are imposed as

$$\phi_i^{t=0}(Z) = \phi_{i,Oxidizer} + Z \cdot (\phi_{i,Fuel} - \phi_{i,Oxidizer}). \quad (4.2)$$

This gives a set of thermochemical quantities

$$\{\rho, Y_i, \omega_i, T\} = \mathcal{M}(Z, t). \quad (4.3)$$

Subsequently, this mapping \mathcal{M} is postprocessed to construct the progress variable from Y_i , as well as the progress variable source term as $\dot{\omega}_C = \dot{\omega}_{CO} + \dot{\omega}_{CO_2} + \dot{\omega}_{H_2O} + \dot{\omega}_{H_2}$. With this, \mathcal{M} can be parametrized by the mixture fraction and progress variable, instead of mixture fraction and time, $\mathcal{M}(Z, t) \rightarrow \mathcal{M}(Z, C)$, which fits neatly into the framework of the flamelet-progress variable approach (FPVA) by Pierce & Moin (2004).

Although autoignition within a turbulent flow field at varying local equivalence ratio is a complex process that involves the effects of turbulence and diffusion, the PSR calcu-

lations include the kinetic dynamics at low temperature, which governs autoignition and therefore seems like a natural choice for such a mapping.

4.2. Convolution with the assumed shape subgrid-scale pdf

In conserved scalar approaches for large eddy simulation (LES) of turbulent combustion, chemical source term closure is performed by convoluting the thermochemical quantities with an assumed shape subgrid-scale probability density function (pdf). For two stream problems, a β -function has proven to be a reasonable approximation of the conserved scalar subgrid-scale pdf.

When such a β -function is used, any thermochemical quantity at the filtered LES grid level, $\tilde{\phi}$, is defined as

$$\tilde{\phi} = \int_0^1 \phi(Z)P(Z)dZ, \quad (4.4)$$

with the pdf as (Cook & Riley 1994)

$$P(Z) = \frac{Z^{a-1}(1-Z)^{(b-1)}}{\int_0^1 \gamma^{a-1}(1-\gamma)^{b-1}d\gamma}, \quad (4.5)$$

with

$$a = \tilde{Z} \left(\frac{\tilde{Z}(1-\tilde{Z})}{\tilde{Z}''^2} - 1 \right) \quad (4.6)$$

$$b = (a/\tilde{Z}) - a. \quad (4.7)$$

This practice is straightforward as long as the conserved scalar is the only independent variable used for parameterization of chemistry. If additional quantities such as a progress variable are used for parameterization, however, it is necessary to ensure adequate statistical independence of the individual independent variable such that established assumed shape pdf's for the conserved scalar can still be invoked.

For the case of non-premixed combustion this is fairly straightforward. The particular choice of Λ , as mentioned in Section 2, ensures its statistical independence from mixture fraction. Therefore, equation (4.4) is evaluated for each individual flamelet solution one wishes to use to compile the chemistry tabulation.

If the solutions to PSR calculations are to be used for tabulation, matters are complicated by the fact that there is no physical process to link two solutions adjacent to each other in mixture fraction space. Hence, a question arises regarding how convolution with the pdf in a local LES control volume should be performed.

Some authors (Domingo *et al.* (2007); Veynante & Vervisch (2002); Vervisch *et al.* (2004)) have suggested scaling the progress variable at a given equivalence ratio with its equilibrium value. They showed reasonable agreement of this assumption with the findings of a direct numerical simulation (DNS).

If, however, one considers the mixture within a given LES control volume and its history in a Lagrangian manner, it seems more natural to assume a common residence time for this mixture. This means the convolution should be carried out over a range of mixtures characterized by a constant residence time, t .

Given the mapping $\mathcal{M}(Z, C)$ for each coordinate of the mapping (Z^*, C^*) , the subset \mathcal{S} of \mathcal{M} is sought which satisfies

$$\mathcal{S}_{\mathbb{Q}(Z^*, C^*)}(Z) = \langle \mathcal{M}(Z, C) | t = t_{C^*} \rangle, \quad (4.8)$$

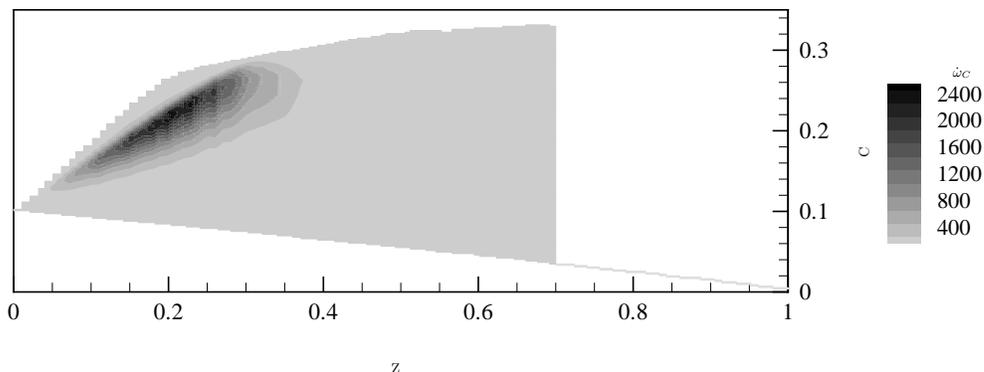


FIGURE 1. PSR-based chemistry tabulation, progress variable source term, $\dot{\omega}_C$

where $\langle \bullet | condition \rangle$ denotes sampling values of \bullet , for which *condition* holds.

