

Hybrid transported-tabulated strategy to downsize detailed chemistry for Large Eddy Simulation

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A strategy to bridge the gap between hydrocarbon combustion detailed chemistry and Large Eddy Simulation (LES) of burners is reported. Significant progress has been made recently in terms of accuracy and robustness in both chemical kinetics and flow computations. However, the highest resolution reached in LES of practical burners does not yet ensure that the response of intermediate radical species is fully captured. In the method discussed, the full set of species and elementary reaction rates of the detailed mechanism are kept, but only species featuring non-zero concentration in fresh and burnt gases are transported with the flow. Intermediate chemical species, developing within thin flame layers, are expressed resorting to their self-similar properties observed in a series of canonical combustion problems. The method is tested with success in various adiabatic or non-adiabatic laminar and strained flames.

1. Introduction

Developing tools for accurately simulating ignition, flame stability, quenching and pollution implies introducing detailed chemistry into the modeling loop. Indeed, oversimplified descriptions of the chemical reactions using global or reduced chemical schemes are known to lack generic character and to strongly reduce prediction capabilities (Bilger *et al.* 2005). Along these lines, many methods have been discussed in the literature to downsize large hydrocarbon chemical schemes, through the systematic tabulation of chemical responses from canonical combustion problems; typically, the chemical response of one-dimensional laminar flames is projected into a progress variable space to build a look-up table (Gicquel *et al.* 2000; van Oijen *et al.* 2001; Nguyen *et al.* 2010; Lodier *et al.* 2011), and only this progress variable is transported with the flow.

This approach derives from the flamelet hypothesis (Bray 1996; Bradley *et al.* 1988; Peters 2000) and it has been successful in Large Eddy Simulation (LES) for reproducing the major properties of burners (Pierce & Moin 2004; Ihme *et al.* 2005; Domingo *et al.* 2008; Subramanian *et al.* 2010). However, each chemical look-up table is hooked up to one set of boundary conditions in terms of fuel and oxidizer compositions. Dilution by burnt gases and heat losses are difficult to introduce because they drastically increase the size of the tables to be stored; this leads to huge look-up tables that are not well adapted to high performance computing.

An alternative is explored in this work, whereby the detailed chemical scheme is kept unaltered; nonetheless, the number of species transported is reduced by expressing intermediates from a transformation grounded on their self-similar behavior observed in all canonical problems used so far for generating look-up tables. The outcome is a method

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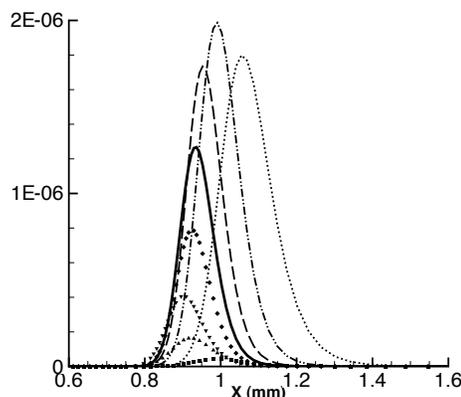


FIGURE 1. CH mass fraction versus physical space. Equivalence ratio (ϕ): ■ 0.6, ▲ 0.7, ▼ 0.8, ◆ 0.9, — 1.0, - - - 1.1, - · - 1.2, · · · 1.3.

preserving most of the features of the detailed chemical scheme, with resolution needs reduced by at least an order of magnitude.

2. Transported-tabulated chemistry

2.1. Self-similar behavior of minor species

First, note that many intermediate species play a crucial role in the thin reaction zone where heat is released, but not in calculating the composition and temperature just downstream of these very thin layers, where radical concentrations actually vanish as for instance, in the relaxation towards chemical equilibrium, which mainly depends on major species.

To preserve most of the generic character of detailed chemical schemes but downsize the size of the problem, it would be convenient to dynamically express minor species from major ones during the simulation. This is achieved by using the self-similar flame theory (Ribert *et al.* 2006), studied during previous CTR summer programs (Naudin *et al.* 2006; Veynante *et al.* 2008), and recently examined further for diluted flames with heat loss and cool-flame effects (Wang *et al.* 2010; Ribert *et al.* 2012). Here it is extended to flame inner reaction zones, controlled by intermediate and radical species.

