Modeling Soot Formation in LES of Turbulent Flames Using Virtual Chemistry

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1 Introduction

To reduce pollutant emission in practical combustion applications, one needs the development of more efficient combustion devices. Soot formation is one of the most challenging issues in this topic, characterized by the complexity of the phenomena occurring throughout the combustion processes. In fact, soot production is highly coupled with turbulent flow motion as well as chemical reactions. Numerical simulations of turbulent combustion are particularly high CPU demanding in sooting flames.

A typical solution to decrease simulation time is to use reduced gas kinetics and simplified models for the solid particles. A virtual chemistry model developed by [1] combines these features. The virtual scheme bypasses the gas-solid complex formalism without explicit mathematical distinction between both phases. To account for radiative heat losses in sooting flames, the optimization of the virtual mechanism includes a new virtual parameter describing the Planck absorption coefficient [2]. This model has been validated in several 1-D and 2-D laminar ethylene-air premixed and non-premixed flame configurations [2] such as counterflow flames, burner-stabilized burner, premixed slot burner and Santoro's coflow burner [3]. The objective is to allow the final user to perform Large Eddy Simulations (LES) of turbulent sooting flames using virtual chemistry at a low CPU cost and accurately predict the flame structure and soot formation, at any regime, and in a wide range of operating conditions.

The virtual chemistry model describing the heat release and flame temperature including radiative heat transfer is briefly presented Section 2. The virtual chemistry model for soot formation prediction is also presented. Section 3 presents the application of the virtual model to a 3-D turbulent sooting jet flame. A brief description of the experimental and numerical setups is given, followed by the results. Then, the conclusion and final discussion is presented.

2 Virtual Chemistry for soot formation

Virtual chemistry approach is based on virtual species and reactions whose thermodynamic and chemical properties are optimized using machine learning algorithms in order to retrieve properties of reference flames gathered in a learning database [4]. The virtual reactions between the virtual species do not represent real chemical processes but form a mathematical architecture designed to reproduce user-specified targets such as temperature and mass fractions of combustion products and pollutant species.

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A virtual scheme consists of a main block that models the heat release from the flame and one or more sub-mechanisms each one dedicated to a particular pollutant.

2.1 Main virtual mechanism including radiation

The main virtual mechanism structure presented in Ref. [4] has shown that a reaction scheme composed of 8 species and 2 reactions accurately captures the heat release and the flame temperature for a wide range of fuel compositions. Thermodynamic and transport properties of the virtual species are calibrated to recover averaged properties of the multi-component real mixture [4].

Radiative heat transfer in the gas phase may significantly impact the heat release and the flame temperature. To account for this phenomenon, radiative properties of the reactive flow are also included in the optimization process together with the main virtual mechanism. These properties depend on gas temperature and mixture composition and are captured through the gaseous Planck mean absorption coefficient modeled by a set of virtual parameters [2].

2.2 Soot virtual sub-mechanism

A soot model requires first a fine description of the detailed chemistry leading to the soot precursor formation (PAHs) [5]. Then the formation of the particles is described though a nucleation model [6]. Finally, the solid phase is modeled employing dedicated particles interaction and distribution formalisms such as for instance sectional [7] or MOM [8] approaches. These successive modeling stages lead to the prediction of the soot characteristics such as volume fraction, particle number density, etc.

Most of chemical reduction methods, such as tabulated [9] or analytically-reduced [10] approaches, are limited to the gas-phase kinetics, the description the solid phase remaining very time consuming. The soot virtual sub-mechanism proposed in the present work has the advantage of reducing the overall process by targeting only the final quantities of interest. Here we focus on the soot volume fraction f_v^s as a target.

Soot particles are represented by a virtual species $\mathcal{S}$. The soot volume fraction f_v^s is calculated from the soot species mass fraction $Y_\mathcal{S}$: $f_v^s = (\rho/\rho_s)Y_\mathcal{S}$; where ρ is the gaseous mixture density, and ρ_s is the soot density considered constant, ρ_s =1800 kg m^{-3}.

