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Large Eddy Simulation of Supercritical $\mathrm{CH_4/CO_2/O_2}$ Non-Premixed Turbulent Oxy-Combustion

Eugenio Giacomazzi, Donato Cecere
Franca Rita Picchia, Nunzio Arcidiacono, Giuseppe Messina
Process and Energy Systems Engineering Laboratory, ENEA
Casaccia Research Center, Rome - ITALY

ABSTRACT

The aim of this work is to investigate turbulent non-premixed methane oxy-combustion in an atmosphere rich in $\mathrm{CO_2}$ at supercritical conditions by means of Large Eddy Simulation. A simple shear-layer configuration typical of slot burners is considered: the fuel is $\mathrm{CH_4}$ and flows through the central slot, the oxidant $\mathrm{O_2/CO_2}$ (90, 30, 10% $\mathrm{O_2}$ by mass) mixture flows on both sides of the slot, and a pilot flame is imposed at the inlet between the two streams of reactants to force ignition.

Fully compressible Navier-Stokes equations coupled to the Peng-Robinson cubic equation of state for real gases in its improved translated volume formulation are solved adopting the Large Eddy Simulation approach. The dynamic Smagorinsky and the LTSM subgrid scale models are used for turbulence and combustion closures. Among the diffusive mechanisms only the Dufour effect is neglected; transport properties are accurately calculated. The M_1 radiant transfer of energy model is adopted without considering turbulence-radiation interactions.

Numerical results are examined to highlight different physical aspects of the flow: the effect of oxygen stream dilution on the flame anchoring and topology; the effect of the radiant transfer of energy on the flame structure; diffusive mechanisms, to identify the fastest processes through the comparison of characteristic times.

INTRODUCTION

ENEA's interest on supercritical $\mathrm{CO_2}$ (s-$\mathrm{CO_2}$) gas turbine cycles comes from the current development of the European electric system and its needs. In electric systems with a large share of variable renewable energy sources (vRES) such as solar and wind, thermal power generation is transforming from an electrical energy supplier to a flexibility supplier. The electrical system flexibility is a key pillar to make effective the contribution of the current share of vRES and, hopefully, to strongly increase it. In fact, by increasing the share of vRES also the electrical system power fluctuations increase due to the inherent uncertainty of sun and wind as primary energy sources; consequently, the need of both dispatchable and flexible back-up power generation increases. Hydroelectric power generation, jointly with gas turbine (GT) based power generation, are currently the best options to provide both highly flexible and low emission back-up power.

Operational flexibility relates at a "family of performance" defining how quickly a power plant can respond to load changes, referring to the whole operational envelope, including load gradients, minimum load, part-load efficiency and both start-up and shut-down operation times. Apart from hydroelectric power generation, whose deployment is strongly constrained by the land orography,

gas turbines both in open cycle configuration (OCGT) and in combined cycle (CCGT) are often the best solution to provide the required flexibility. Specifically OCGTs are very suitable for flexible generation and as peak-load suppliers, but they exhibit low efficiency and consequently, both high emissions and high costs. CCGTs are currently the best in class in terms of efficiency at nominal power, but both frequent start-ups and thermal cycling due to flexible operations negatively impact on the power plant's economics, increasing the maintenance costs and reducing its technical life. Besides these issues, current Carbon Capture and Storage (CCS) technologies are not suited for gas turbine based power plants running as flexible back-up generation. This is more true in the near future scenario with an increased share of vRES. Furthermore, CCS' costs are high.

In the recent years it is growing the interest on s-$\mathrm{CO_2}$ power cycles equipped with oxy-combustion as a new technology able to solve simultaneously all the above issues, integrating the OCGTs typical operational flexibility with an efficiency target closer to CCGTs and with both flexible and efficient carbon capture capabilities. Oxy-combustion s-$\mathrm{CO_2}$ (OXYSCO2) power plants are internal combustion engines using s-$\mathrm{CO2}$ as working fluid, conceptually similar to conventional gas turbines. As an emerging technology, the OXYSCO2 engine capabilities are not yet demonstrated, although there are on-track two important research project in US. Concerning their operational flexibility, it is likely that the heat exchangers will be the most important barrier at plant component level and combustion dynamics at process level. Combustion dynamics knowledge related to conventional gas turbines can lead to a basic understanding of the relationships between the engine operational flexibility and combustion dynamics. However, the strong differences in thermo-physical properties between the perfect gas model and the supercritical fluid model require a complete re-shaping of the combustion modelling strategies as well as new customized numerical tools.

High-pressure combustion of reactants exhibiting real gas behaviour is becoming an important research topic not only for the growing interest in supercritical $\mathrm{CO_2}$ gas turbine cycles, but also in other industrial applications, such as organic Rankine gas turbine cycles, diesel engines with higher and higher pressures, liquid oxygen rocket engines, cooling systems. Injection pressures are usually above the critical values of common propellants, like oxygen, hydrogen and hydrocarbons; for example, oxy-combustors of s-$\mathrm{CO_2}$ gas turbines are being designed to operate at $300\,\mathrm{bar}$. Furthermore, in some applications, e.g., rocket engines, $\mathrm{O_2}$ may be injected at cryogenic temperatures: these conditions are far away from ideal thermodynamics, and the different fluid behaviour has to be accounted for by means of real gas equations of state and specific models for molecular transport properties.

Experimental work at such high-pressure conditions may be prohibitive: the use of advanced laser diagnostics is not an easy task to achieve, and very few examples exist in literature; besides, facilities themselves may be very expensive. Hence, most of research and design in this area is based on numerical simulation. However, also the numerical approach is not an easy task, suffering problems of both modelling accuracy (e.g., the ability to capture huge variations of fluid properties when crossing the pseudo-boiling line) and computational efficiency (too complex and accurate equations of state cannot be used in time-consuming simulations, like LES and DNS). Furthermore, numerical schemes are stressed hardly: the high-density gradients typical of some applications (as those involving liquid injection) and the multi-species transport enhance wiggle formation in fully compressible solvers.

This work is devoted to numerical simulation of methane oxy-combustion in supercritical carbon dioxide at $300\,\mathrm{bar}$. Although adopting a simple chemical scheme, simplifying assumptions on the radiative transfer of energy and a two-dimensional computational domain, some interesting observations are highlighted, and needs for future work identified.