The detailed chemistry mechanism developed by Lindstedt (1998) and Juchmann *et al.* (1998) (29 chemical species, 282 elementary reaction steps) is used for methane/air combustion under the hypothesis of unity Lewis number transport, usually formulated in chemistry tabulation in view of turbulent flame simulations. Various canonical flame problems generally used to construct chemical look-up tables are considered: freely propagating, or strained (steady and unsteady) fresh-against-burnt one-dimensional premixed flames, zero-dimensional auto-ignition and one-dimensional strained diffusion flames. The laminar flame solver REGATH (Real Gas THERmodynamics) is used (Candel *et al.* 2011).

It was found that minor species within thin reaction zones also feature a self-similar behavior, as temperature and major species do. Figure 1 shows the CH radical response in physical space for various equivalence ratios in laminar unstrained premixed flames. Every profile of such j -th intermediate species may be normalized by its maximum

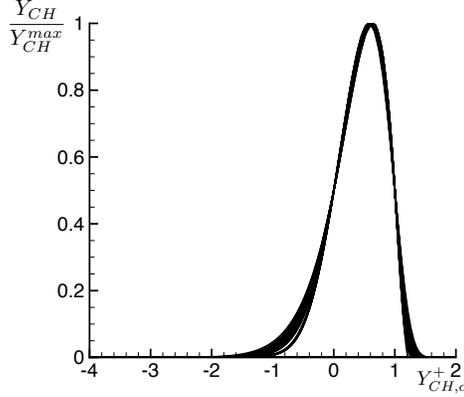


FIGURE 2. Reduced CH radical mass fraction, $Y_{CH}^+ = (Y_{CH})/(Y_{CH}^{max})$ versus $Y_{CH,c}^+$ (Eq. (2.1)) for 18 canonical combustion problems: laminar premixed ($0.6 \leq \phi \leq 1.3$), laminar strained premixed ($\phi = 0.9$, $120 \leq a_T \leq 6500 \text{ s}^{-1}$), diffusion flames ($80 \leq a_T \leq a_T^q \text{ s}^{-1}$) and zero-dimensional self-ignition ($10 \leq P_0 \leq 20 \text{ atm}$, $0.8 \leq \phi \leq 1.2$, $900 \leq T_0 \leq 1300 \text{ K}$).

$Y_j^+ = Y_j / Y_j^{max}$ and examined in a reduced composition space:

$$Y_{j,c}^+(\phi) = \frac{Y_c - Y_{j,c}^A(\phi)}{Y_{j,c}^B(\phi) - Y_{j,c}^A(\phi)}, \quad (2.1)$$

where $Y_{j,c}^A(\phi)$ and $Y_{j,c}^B(\phi)$ are the two values of the progress variable Y_c when $Y_j/Y_k^{max} = 0.5$. The parameter ϕ denotes the equivalence ratio of the mixture, and the progress variable is defined as $Y_c = Y_{CO} + Y_{CO_2}$. The response of Y_{CH}^+ versus Y_c^+ for the major canonical combustion problems considered is shown in Figure 2. All the CH distributions in both time and physical space can be approximated by a single response when analyzed in reduced coordinates. A systematic study of intermediate species distribution was conducted for CH4/Air combustion. Two main groups of self-similar response emerge, as shown in Figures 3 and 4.

Thus, within intense reaction zones, minor species can be approximated in a self-similar manner as

$$Y_j = Y_j^{max}(\phi) \times Y_j^+(Y_{j,c}^+(\phi)). \quad (2.2)$$

Depending on the degree of precision required, $Y_j^+(Y_{j,c}^+(\phi))$ may be determined for every radical or a single generic form may be used for a given group of species. The parameters $Y_{j,c}^A(\phi)$ and $Y_{j,c}^B(\phi)$ of Eq. (2.1) must also be known. From the database of canonical problems, expressions for these parameters have been optimized and are given in Tables 2, 3 and 4. Figure 5 shows the comparison between these expressions and the exact values found in the reference database.

From this analysis, a set \mathcal{B} of species which do not need to be transported with the flow is found and given in Table 1. The size of this set \mathcal{B} is directly linked to the precision actually required in the flame simulations. In other words, more species may be tabulated if only major species need to be reproduced in the burner simulation.