Subsequently, the convolution is carried out,

$$\tilde{\phi}_i(\tilde{Z}, \tilde{C}, \tilde{Z}'^2) = \int_0^1 \mathcal{S}_{@(\tilde{Z}^*, \tilde{C}^*)}(Z) P(Z) dZ, \quad (4.9)$$

and the dimension of conserved scalar variance \tilde{Z}'^2 is therefore added to the chemistry tabulation, \mathcal{M} . Currently, the conditional pdf of progress variable is assumed to be a Dirac- δ function. This is a strong but natural assumption for an initial assessment of the present model.

Figure 1 shows the resulting map of progress variable source term, $\dot{\omega}_C$, for vanishing mixture fraction variance. In the very rich part of the flame, values are linearly interpolated towards the values of pure fuel and $\dot{\omega}_C$ is neglected, since for rich mixtures autoignition is an extremely slow process.

5. Results

The vitiated coflow burner studied by Cabra *et al.* (2005) was chosen as a test case for the present method. They investigated a lifted flame formed by a methane jet, issuing from a nozzle into a flow of hot combustion products. The coflow was generated by about 2200 lean H₂-air flames. The coflow served to mimic the effect of exhaust gas recirculation (EGR) or of a similar technique, in which combustion products are at least partially fed back into the burner to increase efficiency.

5.1. Boundary conditions

Conditions for the mixture composition in the fuel and oxidizer stream, respectively, were taken from Cabra *et al.* (2005). However, since the information they provide does not satisfy mass conservation, meaning the necessary condition $\sum_i X_i = 1$ is violated, the amount of nitrogen was adjusted to compensate for this. Also, components given with a concentration of less than one ppm were neglected in the boundary conditions. The resulting conditions are summarized in Table 1.

	Re	d [mm]	V [m/s]	T [K]	X _{O₂}	X _{N₂}	X _{H₂O}	X _{OH}	X _{H₂}	X _{CH₄}
Jet	28,000	4.57	100.0	320	0.15	0.517	0.0029	0	1×10^{-4}	0.33
Coflow	23,300	210.0	5.4	1350	0.12	0.7294	0.15	2×10^{-4}	1×10^{-4}	0.0003

TABLE 1. Conditions and specifications for the vitiated methane-air flame

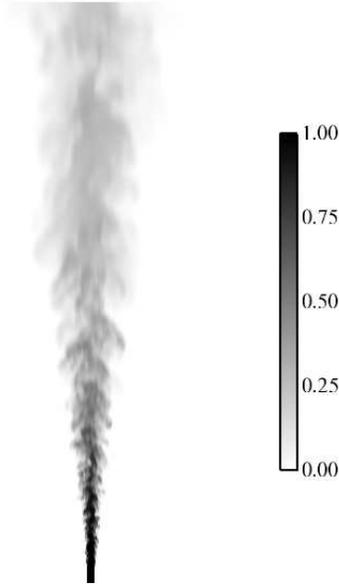


FIGURE 2. Instantaneous field of mixture fraction

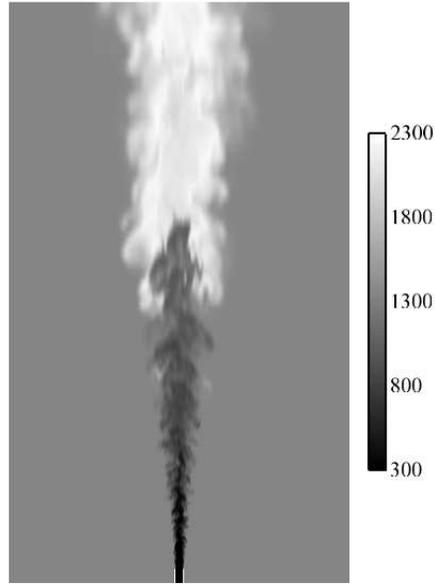


FIGURE 3. Instantaneous field of temperature

5.2. Assessment of autoignition tabulation

Early exploratory studies of this configuration, using chemistry tabulation based solely on steady diffusion flamelets or the steady regime indexing procedure as used by Knudsen & Pitsch (2009), showed a consistent trend to significantly underpredict the lift-off height of the flame, meaning that the flame base is predicted to be too close to the nozzle.