THE RADIATIVE TRANSFER OF ENERGY

The Radiative Trasfer of Energy (RTE) is a very important mechanism in several applications. At industrial furnace and combustion chamber temperatures the gaseous species that absorb and emit significantly are $\mathrm{CO_2}$, $\mathrm{H_2O}$, CO, $\mathrm{SO_2}$, NO, and $\mathrm{CH_4}$. Other gases, such as $\mathrm{N_2}$, $\mathrm{O_2}$ and $\mathrm{H_2}$, are transparent to infrared radiation and do not emit significantly; however, they become important absorbing/emitting contributors at very high temperatures. A non-negligible contribution to radiation is also providied by hot carbon (soot) particles within the flame and from suspended particulate material (as in pulverized-coal combustion).

In gases, the absorption coefficient κ_λ often varies strongly with wavelength, temperature and pressure: $\kappa_\lambda = \kappa_\lambda(T, p)$. Emission and absorption spectra can exhibit lines and continuous regions. Differently from solid surfaces that can be considered opaque, gas properties exhibit very irregular wavelength dependencies: absorption or emission by gases is significant only in certain wavelength regions, especially when the gas temperature is below a few thousand Kelvin.

Emission and absorption coefficients of gases increase proportionally to the concentration of the participating species, and hence to the mixture pressure for a given species mass or molar fraction. Besides, the spectral coefficients vary with temperature but also with pressure. Increasing pressure results in spectral line broadening, mainly due to molecular collisions (since the gas density increases) [1], up to wider and more overlapping lines than at lower pressures: the result is that the gas becomes "grayer" (opaque). The spectral absorption coefficient amplitude may increase by more than 1000 times moving from 1 to $200\,\mathrm{bar}$, as shown in [2, p. 138] or [3]. The absorption coefficient of cold lines decreases when the temperature increases, but towards high wavelength, hot lines appear thus resulting in a slight increase of the absorption coefficient.

When the total emission from a volume element of absorbing medium is to be calculated, the appropriate mean absorption coefficient is the Planck mean absorption coefficient, $\kappa_P(T, p)$. The Planck mean is convenient since it depends only the local properties and it can be tabulated. Values for κ_P have been estimated in the past [4, p. 253-324], but these are today known to be seriously in error, especially at higher temperatures and pressures. Today, the Planck mean absorption coefficient for single species can be calculated [5] directly from high-resolution spectroscopic databases such as HITRAN [6] and HITEMP [7]: the new databases, with the inclusion of more lines from higher vibrational energy levels provide more accurate absorption data, thus resulting in κ_P at high temperature generally larger than those obtained through older databases.

Looking at available literature, some interesting results are here reported. Numerical simulations of the heat transfer from the hot wall of a pipe containing a flow of $\mathrm{CO_2}$ [3] showed that the influence of radiation on the flow temperature and velocity distribution decreases with an increase of the operating pressure and becomes negligible above 200 bar. This means that the radiant transfer of energy becomes weak at very high pressure since the optical thickness increases and radiation is absorbed in a very thin layer; for the problem investigated in [3], this layer is near the hot inner surface of the pipe. However, the present authors note that such conclusions should be confirmed by analysing the competition between thermal radiation and conductivity to identify the most effective heat transfer mechanism in the thin layer close to the wall, e.g., by looking at the associated budgets in the energy transport equation as done in this work.

Concerning flames, it is well known that neglecting radiation at atmospheric pressure conditions may lead to overprediction of temperature of up to $200\,\mathrm{K}$. However, it has to be noted that numerical predictions strongly depend on the RTE model adopted: the usually-employed optically-thin or gray radiation models lead to underprediction of temperature of up to $100\,\mathrm{K}$ and more [8, 9]. Furthermore, radiation is enhanced by turbulence through strong nonlinear interactions between

temperature and radiative property fluctuations (TRI): such interactions increase the heat loss from a flame leading to a reduction in the local gas temperature of $200\,\mathrm{K}$ and more. The TRI cooling effect is even enhanced in high-pressure combustors that typically exhibit larger optical thicknesses [10, 11]. For systems where RTE is important, radiation was shown to be equivalent to a large nonlinear diffusion term: in fact, the radiant transfer of heat can work as a preheating mechanism as conduction of heat, thus resulting in higher flame speeds [12].

Focusing on peak flame temperature, radiation typically reduces it in low pressure flames. Although radiation effects become more pronounced as the pressure is increased (due to increased emission/absorption), the peak flame temperature is less affected by radiation because of the faster chemical reactions, as also observed in [2, p. 139]. In fact, according to the Arrhenius law, the chemical reaction rate is proportional to reactant molar concentrations raised to a power that is equal to the number of moles that have to collide for the reaction to occur. Hence, the reaction rate is proportional to p for first-order reactions and to p^2 for second-order reactions; three-body reactions, are even more enhanced [13].

PHYSICAL AND NUMERICAL MODELS ADOPTED

In this article the compressible Navier-Stokes equations are solved for a reacting real gas flow at supercritical conditions for which the Peng-Robinson cubic equation of state in its improved volume translated formulation is assumed. The mathematical models adopted are derived for a fluid of N_s chemical species. The constitutive laws assumed to describe the behaviour of the fluid are here reported. They simply model the microscopic molecular diffusion of momentum, energy and mass, i.e., they model the momentum flux $\boldsymbol{S}$, the heat flux $\boldsymbol{Q}$ and the species mass flux $\boldsymbol{J_i}$.

A Newtonian fluid is considered and the Stokes' assumption is made: it is characterized by the following constitutive relation between the stress, $\boldsymbol{S}$, and the strain rate, $\boldsymbol{E}$,

$$\boldsymbol{S} = -\left(p + 2/3\,\mu\nabla\cdot\boldsymbol{u}\right)\boldsymbol{I} + 2\mu\boldsymbol{E} = -p\boldsymbol{I} + \boldsymbol{\mathcal{T}}\,, \tag{1}$$

μ being the viscosity; $\boldsymbol{\mathcal{T}}$ is the viscous part of the stress tensor.

The mass diffusion flux has three contributions [14]. The first one is due to concentration gradients (here modelled through the Hirschfelder and Curtiss' law for multi-component mixtures) [15], the second due to pressure gradients (the baro-diffusion mechanism) [16], and the third one due to temperature gradients (the thermo-diffusion or Soret effect) [17]:

$$\boldsymbol{J}_i = \rho Y_i \boldsymbol{V}_i = \boldsymbol{J}_i^{HC} + \boldsymbol{J}_i^{BD} + \boldsymbol{J}_i^{S} = -\rho Y_i D_i \left[\frac{\nabla X_i}{X_i} + \frac{X_i - Y_i}{X_i}\frac{\nabla p}{p}\right] - \mathcal{D}_i^T \frac{\nabla T}{T}. \tag{2}$$

The diffusion coefficient D_i is an *effective* diffusion coefficient of the i-th species into the mixture.