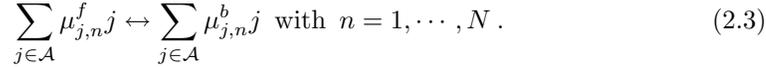
2.2. HTTC formalism

The practical use of the relation (2.2) in the solution of detailed chemistry is now discussed. Let us consider a mixture that involves a set \mathcal{A} of distinct chemical species j ,

TABLE 2. Coefficients for $Y_{k,c}^A = a\phi^3 + b\phi^2 + c\phi + d$

Species	a	b	c	d
CH	0.0	0.0	0.0845	0.0214
CH ₂ (S)	0.0	1.9942163E-2	3.918688E-2	3.8169E-2
CH ₂	0.0	4.225742E-2	-6.7411432E-3	5.60958932E-2
CH ₃ O	1.7968216E-1	-3.5179283E-1	1.813892E-1	0.02333
CH ₂ OH	0.0	0.04183293	-0.012457212	0.0588273764
C ₂ H	0.0	-0.001360763	0.08458736	0.0233338
HCCO	0.0	-0.0193	0.112	0.013
C ₂ H ₂	0.0	0.0	4.22905489E-2	1.106794E-2
CH ₂ CO	1.321594E-1	-2.9526713E-1	2.213232E-1	-2.81894E-2
C ₂ H ₃	0.0	0.0	6.35226E-2	3.0928379E-2
C ₂ H ₄	0.0	0.0	2.711796E-2	9.68501E-3
C ₂ H ₅	0.0	1.00437484E-1	-1.4992E-1	1.07634345E-1
C ₂ H ₆	0.0	2.565784E-2	-3.346162E-2	3.285254E-2
C	0.0	1.84E-3	0.08755	0.02254
C ₂	0.0	0.0	0.08766	0.02431

interacting in a detailed chemical scheme according to N elementary reactions:



$\mu_{j,n}^f$ (resp. $\mu_{j,n}^b$) is the forward (resp. backward) stoichiometric coefficient of the n^{th} reaction rate, which may be written

$$\dot{\omega}_n = \prod_{j \in \mathcal{A}} \left(\frac{\rho Y_j}{W_j} \right)^{\mu_{j,n}^{j,n}} A_n T^{\nu_n} \exp(-T_{A,n}/T), \quad (2.4)$$

where the usual notations have been introduced (Williams 1985). The overall reaction rate of the j -th species reads

$$\dot{\omega}_j = W_j \sum_{n=1}^N \left(\mu_{j,n}^b - \mu_{j,n}^f \right) \dot{\omega}_n. \quad (2.5)$$

\mathcal{B} (Table 1) is the subset composed of intermediate species expressed with relation (2.2). In the calculation of elementary burning rates (2.4), if $j \in \mathcal{B}$, then $Y_j = Y_j^{\max}(\phi) \times Y_j^+(Y_{j,c}^+(\phi))$. The full chemical mechanism is explicitly used to compute burning rates (2.5) of the remaining set of species $\mathcal{A}-\mathcal{B}$ that are transported with the flow. The species of set \mathcal{B} need no longer to be transported; still, the full chemical mechanism is explicitly operating to solve the reactive flow problem.

3. Results

The Hybrid Transported-Tabulated Chemistry (HTTC) approach is tested against the full transported chemistry for various premixed flames. First, the spatial resolution needed for full chemistry transport is retained, to be decreased down to ten times coarser within the flame zone (see Figure 6). The chemical flame structure is fully reproduced when decreasing the mesh resolution, despite a small deviation of the CH mass fraction on the burnt side.