Based on this experience, the source term of the progress variable provided by the autoignition tabulation should yield a flame at least as far away from the nozzle as seen in the experiments. Otherwise, even a physically informed blending of those tabulations is unlikely to be able to predict the flame lift-off accurately. Therefore, a first step is a calculation of the flame with the progress variable source term from the autoignition table only, in order to assess the performance of the tabulation and convolution procedure in conjunction with a model for the subgrid-scale variance of mixture fraction. Figures 2-5 show some snapshots of this calculation. Once this limiting case is established, a blending based on the proposed regime indexing procedure will be assessed and developed further.

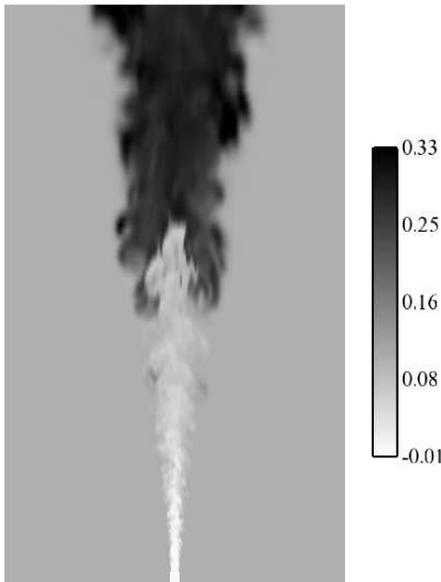


FIGURE 4. Instantaneous field of progress variable

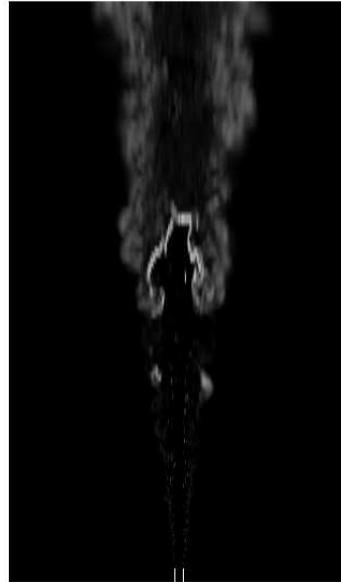


FIGURE 5. Instantaneous field of progress variable source term

6. Future work

As outlined, the goal is to develop a reliable model to use in predicting complex multi-physics combustion processes.

Once the autoignition tabulation based on PSR calculations is verified, we plan to proceed with using the regime indexing procedure as proposed to assess its performance and improve on its formulation.

Acknowledgments

This work is funded by NASA and their financial support is gratefully acknowledged.

REFERENCES

- CABRA, R., CHEN, J.-Y., DIBBLE, R., KARPETIS, A. & BARLOW, R. 2005 Lifted methane-air jet flames in a vitiated coflow. *Combustion and Flame* **143**, 491–506.
- COOK, A. & RILEY, J. 1994 A subgrid model for equilibrium chemistry in turbulent flows. *Physics of Fluids* **6** (8), 2868–2870.
- DOMINGO, P., VERVISCH, L. & VEYNANTE, D. 2007 Large-eddy simulation of a lifted methane jet flame in a vitiated coflow. *Combustion and Flame* **152**, 415–432.
- IHME, M. 2007 Pollutant formation and noise emission in turbulent non-premixed flames. PhD thesis, Stanford University.
- IHME, M., CHA, C. & PITSCH, H. 2005 Prediction of local extinction and re-ignition

- effects in non-premixed turbulent combustion using a flamelet/progress variable approach. *Proceedings of the Combustion Institute* **30**, 793–800.
- IHME, M. & PITSCH, H. 2005 LES of a non-premixed flame using an extended flamelet/progress variable model. In *Proceedings of the 43rd AIAA Aerospace Sciences Meeting and Exhibit*. AIAA.
- KNUDSEN, E. & PITSCH, H. 2009 A general flamelet transformation useful for distinguishing between premixed and non-premixed modes of combustion. *Journal of Computational Physics* **156**, 678–696.
- PETERS, N. 1984 Laminar diffusion flamelet models in non-premixed turbulent combustion. *Progress in Energy and Combustion Science* **10**, 319–339.
- PETERS, N. 1986 Laminar flamelet concepts in turbulent combustion. *Twenty-first Symposium (International) on Combustion* pp. 1231–1250.
- PETERS, N. 2000 *Turbulent Combustion*. Cambridge University Press.
- PIERCE, C. & MOIN, P. 2004 Progress-variable approach for large-eddy simulation of non-premixed turbulent combustion. *Journal of Fluid Mechanics* **504**, 73–97.
- PITSCH, H. & IHME, M. 2005 An unsteady flamelet/progress variable model for LES of nonpremixed turbulent combustion. In *Proceedings of the 43rd AIAA Aerospace Sciences Meeting and Exhibit*. AIAA.
- VERVISCH, L., HAUGUEL, R., DOMINGO, P. & RULLAUD, M. 2004 Three facets of turbulent combustion modelling: DNS of premixed V-Flame, LES of lifted nonpremixed flame and RANS of jet-flame. *Journal of Turbulence* **5** (4).
- VEYNANTE, D. & VERVISCH, L. 2002 Turbulent combustion modeling. *Progress in Energy and Combustion Science* **28**, 193–266.