Keeping apart the radiative heat transfer of energy, the heat flux has three contributions too. The first due to temperature gradients (the Fourier diffusion), the second due to mass diffusion fluxes, and the third one is the Dufour effect (reciprocal of the Soret effect):

$$\boldsymbol{Q} = \boldsymbol{q}_F + \boldsymbol{q}_{V_i} + \boldsymbol{q}_D = -K\nabla T + \rho\sum_{i=1}^{N_s} h_{s_i} Y_i \boldsymbol{V_i} + \boldsymbol{q}_D. \tag{3}$$

Note that this is the heat flux expression entering into the energy transport equation where formation energies are isolated in a source term, i.e., not included in the energy definition. Usually the Dufour effect (the third term) is negligible even when thermo-diffusion is not [17, p. 768] and hence it is neglected in the present work.

Molecular transport properties for individual species are accurately modelled through NIST models [18] for viscosity and thermal conductivity. The diffusion coefficient D_i of the i-th species into the rest of mixture is modelled according to the Hirschfelder and Curtiss expression [15], where the required binary diffusion coefficient is calculated by means of kinetic theory. The thermo-diffusion coefficient $\mathcal{D}_i^T$ is estimated by means of the EGLIB routines [19].

A simplified chemical mechanism consisting of 4 reactions and 6 species developed for oxy-combustion is adopted [20, Table 2]. Since main radical species are not included in the mechanism, temperature is overestimated by more than 300 K [21]. This is a first step before facing a more complex kinetics.

The transport equations are solved in the framework of Large Eddy Simulation. Unclosed turbulent combustion subgrid terms of the filtered compressible Navier-Stokes equations are modelled through the dynamic Smagorinsky model and the authors' LTSM (Localised Turbulent Scale Model) [22] turbulent combustion model.

The radiant transfer of energy is also taken into account by means of the M_1 diffusive model. For the time being, turbulence-radiation interaction is neglected, although it is expected to play an important role in flame cooling at the high-pressure conditions of the present simulations. Another simplification is the adoption of the individual species' Planck mean absorption coefficients typically used at atmospheric pressure. Since they are expected to increase by increasing pressure, the effect of multiplying them by 1000 is investigated. In the future they will be accurately calculated by using the high-resolution spectroscopic databases HITRAN [6] and HITEMP [7].

In this work, a macroscopic radiation model, the M_1 model, is adopted. It is based on field equations for the radiative energy and the radiative heat flux vector. The limit is that it is valid for non scattering media. The main advantage is that it is independent of the opacity of the media, i.e., it adapts itself and works from thin to thick optical thickness. An averaged form of the M_1 model for turbulent flows exists [23, 24] and a simplified formulation of this model was finally developed [25]: this is better suited for combustion problems and thus applied in present simulations. In particular, since the original unsteady equations in [23] are very stiff and difficult to solve, a more friendly form is obtained assuming that radiative energy rapidly reaches its equilibrium state, i.e., neglecting the time derivatives. In this way two diffusion equations are obtained for the radiant energy density, E_r $\left[\mathrm{J\,m^{-3}}\right]$, and for its flux vector, $\boldsymbol{F}_r$ $\left[\mathrm{J\,m^{-2}\,s^{-1}}\right]$,

$$\nabla \cdot \left[\frac{1}{\sigma_F}\nabla \cdot (\boldsymbol{D}_r E_r)\right] = -\left(\sigma_P a T^4 - \sigma_E E_r\right) \tag{4}$$

$$-\nabla \cdot \left[\frac{\boldsymbol{D}_r}{\sigma_E}\nabla \cdot \boldsymbol{F}_r\right] + \sigma_F \boldsymbol{F}_r = -c\nabla \cdot \left[\frac{\sigma_P}{\sigma_E}\boldsymbol{D}_r a T^4\right] \ , \tag{5}$$

where T is the temperature, $c = 299792458\,\mathrm{ms^{-1}}$ is the speed of light, $a = 4\,\sigma_{SB}/c = 7.565767 \cdot 10^{-16}\,\mathrm{J\,m^{-3}\,K^{-4}}$, σ_{SB} being the Stefan-Boltzmann constant, σ_P is the Planck mean absorption coefficient of the mixture, σ_E and σ_F its two effective absorption coefficients, $\boldsymbol{D}_r$ the nondimensional Eddington tensor that takes into account the local opacity of the flame.

It is observed that in the present numerical experiment, since it deals with oxy-combustion in a supercritical $\mathrm{CO_2}$ atmosphere and since the $\mathrm{CO_2}$ becomes optically thick by increasing pressure, the radiation transport can be correctly modelled by a diffusion process.

The quasi-steady radiative transfer equations are solved (by using a SOR technique coupled to a median spatial filter) periodically at some time steps to update the radiative sink/source term, $\dot{S}_{rad} = -\nabla \cdot \boldsymbol{F}_r$ $\left[W/m^3\right]$, in the transported energy equation.

NUMERICAL SCHEMES AND BOUNDARY CONDITIONS

The numerical simulations are performed by means of the in-house parallel code HeaRT and ENEA's supercomputing facility CRESCO [26]. The HeaRT code solves the compressible Navier-Stokes equations discretised through staggered finite-difference schemes. A second-order accurate centered scheme is adopted for diffusive fluxes; convective terms are modelled through the $AUSM^{+}$-up method [27] coupled with a third/fifth-order accurate $WENO$ interpolation to reduce spurious oscillations (strongly experienced using centered schemes in high-pressure tests); such numerical spatial scheme was extensively tested by the present authors proving its robustness and accuracy [28]. The low-storage third-order accurate Runge-Kutta method of Shu-Osher is used for time integration. The total energy is defined as sum of internal (thermal) and kinetic energy only. The authors found this choice mandatory [29, 30] to avoid, or at least reduce, unphysical energy and temperature oscillations, mainly driving to the divergence of calculation. No spurious waves were experienced in previous simulations of premixed flames, when the total energy was defined including the chemical formation contribution.

Non-reflecting boundary conditions [31, 32, 33] are implemented at open boundaries in their extended form to take into account the effect of variable transport properties [34], local heat release [35] and real gas effects [36]. It is observed that in previous and present real gas simulations the authors had to impose a higher value of the relaxation constant in the partially non-reflecting treatment of the outlet with respect to the ideal gas theoretical value (1.5 against 0.27) [31, 37] to avoid unphysical pressure drift in the whole computational domain. A synthetic turbulence generator is adopted at flow inlets [38].