TABLE 3. Coefficients for $Y_{k,c}^B = a\phi^4 + b\phi^3 + c\phi^2 + d\phi + e$

Species	a	b	c	d	e
CH	0.0	0.0	-0.0274	0.1479	0.0049
CH ₂ (S)	0.0	0.0	-2.471478E-2	1.42096415E-1	6.33512E-3
CH ₂	0.0	0.0	-1.75764E-2	1.292931E-1	1.05477E-2
CH ₃ O	0.0	0.0	0.04536545	2.3985576E-4	0.057465564
CH ₂ OH	0.0	0.0	-0.02028697	0.1324558	0.009652014
C ₂ H	0.0	0.0	-0.03012222	0.1526458	0.00392675
HCCO	0.0	0.0	-0.03358	0.1526458	0.00392675
C ₂ H ₂	0.0	0.0	-1.9518272E-2	1.3055479E-1	1.0543782E-2
CH ₂ CO	6.954615E-2	-3.93172E-1	7.21385E-1	-4.2987445E-1	1.510963E-1
C ₂ H ₃	0.0	0.0	-2.92186788E-2	1.43572433E-1	7.25358E-3
C ₂ H ₄	0.0	0.0	-2.1346288E-2	1.205908E-1	1.230619E-2
C ₂ H ₅	0.0	0.0	0.0	7.9415492E-2	2.881E-2
C ₂ H ₆	0.0	0.0	1.20361515E-2	4.20056E-2	3.979535E-2
C	0.0	0.0	-0.0316	0.1571	0.0016
C ₂	0.0	0.0	-0.0292	0.1516	0.0048

TABLE 4. Coefficients for $\max(Y_k) = a\phi^5 + b\phi^4 + c\phi^3 + d\phi^2 + e\phi + f$
 $a = 0$, but for C $a = -1.858543177E-4$

Species	b	c	d	e	f
CH	-2.81288E-5	8.81513E-5	-9.62511E-5	4.54854E-5	-7.99508E-6
CH ₂ (S)	0.0	-2.33645744E-5	6.187499E-5	-4.79645378E-5	1.18586E-5
CH ₂	0.0	-2.5770894E-4	6.5466313E-4	-4.552792E-4	1.016925667E-4
CH ₃ O	2.50804E-5	-8.8027946E-5	1.02314168E-4	-4.2845804E-5	6.1491311E-6
CH ₂ OH	1.798549E-5	-7.6498997E-5	1.0743648E-4	-5.628127E-5	9.9749746E-6
C ₂ H	-1.0853788E-5	4.053624E-5	-5.324172E-5	2.996812E-5	-6.169033E-6
HCCO	-1.616593E-4	5.727109E-4	-7.13716E-4	3.827051E-4	-7.54347E-5
C ₂ H ₂	3.254375E-3	-9.4537368E-3	1.090194E-2	-5.67022822E-3	1.1076645E-3
CH ₂ CO	1.91581E-4	-6.4926484E-4	7.9367146E-4	-3.9599E-4	7.2244946E-5
C ₂ H ₃	-1.77516E-4	5.955E-4	-6.931788E-4	3.529063E-4	-6.7195404E-5
C ₂ H ₄	0.0	0.0	1.16031772E-3	-1.0797324E-3	4.752937E-4
C ₂ H ₅	0.0	-4.80878779E-5	8.8960738E-5	-1.5045318E-5	-2.424657E-6
C ₂ H ₆	0.0	0.0	-2.67984627E-5	1.4164448E-3	-4.1586346E-4
C	8.01937571E-4	-1.3490183E-3	1.1164719145E-3	-4.56431545E-4	7.3856406E-5
C ₂	5.5816157E-9	-1.765406E-8	2.08911E-8	-1.093574E-8	2.13257E-9

The impact of $Y_{j,c}^A(\phi)$ and $Y_{j,c}^B(\phi)$ (Eq. 2.1) is shown in Figure 7; $S_L(\phi)/S_L^o$, the flame speed normalized by its value under the stoichiometric condition, is plotted versus equivalence ratio. Accounting for equivalence ratio variation in $Y_{j,c}^A(\phi)$ and $Y_{j,c}^B(\phi)$ reproduces the detailed chemistry response, but applying the unity equivalence ratio value ($Y_{j,c}^A(\phi = 1)$ and $Y_{j,c}^B(\phi = 1)$) for the full range lacks prediction capabilities, necessitating optimal tables as shown above.