To predict the formation of the soot virtual species $\mathcal{S}$, the following chemical structure is proposed:

$$\alpha^s_{F,1}F + \alpha^s_{Ox,1}Ox \rightarrow \alpha^s_{V_1,1}V_1 + \alpha^s_{V_3,1}V_3 \qquad (R^s_1)$$

$$V_1 \rightarrow \alpha^s_{s,2}\mathcal{S} + \alpha^s_{V_2,2}V_2 \qquad (R^s_2)$$

$$\mathcal{S} + V_1 + V_2 \rightarrow \alpha^s_{\mathcal{S},3}\mathcal{S} + \alpha^s_{V_3,3}V_3 \qquad (R^s_3)$$

$$\mathcal{S} + Ox + V_2 \rightarrow Ox + 2V_3 \qquad (R^s_4)$$

where F represents the fuel, Ox the oxidizer, and V_1, V_2 and V_3 are three virtual species. $\alpha^s_{k,r}$ is the stoichiometric coefficient per unit mass of the k^{th} virtual species in the r^{th} virtual reaction. The superscript s stands for soot virtual sub-mechanism.

The soot production is initiated by reaction R^s_1. The rate of progress of this reaction must be the as in the main virtual mechanism in order to be consistent with the fuel oxidation. The three virtual species V_1, V_2 and V_3 together with virtual reactions R^s_2, R^s_3 and R^s_4 represent the nucleation, surface growth and coagulation, and oxidation of soot particles.

A cost function is defined to calibrate the set of reaction rate parameters of the virtual sub-mechanism. As in Ref. [1], a reference flame database composed of 1-D laminar premixed and non-premixed ethylene-air flames [2] is generated using the detailed soot kinetics from [5]. Radiative heat losses are calculated using the optically-thin narrow-band radiation model based on Ref. [11], included in REGATH package [12].

3 Application to a 3-D turbulent sooting flame

The virtual chemistry and radiative models for sooting flames are used to perform LES of the 3-D turbulent ethylene non-premixed jet flame studied by [13], included the International Sooting Flame (ISF) workshop [14].

3.1 Sandia burner experimental setup

The experimental configuration comprises a piloted jet burner surrounded by a coflow of air. The piloted jet burner consists of a central jet tube and a pilot tube.The central tube is fed with pure ethylene with a bulk velocity of $u_{fuel}^{\text{bulk}} = 54.7\ \text{cm}\,\text{s}^{-1}$ and exit Reynolds number of $Re = 20000$. An ethylene-air mixture of $\phi = 0.9$ and mass flow rate of $1.772 \times 10^{-4}\ \text{kg}\,\text{s}^{-1}$ is injected through the pilot tube, corresponding to 2% of the heat release rate of the central jet. The air coflow mean velocity is $0.6\ \text{m}\,\text{s}^{-1}$. All gases are injected at atmospheric pressure and $T = 294$ K. Please refer to Ref. [13] for further details on this configuration.

3.2 Numerical setup

Downstream the burner and coflow exits, the computational domain has a cylindrical shape with a radius of $94D$ and a length of $500D$, where $D = 3.2$ mm is the diameter of the central jet tube. An unstructured grid consisting of 43.4 millions of tetrahedral cells was built.

Boundary conditions in the Sandia burner include: the injector and coflow inlets, injector and lateral no slip walls, and the outlet. For the fuel stream, fully-developed velocities profiles fitted in a pipe flow simulation [7] are imposed at the inlet. The pilot inlet boundary condition is modeled as a uniform annular gas-mixture flow at the given mass flow rate, with composition and temperature corresponding to the burnt gases at equilibrium in adiabatic condition, for initial composition $\phi = 0.9$ and inlet temperature 294 K. A temperature distribution of 1000 K is imposed at the pilot walls. Lateral walls are adiabatic.