THE NUMERICAL EXPERIMENT AND ITS SET-UP

The numerical experiment here simulated has a nominal pressure around $300\,\mathrm{bar}$ and consists in a simple shear-layer configuration that can be encountered in slot burners; the flow is confined by means of two no-slip adiabatic walls (at left and right of the domain). At the bottom of the computational domain there is the inlet, while the outlet is located at the top.

In this preliminary work, simulations are perfomed in a two-dimensional framework to reduce computational time. The domain $y \times z$ is $8 \times 10\,\mathrm{mm}$, y and z being the transversal and the streamwise dircetions, respectively: it is discretised by means of 396×700 nodes. The computational nodes are uniformly distributed along the streamwise direction z, with $\Delta z \sim 1.42 \cdot 10^{-5}\,\mathrm{m}$, and stretched from the center towards the sides along the transversal direction y, with $\Delta y_{min} \sim 8.8 \cdot 10^{-6}\,\mathrm{m}$ and $\Delta y_{max} \sim 5.74 \cdot 10^{-5}\,\mathrm{m}$; the aspect ratio $\Delta z/\Delta y$ is in the range $[0.25, 1.61]$.

The fuel is $\mathrm{CH_4}$ and is injected centrally at $50\,\mathrm{m/s}$ and $388\,\mathrm{K}$; its width is $4 \cdot 10^{-4}\,\mathrm{m}$. The oxidant $\mathrm{O_2/CO_2}$ mixture flows on both sides of the methane, and a pilot flame is imposed at the inlet between the two streams of reactants to force ignition. Each oxidant jet flows at $100\,\mathrm{m/s}$ and $450\,\mathrm{K}$; their width is $1.95 \cdot 10^{-4}\,\mathrm{m}$. The pilot flames imposed at the inlet exhibit a temperature distribution ranging from 900 to $2049\,\mathrm{K}$ and a coherent distribution of chemical species; such data come from separate calculations previously performed; these hot gases flow at $5\,\mathrm{m/s}$ and are $10^{-4}\,\mathrm{m}$ widen. Adjacent to each of the oxidant streams, there is a coflowing stream of $\mathrm{CO_2}$ at $50\,\mathrm{m/s}$ and $973\,\mathrm{K}$.

The (isotropic) turbulence characteristics of the jets are specified in terms of velocity fluctuations and auto-correlation length-scales: $u' = 5\,\mathrm{m/s}$ and $l_y = 3 \cdot 10^{-5}\,\mathrm{m}$ for the methane jet; $u' = 1\,\mathrm{m/s}$ and $l_y = 5 \cdot 10^{-5}\,\mathrm{m}$ for the pilot flames; $u' = 10\,\mathrm{m/s}$ and $l_y = 5 \cdot 10^{-5}\,\mathrm{m}$ for the oxidant jets; $u' = 5\,\mathrm{m/s}$ and l_y ranging from 2 to $6 \cdot 10^{-5}\,\mathrm{m}$ for the $\mathrm{CO_2}$ coflowing streams. The length-scale l_y is the (transversal) scale on the inlet plane, while the orthogonal one is $l_z = 2l_y$. The methane jet Reynolds number (based on its bulk velocity and diameter) is nearly 142500 while its turbulent

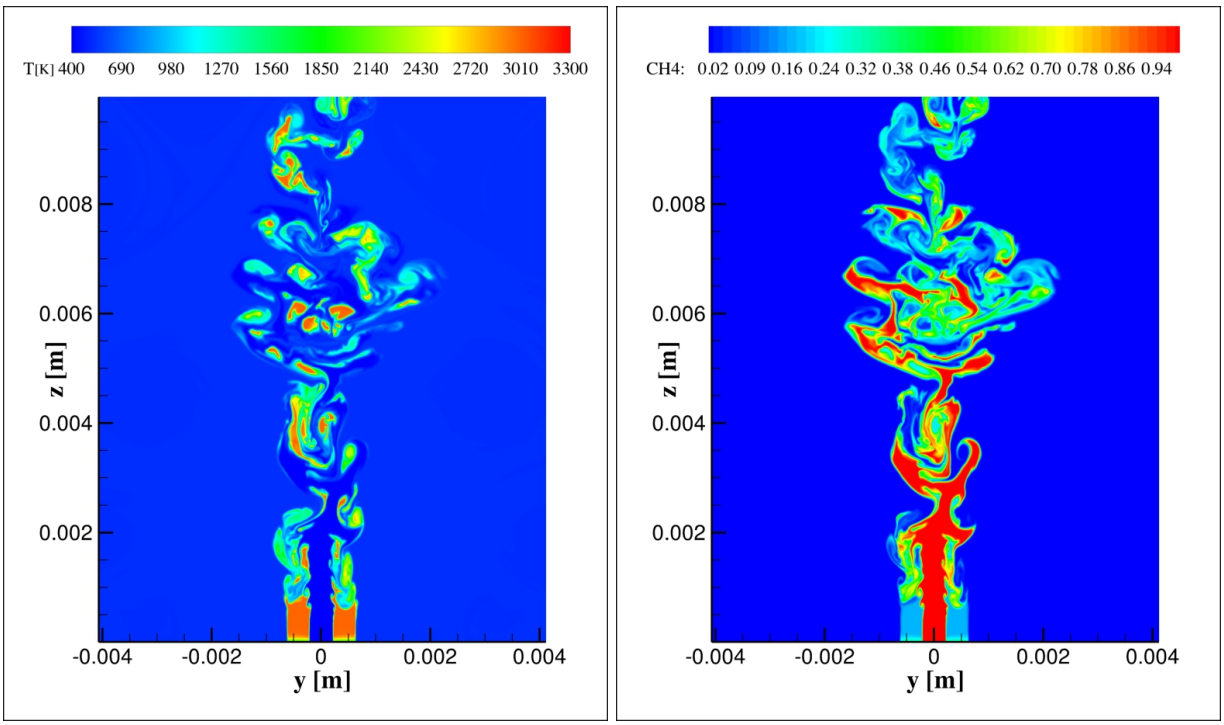

Figure 1: Case 10% O_2, $10^3 \times \kappa_P$, with a forced wide pilot flame. Instantaneous distribution of temperature and methane mass fraction.

counterpart (based on the imposed velocity fluctuation and length-scale) is around 1070. The oxidant jet Reynolds number is nearly 141500, while its turbulent counterpart (Re^t_{Ox}) is around 3600 for the 90% O_2 condition; inceasing dilution with CO_2 results in higher values. The smallest dissipative scale expected comes from the Re^t_{Ox} and is nearly 10^{-7} m.