The fresh against burnt gases premixed strained flame is also reproduced by the hybrid method, as illustrated in Figure 8 displaying the response of CO and HO₂ versus physical space for two temperatures of burnt gases, one mimicking heat-loss effects and for a rate of strain $a_T = 50 \text{ s}^{-1}$. Figure 9 shows the response of the oxygen radical to a pulse of strain

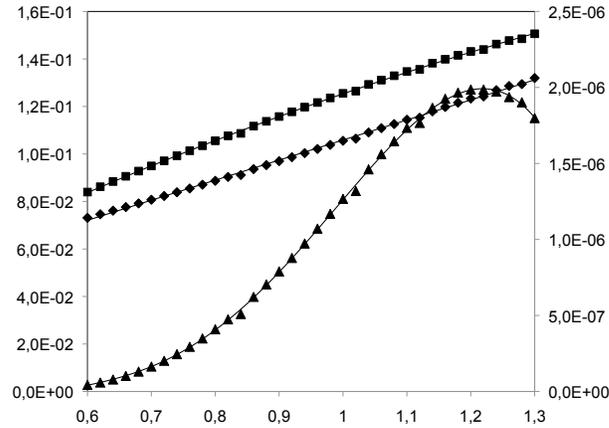


FIGURE 5. CH radical scaling relations versus equivalence ratio. Symbols: from freely premixed flames database. \blacklozenge $Y_{CH,c}^A$, \blacksquare $Y_{CH,c}^B$, \blacktriangle $\max(Y_{CH})$. Line: Approximation from Tables 2-4.

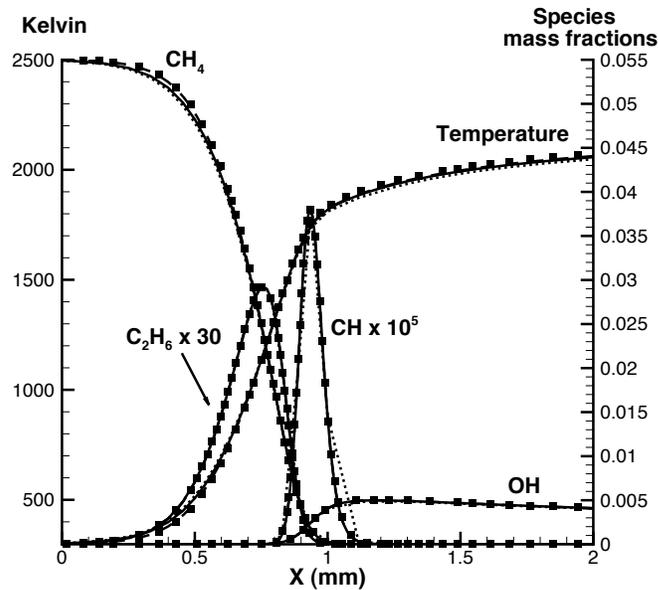


FIGURE 6. Stoichiometric methane/air flame. \blacksquare Detailed chemistry with reference mesh, minimum resolution $h_{min} = 7.28 \mu\text{m}$ (non-uniform mesh), 78 points in the reaction zone. HTTC: - - - reference mesh, — keeping h_{min} , but with 65 points in the reaction zone, \cdots $10h_{min}$ with 30 points.

rate, the species oscillates after the 10% increase of strain and the characteristic time of relaxation along with the intermediate amplitude oscillations are well reproduced; even though the rate of strain does not explicitly enter the definition of $Y_{j,c}^A$, $Y_{j,c}^B$ and Y_j^{\max} . This result suggests that the proposed approach can handle responses of the strongly non-linear chemistry to unsteadiness.

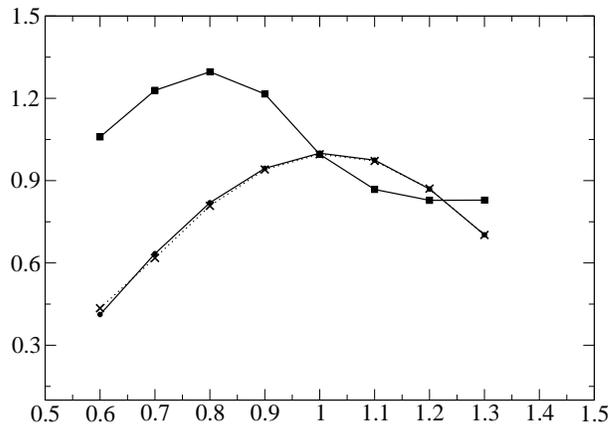


FIGURE 7. Normalized freely propagating flame speed versus equivalence ratio. \times Fully detailed chemistry. \bullet HTTC. \blacksquare $Y_{j,c}^A$ and $Y_{j,c}^B$ fixed at $\phi = 1$.