LES of the Sandia burner are carried out with the YALES2 low-Mach number, unstructured finite-volume code [15], employing a fourth order spatial and temporal schemes, and sigma model for turbulence [16]. The main virtual mechanism and the soot virtual sub-mechanism combustion models are coupled to the LES solver.

The turbulence-chemistry interactions are solved using the Thickened Flame model for LES (TFLES) [17], with sub-grid scale flame wrinkling from [18]. The turbulent Schmidt number of $\mathcal{S}$ and virtual species are dynamically evaluated [7].

3.3 Results

A qualitative analysis of the LES results is first introduced. Figure 1 shows instantaneous fields of temperature and soot volume fraction using the virtual main and soot sub-mechanism models. The

length of the flame is achieved as observed in experimental results of about 830 mm [13]. A maximum value of about 1.6 ppm for the soot volume fraction is obtained, which is of the same order of magnitude as in experimental measurements. It is also noticeable that soot convects towards the flame front and is exclusively formed in the rich zone of the flame.

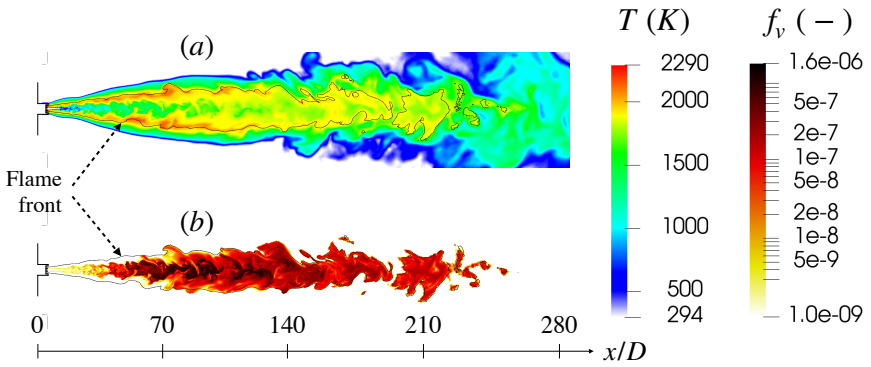

Figure 1: Sandia burner: Instantaneous fields of (a) temperature and (b) soot volume fraction.

A quantitative analysis will now be carried out by extracting characteristic profiles out of the simulation results and comparing them against experimental data (black dots). Also, in order to validate the virtual chemistry and radiative models in the turbulent regime, numerical results using the adiabatic model (blue lines) and the radiative model including gas and soot contributions in the heat loss term (red lines) will be presented. Radial profile measurements of temperature from [19] are now taken into account for further validation of the flame structure. Figure 2 shows radial profiles of the mean (left) and RMS (right) temperature profiles at x/D =175. As expected, mean temperature radial profiles show better agreement when radiation is considered. Despite having a small overestimation, results are satisfactory and are comparable to detailed numerical results of other works in the literature, as the one from Ref. [7]. Concerning RMS, virtual non-adiabatic results show a very good agreement with experimental measurements.