Three compositions of the oxidant mixture are investigated, 90, 30 and 10% of O_2 by mass. The less diluted condition is simulated firstly without considering the radiant transfer of energy (case $NO\text{-}RTE$), then switching on the M_1 model with two different levels of absorption: in the case named $1 \times \kappa_P$ the individual species Planck mean absorption coefficients used at atmospheric pressure are adopted, while in the case $10^3 \times \kappa_P$ they are intensified by a factor 10^3. The 30% O_2 case is simulated only at the intensified condition $10^3 \times \kappa_P$. The 10% O_2 case is simulated without the RTE model and with a wider pilot flame at the inlet, necessary to force ignition.

It is observed that the present simulations are aimed at better understanding the physics of methane oxy-combustion in a s-CO_2 environment: no particular strategies are adopted to enhance injection of reactants from the point of view of mixing. With the simple shear-layer configuration chosen, the 30% O_2 case with a small pilot flame results in no ignition of the mixture apart from some small flame pockets released downstream from the forced anchoring regions, rapidly quenched by the high aerodinamic stretching. The 10% O_2 case with a wider pilot flame results in more and larger reacting pockets characterizing a not efficient combustion (see Fig. 1): this happens for both $NO\text{-}RTE$ and $10^3 \times \kappa_P$ cases. These results higlight that future studies should be focused on injection strategies to ensure efficient combustion with diluted oxygen streams, at least 30% by volume as imposed primarily by safety reasons in power plants [39]. Hence, since stable and efficient combustion has been achieved only in the 90% O_2 by mass case (corresponding to nearly 92% by volume), in the following the attention will be focused on the analysis of this case.

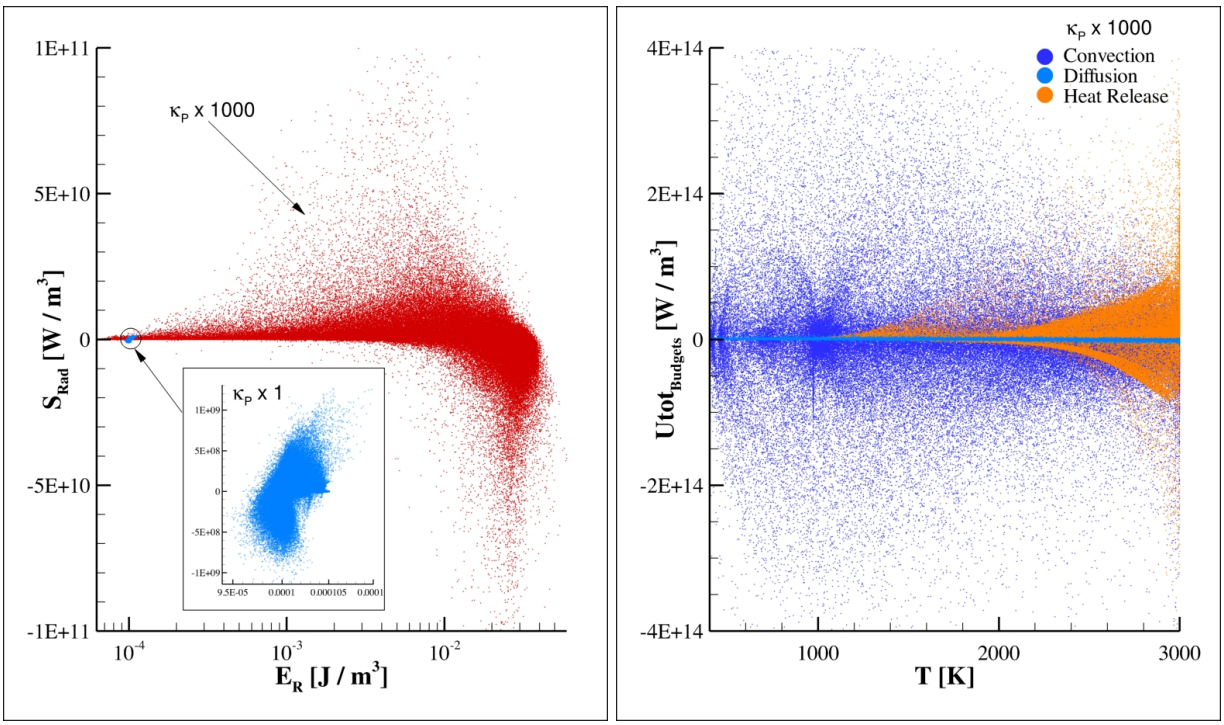

Figure 2: Case $90\%\ O_2$. Instantaneous distributions of the radiative source term in the total energy transport equation versus the radiant energy density for two intensifying factors of the Planck mean absorption coefficient κ_P (left). Instantaneous distributions of energy budgets related to convection, diffusion and heat release terms contributing to the total energy transport equation (right); contributions from viscous and gravity force works are lower and hence not reported.

EFFECT OF RADIANT TRANSFER OF ENERGY AND FLAME STRUCTURE

As already stated, only the results obtained for the lowest dilution level of the oxygen stream, i.e., 90% O_2 by mass, will be analysed in detail since it is the only condition resulting in a stably anchored flame. Radiation effects were examined by performing three simulations related to the three cases $NO\text{-}RTE$, $1 \times \kappa_P$ and $10^3 \times \kappa_P$.

Comparing results obtained without considering the RTE model with those obtained using different intensification factors of the Planck mean absorption coefficients, very little differences appear in the flame structure. This happens despite the large differences in the radiation source/sink term of the energy transport equation shown in Fig. 2 (left). This behaviour can be easily understood by comparing the energy budgets of the different terms contributing to the total energy transport and shown in Fig. 2 (right). The order of magnitude of the work done by the gravity force is $10^5\,\mathrm{W/m^3}$; the viscous work is of the order of $10^8\,\mathrm{W/m^3}$; since they are order of magnitudes lower than the other terms, they are not shown. Then, in the ordered list of budgets there is the radiative source term with $10^{10}\,\mathrm{W/m^3}$, followed by the heat diffusion term with $10^{11}\,\mathrm{W/m^3}$. It can be concluded that in the present simulation the effect of radiant transfer of energy is negligible even with the intensifying factor 10^3 for the κ_P. However, accurate calculation of the Planck mean absorption coefficients from high-resolution spectroscopic databases is necessary to really understand if the intensification factor 10^3 here adopted is justifiable, sufficient or not. Besides this, it is reminded that turbulence/radiation interaction has not been taken into account in this work, although its contribution is expected to be enhanced in high-pressure combustors [10, 11].