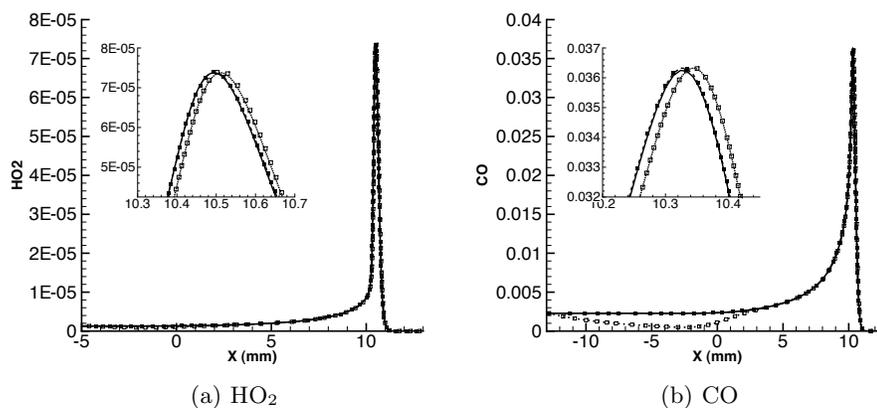


FIGURE 8. Mass fractions versus physical space, lean mixture ($\phi = 0.9$) against burnt gases in premixed counterflowing strained flames, $a_T = 50 \text{ s}^{-1}$. Detailed chemistry: symbols, lines: HTTC. \blacksquare and $—$: $T_o = 300 \text{ K}$ against $T_b = T_{ad}$. \square and \cdots : $T_o = 300 \text{ K}$ against $T_b = 0.8T_{ad}$.

4. Conclusion

A novel approach has been explored to downsize the resolution cost when computing flames with detailed chemistry. The chemical scheme is left unaltered and instead of reducing the number of species and elementary reaction steps, only the intermediate radicals evolving within the thin reaction zones are expressed from global flame parameters (equivalence ratio and progress of reaction). This is done after finding the minor species generic self-similar response in a series of canonical problems: strained and unstrained counterflowing premixed flames with non-adiabatic burnt gases, counterflowing diffusion flames and zero-dimensional self-ignition at various pressures. The resolution needed to

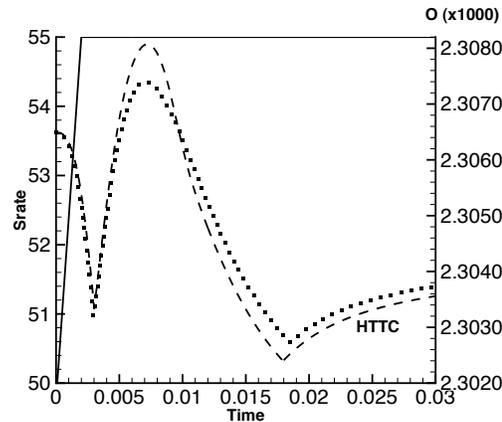


FIGURE 9. Response of the oxygen radical to a 10% pulse of strain rate in a counterflowing lean fresh ($\phi = 0.9$) against stoichiometric burnt gases. - - - HTTC. . . . Detailed chemistry.

properly capture the flame chemical response is then decreased by an order of magnitude in every direction, dramatically decreasing the number of computing cells needed in three-dimensional simulations.

All the species which are non-zero in fresh and burnt gases are transported, allowing combustion systems to be addressed directly with various fuel and oxidizer compositions along with the recirculation of burnt gases in the presence of heat loss. The mesh resolution in Large Eddy Simulation of real burners has recently reached levels (Moureau *et al.* 2011) which are compatible with those needed in this hybrid tabulated transported chemistry strategy (of the order of $70 \mu\text{m}$) hence, opening a new perspective in LES applied to the design and optimization of gaseous burners.