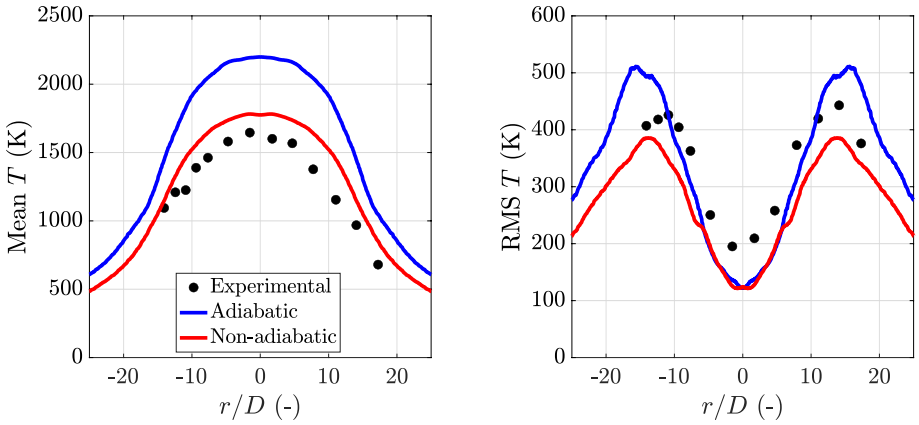

Figure 2: Sandia burner: Mean and RMS temperature radial profiles at $x/D = 175$.

Figure 3 (left) presents mean soot volume fraction profiles along the jet centerline for both adiabatic and non-adiabatic virtual models. Results are compared to experimental measurements from Ref. [13]. The peak of soot volume fraction is properly obtained by the non-adiabatic virtual model, while it is underestimated by the adiabatic model. Figure 3 (right) illustrates the mean soot volume fraction radial

profiles at $x/D = 101.6$, where the non-adiabatic virtual scheme reproduces correctly the soot volume fraction maximum value compared to the experiment but does not capture the double-peaked shape.

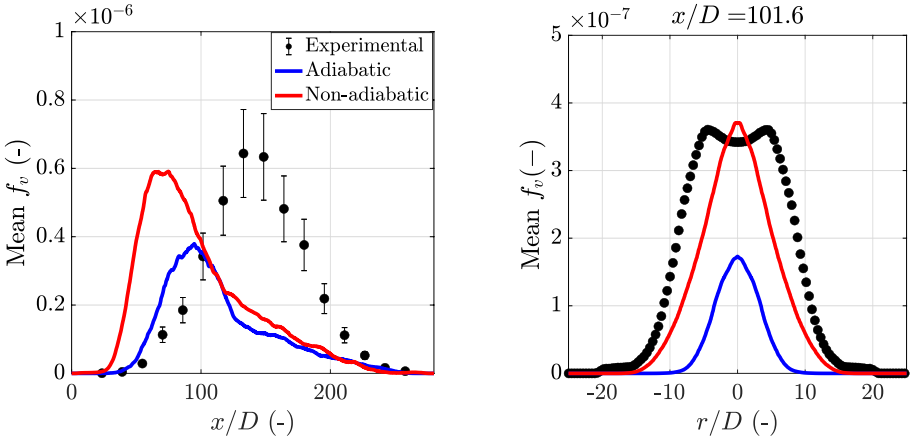

Figure 3: Sandia burner: Mean soot volume fraction profiles: (left) along the jet centerline, and (right) radial at $x/D = 101.6$.

A simulation of 250 ms physical time needed about 2.2×10^5 CPU hours. This CPU time is three times smaller than the total CPU time made by Ref. [7] for the same physical time using a sectional method.

4 Conclusions and discussion

A new reduced chemistry model based on virtual chemistry approach was developed to predict the temperature and heat release rate together with soot volume fraction in a 3-D turbulent ethylene-air flame. Also, a virtual approach was proposed in order to include gas phase radiative heat transfer in the simulations. The models were applied to turbulent flames using LES in the laboratory-scale configuration studied by [13]. The proposed models allow to retrieve flame characteristics, such as temperature, heat release, radiative effects and soot volume fraction, at very low CPU costs.

LES of Sandia flame was performed using the virtual chemistry and virtual radiative models giving good agreement with experimental measurements. The new virtual soot sub-mechanism captures soot physical phenomena with good level of accuracy compared to other numerical works in the literature. Also, the new virtual radiative model well provides the heat losses. In overall, the different turbulence-chemistry-soot-radiative interactions in the Sandia flame are well retrieved by this virtual approach.

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References

[1] H. M. Colmán, A. Cuoci, N. Darabiha, and B. Fiorina. (2019). A reduced virtual chemistry model for soot prediction in hydrocarbon-air flames. 27th ICDERS: 102.