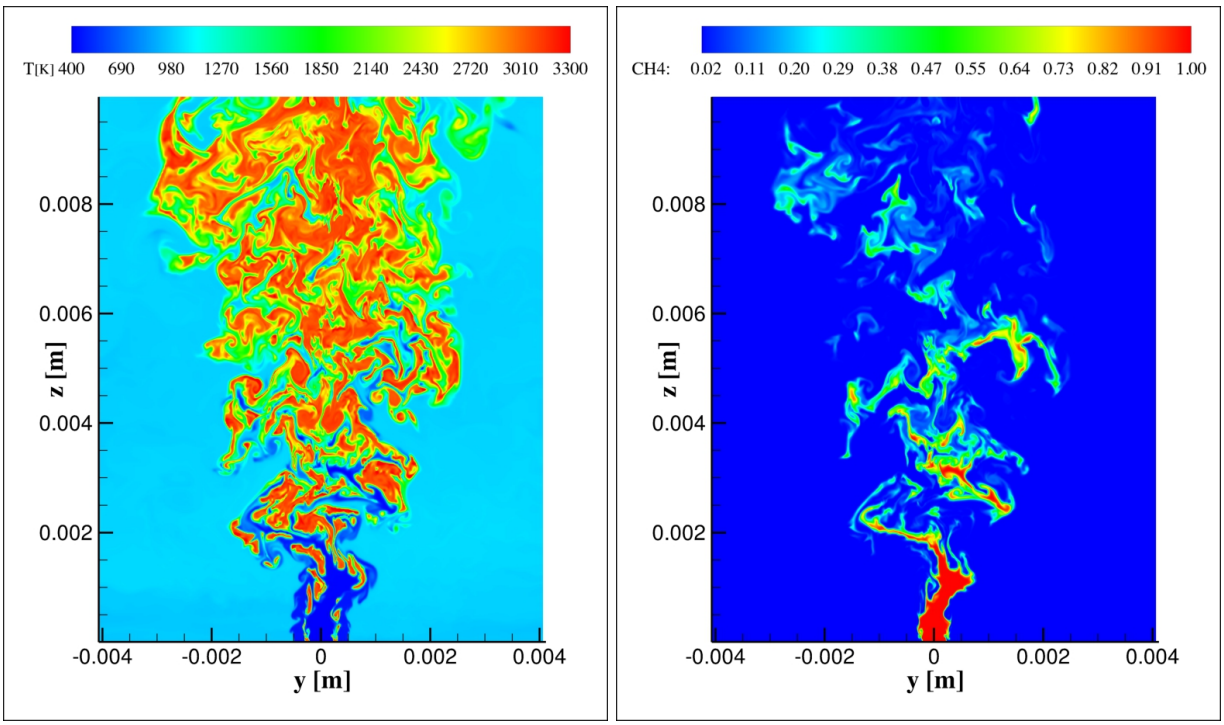

Figure 3: Case 90% O_2, $10^3 \times \kappa_P$. Instantaneous distribution of temperature and methane mass fraction.

It is observed that the distribution in Fig. 2 (left) does not nearly change by substituting the radiant energy density with temperature in the abscissa, i.e., high values of E_R corresponds to high values of T. Hence, radiation cooling is active in the hottest regions of the flame but it produces a negligible effect, i.e., peak temperatures do not decrease due to radiation transport. As a matter of fact, the competition between the enhanced radiative cooling expected and the enhanced chemical kinetics (both due to the high pressure) is here dominated by chemistry. This result is in agreement with what already observed in [2, p. 139] for high-pressure flames in air. This behaviour is here understood by noting that the heat release budget in the energy transport equation is four order of magnitudes greater than the radiative budget, as shown in Fig. 2. Instantaneous distributions of the radiative energy density and of the source term in the total energy transport equation for the case $10^3 \times \kappa_P$ are shown in Fig. 4. In particular, the radiative source/sink term is localized in thin layers. Although not shown, the results evidenced that radiative cooling (associated to negative values of S_{rad}) in this flame is limited to a very thin layer localized around the stoichiometric mixture fraction and it is more frequent in the hot products side ($\mathcal{Z} < \mathcal{Z}_{st}$).

Once clarified that in the present simulations the effect of radiation is negligible, let's examine the structure of the flame. The flame is stably anchored, showing small reacting pockets mainly aligned in the streamwise direction close to the injection, and evolving into larger scale reacting regions moving downstream, as revealed by the temperature snapshot in Fig. 3 (left). The reacting structures are thinner than those typically encountered at lower pressures: this is due to the accelerated chemical kinetics promoted by the high-pressure condition. The methane distribution associated to the same instant is reported in Fig. 3 (right) showing that the jet is corrugated by turbulence without exhibiting any laminar region on its boundaries. High-momentum O_2 ligaments penetrate the methane jet as well as CH_4 fingers also develop into the coflowing stream, thus producing isles of fuels later developing in reacting pockets.