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REFERENCES

- BILGER, R. W., POPE, S. B., BRAY, K. N. C. & DRISCOLL, J. F. 2005 Paradigms in turbulent combustion research. *Proc. Combust. Inst.* **30** (1), 21–42.
- BRADLEY, D., KWA, L. K., LAU, A. K. C., MISSAGHI, M. & CHIN, S. B. 1988 Laminar flamelet modeling of recirculating premixed methane and propane-air combustion. *Combust. Flame* **71** (2), 109–122.
- BRAY, K. N. C. 1996 The challenge of turbulent combustion. *Proc. Combust. Inst.* **26**, 1–26.
- CANDEL, S., SCHMITT, T. & DARABIHA, N. 2011 Progress in transcritical combustion : Experimentation, modeling and simulation. In *23rd ICDERS Conf. (2011)*.
- DOMINGO, P., VERVISCH, L. & VEYNANTE, D. 2008 Large-eddy simulation of a lifted methane-air jet flame in a vitiated coflow. *Combust. Flame* **152** (3), 415–432.
- GICQUEL, O., DARABIHA, N. & THEVENIN, D. 2000 Laminar premixed hydrogen/air

- counterflow flame simulations using flame prolongation of ILDM with differential diffusion. *Proc. Combust. Inst.* **28**, 1901–1908.
- IHME, M., CHA, C. M. & PITSCH, H. 2005 Prediction of local extinction and re-ignition effects in non-premixed turbulent combustion using a flamelet/progress variable approach. *Proc. Combust. Inst.* **30** (1), 793–800.
- JUCHMANN, W., LATZEL, H., SHIN, D., PIETER, G., DREIER, T., VOLPP, H., LINDSTEDT, R. & LEUNG, K. 1998 Absolute radical concentration measurements and modeling of low-pressure CH₄/O₂/NO flames. In *Twenty-seventh Symposium (Int.) on Combustion*, vol. 27, pp. 469–476. The Combustion Institute.
- LINDSTEDT, P. 1998 Modeling of the chemical complexities of flames. In *Twenty-seventh Symposium (Int.) on Combustion*, vol. 27, pp. 269–285. The Combustion Institute.
- LODIER, G., VERVISCH, L., MOUREAU, V. & DOMINGO, P. 2011 Composition-space premixed flamelet solution with differential diffusion for in situ flamelet-generated manifolds. *Combust. Flame* **158** (10), 2009–2016.
- MOUREAU, V., DOMINGO, P. & VERVISCH, L. 2011 From large-eddy simulation to direct numerical simulation of a lean premixed swirl flame: Filtered laminar flamelet modeling. *Combust. Flame* **158** (7), 1340–1357.
- NAUDIN, A., FIORINA, B., PAUBEL, X., VEYNANTE, D. & VERVISCH, L. 2006 *Self-similar behavior of chemistry tabulations in laminar and turbulent multi-fuel injection combustion systems*. Proceedings of the Summer Program - XI, Center for Turbulence Research, Stanford.
- NGUYEN, P., VERVISCH, L., SUBRAMANIAN, V. & DOMINGO, P. 2010 Multidimensional flamelet-generated manifolds for partially premixed combustion. *Combust. Flame* **157** (1), 43–61.
- VAN OIJEN, J. A., LAMMERS, F. A. & DE GOEY, L. P. H. 2001 Modelling of complex premixed burner systems by using flamelet-generated manifolds. *Combust. Flame* **127** (3), 2124–2134.
- 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.
- RIBERT, G., GICQUEL, O., DARABIHA, N. & VEYNANTE, D. 2006 Tabulation of complex chemistry based on self-similar behaviour of laminar premixed flames. *Combust. Flame* **146** (4), 649–664.
- RIBERT, G., WANG, K. & VERVISCH, L. 2012 A multi-zone self-similar chemistry tabulation with application to auto-ignition including cool-flames effects fuel. *Fuel* **91** (1), 87–92.
- SUBRAMANIAN, V., DOMINGO, P. & VERVISCH, L. 2010 Large-eddy simulation of forced ignition of an annular bluff-body burner. *Combust. Flame* **157** (3), 579–601.
- VEYNANTE, D., FIORINA, B., DOMINGO, P. & VERVISCH, L. 2008 Using self-similar properties of turbulent premixed flames to downsize chemical tables in high-performance numerical simulations. *Combust. Theory and Modelling* **12** (6), 1055–1088.
- WANG, K., RIBERT, G., DOMINGO, P. & VERVISCH, L. 2010 Self-similar behavior and chemistry tabulation of burnt-gases diluted premixed flamelets including heat-loss. *Combust. Theory and Modelling* **4** (14), 541–570.
- WILLIAMS, F. A. 1985 *Combustion theory*. The Benjamin/Cummings Publishing Company, Inc.