[2] H. Maldonado Colmán, A. Cuoci, N. Darabiha, and B. Fiorina. (2022). A virtual chemistry model for soot prediction in flames including radiative heat transfer. Combust. Flame. 238: 111879.

[3] R. J. Santoro, H. G. Semerjian, and R. A. Dobbins. (1983). Soot particle measurements in diffusion flames. Combust. Flame. 51: 203.

[4] M. Cailler, N. Darabiha, and B. Fiorina. (2020). Development of a virtual optimized chemistry method. Application to hydrocarbon/air combustion. Combust. Flame. 211: 281.

[5] C. Saggese, S. Ferrario, J. Camacho, A. Cuoci, A. Frassoldati, E. Ranzi, H. Wang, and T. Faravelli. (2015). Kinetic modeling of particle size distribution of soot in a premixed burner-stabilized stagnation ethylene flame. Combust. Flame. 162: 3356.

[6] C. A. Schuetz and M. Frenklach. (2002). Nucleation of soot: Molecular dynamics simulations of pyrene dimerization. Proc. Combust. Inst. 29: 2307.

[7] P. Rodrigues, O. Gicquel, B. Franzelli, N. Darabiha, and R. Vicquelin. (2019). Analysis of radiative transfer in a turbulent sooting jet flame using a monte carlo method coupled to large eddy simulation. J. Quant. Spectrosc. Radiat. Transfer. 235: 187.

[8] M. E. Mueller, G. Blanquart, and H. Pitsch. (2009). A joint volume-surface model of soot aggregation with the method of moments. Proc. Combust. Inst. 32: 785.

[9] B. Fiorina, O. Gicquel, L. Vervisch, S. Carpentier, and N. Darabiha. (2005). Premixed turbulent combustion modeling using tabulated detailed chemistry and pdf. Proc. Combust. Inst. 30: 867.

[10] P. Pepiot-Desjardins and H. Pitsch. (2008). An efficient error-propagation-based reduction method for large chemical kinetic mechanisms. Combust. Flame. 154: 67.

[11] A. Soufiani and J. Taine. (1987). Application of statistical narrow-band model to coupled radiation and convection at high temperature. Int. J. Heat Mass Transf. 30: 437.

[12] N. Darabiha and S. Candel. (1992). The influence of the temperature on extinction and ignition limits of strained hydrogen-air diffusion flames. Combust. Sci. Technol. 86: 67.

[13] J. Zhang, C. R. Shaddix, and R. W. Schefer. (2011). Design of "model-friendly" turbulent non-premixed jet burners for C2+ hydrocarbon fuels. Rev Sci Instrum 82: 074101.

[14] ISF5, International sooting flame, http://www.adelaide.edu.au/cet/isfworkshop/data-sets/.

[15] V. Moureau, P. Domingo, and L. Vervisch. (2011). Design of a massively parallel cfd code for complex geometries. Comptes Rendus Mécanique. 339: 141.

[16] F. Nicoud, H. B. Toda, O. Cabrit, S. Bose, and J. Lee. (2011). Using singular values to build a subgrid-scale model for large eddy simulations. Physics of fluids. 23: 085106.

[17] O. Colin, F. Ducros, D. Veynante, and T. Poinsot. (2000). A thickened flame model for large eddy simulations of turbulent premixed combustion. Physics of fluids. 12: 1843.

[18] F. Charlette, C. Meneveau, and D. Veynante. (2002). A power-law flame wrinkling model for LES of premixed turbulent combustion part ii: dynamic formulation. Combust. Flame. 131: 181.

[19] S. P. Kearney, D. R. Guildenbecher, C. Winters, T. W. Grasser, P. A. Farias, and J. C. Hewson. (2015). Temperature, oxygen, and soot-volume-fraction measurements in a turbulent c2h4-fueled jet flame. Sandia Report 7968.