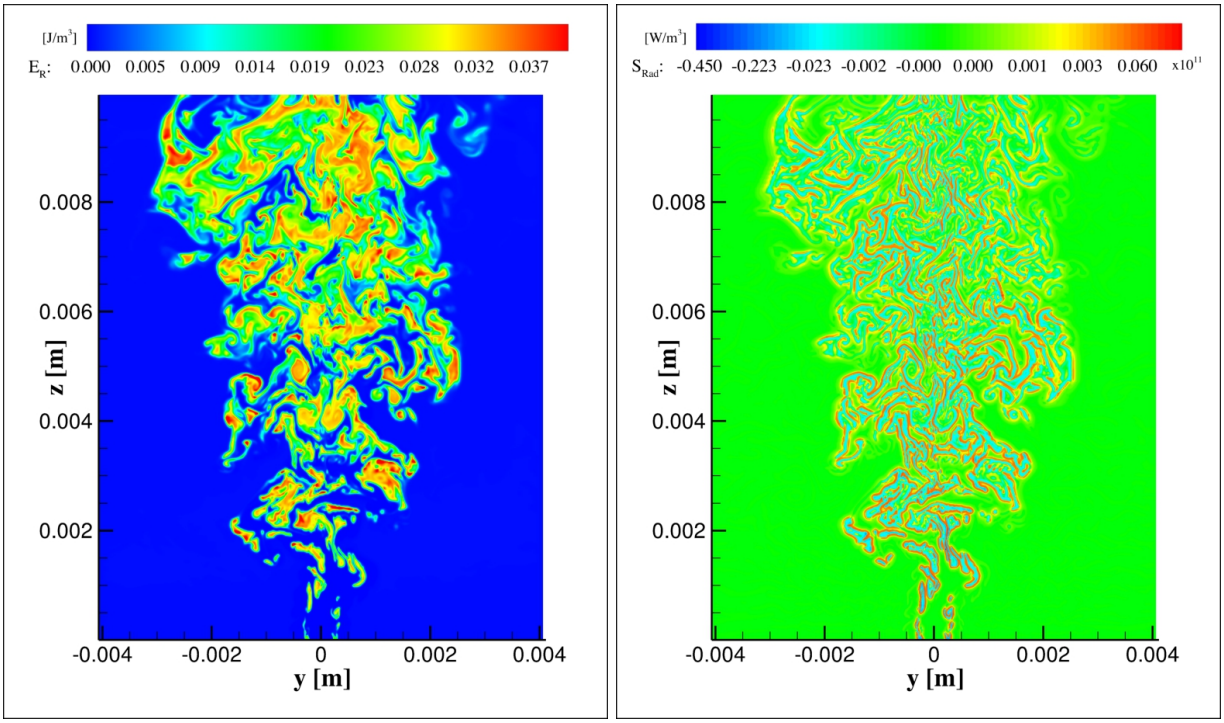

Figure 4: Case 90% O_2, $10^3 \times \kappa_P$. Instantaneous distributions of the radiative energy density and of the source term in the total energy transport equation.

STANDARD AND EFFECTIVE DIFFUSION TIMES AND CHARACTERISTIC NUMBERS

The attention is now focused on diffusion terms only of the Navier-Stokes equations. Characteristic diffusion times may appear when non-dimensionalising Navier-Stokes equations: the momentum diffusion (viscous) time $\tau_\nu = \delta^2/\nu$ (δ being a reference length scale, ν the kinematic viscosity), the mass diffusion time $\tau_{D_i} = \delta^2/D_i$ (D_i being the diffusion coefficient of the i-th species into the rest of mixture), the heat diffusion time $\tau_\alpha = \delta^2/\alpha$ ($\alpha = \kappa/(\rho C_p)$ being the thermal diffusivity, with κ the thermal conductivity, ρ the density and C_p the specific heat at constant pressure). The related non-dimensional numbers are: the Reynolds number Re, defined as the ratio between the momentum diffusion and convective times (the convective time being $\tau_c = \delta/U$ with U a reference velocity), the Schmidt number Sc_i, defined as the ratio between the mass and momentum diffusion times, the Prandtl number Pr, defined as the ratio between the heat and momentum diffusion times, the Lewis number Le_i, defined as the ratio between the mass and heat diffusion times.

Non-dimensional parameters can give a quick information about the relative importance of physical mechanisms. For example, the deviation of the Lewis number from unity is commonly accepted to well represent the unbalanced influence of thermal to mass diffusion. Especially for modelling purposes, some authors addressed the need to define an “effective” Lewis number [40, 41]. Authors in [42] observed that, not only the calculation of Le may be biased by steep gradients, but also its meaning may be questioned due to the off-diagonal terms of the diffusive transport matrix (Soret, Dufour, ...).

In [29, 30] a method to calculate effective characteristic times (and then the related non-dimensional numbers) from the Navier-Stokes equations terms was suggested. In particular, the local momen-

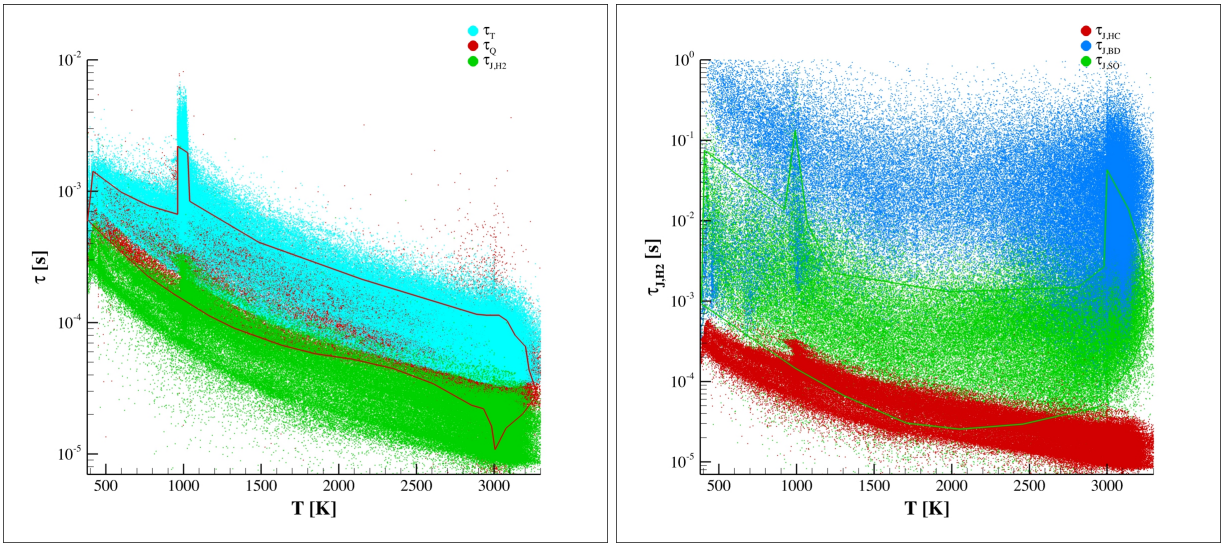

Figure 5: Case 90% O_2, $10^3 \times \kappa_P$. Instantaneous distributions of effective diffusive times (left): the edges of the heat diffusion time distribution are marked by a red line to help the reader. The associated Hirschfelder-Curtiss, baro-diffusion and Soret contributions to the hydrogen effective mass diffusion time are also shown (right): the edges of the Soret diffusion time distribution are marked by a green line.

tum (or viscous), heat and mass diffusion times are modelled as (h_s being the sensible enthalpy):

$$\tau_{\mathcal{T}} \sim \rho \frac{|\nabla \boldsymbol{u}|}{|\boldsymbol{\mathcal{T}}|} \Delta^2, \quad \tau_Q \sim \rho \frac{|\nabla h_s|}{|\boldsymbol{Q}|} \Delta^2, \quad \tau_{J_i} \sim \rho \frac{|\nabla Y_i|}{|\boldsymbol{J}_i|} \Delta^2. \tag{6}$$

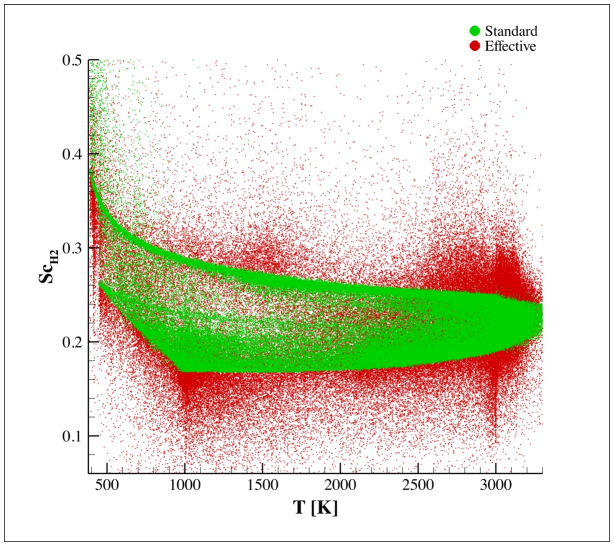

Figure 6: Case 90% O_2, $10^3 \times \kappa_P$. Instantaneous distributions of the standard and effective Schmidt numbers of hydrogen.

Effective diffusion time distributions are reported in Fig. 5 (left). The hydrogen is selected as the most appropriate species to evaluate the mass diffusion time, due to its low mass. Comparing the three diffusion times, it is deduced that the hydrogen mass diffusive mechanism is the fastest, followed by the heat and momentum diffusion. It is also observed that there is an overlapping region, and in particular heat and H_2 mass diffusion mechanisms can interact. Among the different mechanisms contributing to H_2 mass diffusion, the most important is that related to the Hirschfelder-Curtiss law, as shown in Fig. 5 (right). The Soret effect can be as fast as it at intermediate temperatures, while the baro-diffusion can be neglected.

Through the effective diffusion times, effective characteristic numbers can be evaluated and compared to their standard counterpart. Figure 6 shows the instantaneous distributions of the hydrogen Schmidt number: the effective and standard distributions look very similar, although the

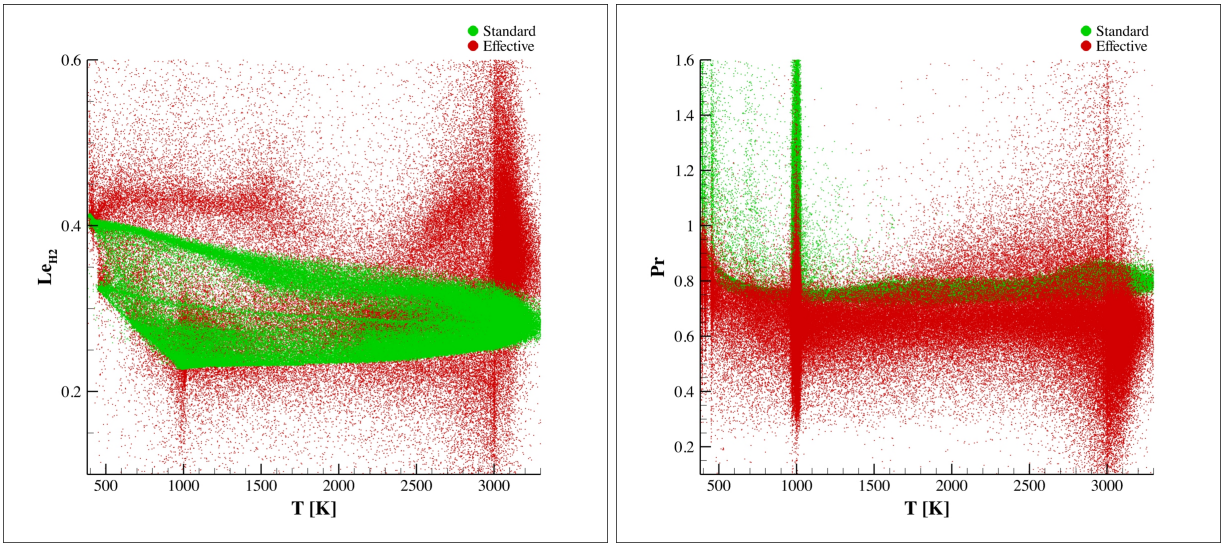

Figure 7: Case 90% O_2, $10^3 \times \kappa_P$. Instantaneous distributions of the standard and effective Lewis number of hydrogen (left) and of the Prandtl number (right).

effective one is spread over the standard one. Instead, the effective Lewis number shows higher values than the standard one, especially in the high temperature regions, as reported in Fig. 7 (left). Similar results were found in [29, 30, 42].

Being $Le = Sc/Pr$, and since the Prandtl number distribution shown in the same figure (right) reveals effective values lower than the standard ones, it can be concluded that the different behaviour of the effective Lewis number with respect to the standard one is due to the effective heat diffusion mechanism. It is observed that in [30] it was found that the effective heat diffusion time largely deviated from its standard quantity mainly for its mass diffusion contribution; although not checked here, this motivation is expected to be confirmed also in the present data.

CONCLUSIONS

The experience gained with the present real gas simulations proves the robustness of the numerical schemes implemented in the HeaRT code to simulate high-pressure oxy-combustion in s-CO_2 atmosphere.

Dilution effect of the oxygen stream by means of carbon dioxide was explored. Fixed the pilot flame imposed at the inlet between the reactant jets, it is found that dilution produce flame anchoring problems and not efficient combustion in the present shear-layer configuration. Hence, in the future attention should be focused on the design of injection strategies to enhance turbulent mixing at highly diluted O_2/CO_2 conditions.

The effect of radiant transfer of energy was taken into account by means of the M_1 diffusive model. Individual species' Planck mean absorption coefficients typically used at atmospheric pressure were assumed. Since they are expected to increase by increasing pressure, the effect of simply multiplying them by 1000 was investigated. In the future they will be accurately calculated by using high-resolution spectroscopic databases. It is observed that turbulence-radiation interaction is neglected for the time being, although it is expected to play an important role in flame cooling at the high-pressure conditions of the present simulations.

The chemical mechanism adopted in this work is too simple, it lacks of the main radical species and it is not validated at the extreme pressure conditions of the present numerical experiment. Hence,

these simulations have to be considered as a first step before facing a more complex kinetics. The inclusion of radical species is expected not only to decrease the peak temperatures but also to affect ignition delay times and flame speed. Furthermore, the methane cracking reactions should be included in case of temperatures as high as those found in these simulations.

An original procedure already developed by the authors was adopted to estimate effective characteristic diffusion times. Effective characteristic numbers were also derived as ratio of effective times and compared to their standard counterparts. Conclusions already found in previous work by using different real gas flow simulations are here confirmed: the effective Prandtl and Lewis numbers differ from their standard counterparts. This may have strong implications in subgrid scale modelling of turbulent combustion in real gas flow simulations.

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