



# Article The Effects of Differential Diffusion on Turbulent Non-Premixed Flames LO<sub>2</sub>/CH<sub>4</sub> under Transcritical Conditions Using Large-Eddy Simulation

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Abstract: In this paper, a large-eddy simulation (LES) of turbulent non-premixed  $LO_2/CH_4$  combustion under transcritical conditions is performed based on the Mascotte test rig from the Office National d'Etudes et de Recherches Aérospatiales (ONERA), and the aim is to understand the effects of differential diffusion on the flame behaviors. In the LES, oxygen was injected into the environment above the critical pressure while the temperature was below the critical temperature. The flamelet/progress variable (FPV) approach was used as the combustion model. Two LES cases with different species diffusion coefficient schemes-i.e., non-unity and unity Lewis numbers-for generating the flamelet tables were carried out to explore the effects of differential diffusion on the flame and flow structures. The results of the LES case with non-unity Lewis numbers were in good agreement with the experimental data. It was shown that differential diffusion had evident impacts on the flame structure and flow dynamics. In particular, when unity Lewis numbers were used to evaluate the species diffusion coefficient, the flame length was underestimated and the flame expansion was more significant. Compared to laminar counterflow flames, turbulence in jet flames allows chemical reactions to take place in a wider range of mixture fractions. The density distributions of the two LES cases in the mixture fraction space were very similar, indicating that differential diffusion had no significant effects on the phase transition under transcritical conditions.

**Keywords:** large-eddy simulation; transcritical conditions; differential diffusion; flame structure; flow dynamics

## 1. Introduction

Combustion under high pressure is used to improve thermodynamic efficiency and reduce pollutant emission in many combustion devices, such as liquid rocket engines, aircraft engines, and diesel engines [1]. The fluids in these engines can locally or even globally be in a supercritical state, under which condition the properties of the fluid are significantly different from those of a classical atmospheric fluid [2]. In practical engines, such as liquid rocket engines, the transcritical state (i.e., when the pressure exceeds the critical pressure of the fluid, but the temperature is lower than its critical temperature) is a common thermodynamic state for reactant injection, with the surface tension and latent heat of vaporization of the liquid being reduced, where jet dynamics are controlled by mass transfer through turbulent mixing rather than breakup and atomization [3]. In order to gain a better understanding of the complex process in practical engines, it is necessary to carry out fundamental studies on turbulent flames under transcritical conditions.

Some experimental studies have been devoted to turbulent flames, where oxygen was injected in a dense transcritical state, while the fuel (either  $H_2$  or  $CH_4$ ) was a light supercritical fluid (i.e., at a temperature above the critical value). In experiments based on the Mascotte test rig from the Office National d'Etudes et de Recherches Aérospatiales (ONERA) [4–6], it was observed that the structures of flames under transcritical conditions



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**Copyright:** © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). were significantly different from those in which the reactants were all subcritical or supercritical. In transcritical oxygen/gaseous hydrogen or methane flames, turbulent flames generated by the injector lip wrapped around the central cold oxygen jet. Turbulent mixing has become the most important factor in the combustion process. The mass flux from the high-density area to the low-density area determines the rates of reactions. However, due to the limitations of experimental diagnostics, quantitative results for the analyses of transcritical flames are still lacking.

Numerical simulations were also carried out to understand turbulent flames under high-pressure conditions. Reynolds-averaged Navier-Stokes (RANS) simulations were able to provide good predictions of the mean flame shape [7–9] for turbulent combustion under transcritical conditions. However, it is impossible to provide information on unsteady flame and turbulence structures in RANS simulations. With the increase in computing power, large-eddy simulations (LESs) have been used to predict turbulent flow [10] and combustion [11-13] under supercritical conditions. Unsteady turbulent flame structures in transcritical flows have also been studied using LES [14–17]. For example, Laurent et al. [18,19] studied flame-wall interactions near the injector lip and the heat release response to fuel inflow acoustic harmonic oscillations in CH<sub>4</sub> oxy-combustion at high pressures. Lacaze et al. [20] and Dahms et al. [21] explained the dynamics of the transition from the classical spray atomization process to a single-phase continuous dense fluid at high pressures. Wang et al. [22,23] investigated the flame dynamics of liquid biswirl injectors in transcritical conditions. Various geometric parameters were examined in depth to explore their influence on flame characteristics. Gao et al. [24] extended the flamelet/progress variable (FPV) model for non-adiabatic combustion with the heat release damping approach. The calculation results correctly predicted the flame lift heights and maximum temperature position. Wada et al. [25] investigated the breakup mechanism of an LOX core in liquid oxygen/gaseous hydrogen (LOX/GH<sub>2</sub>) supercritical combustion. In the reactive case, the LOX core was found to break up further downstream than in the inert case due to the turbulence being suppressed by the volume dilatation and viscous forces around the LOX core.

Differential diffusion, which characterizes the physical process of specific transport rates of species in mixtures, has been studied to some extent by previous researchers. Sevault et al. [26] experimentally reported the differential diffusion phenomenon in CO<sub>2</sub>diluted non-premixed oxy-fuel flames. Hansinger et al. [27] performed a numerical simulation of this experiment by using the transported probability density function (TPDF) method. Differential diffusion was considered by incorporating species' individual molecular diffusivities. The local extinction and the differential diffusion effect in the near field were well reproduced in the simulation. Jung et al. [28] studied auto-ignited laminar lifted methane/hydrogen jet flames and revealed that the differential diffusion of hydrogen molecules led to an unusually decreasing liftoff height.

Many combustion models applied in LESs are based on the mixture fraction, which is considered a passive scalar. One condition for the mixture fraction to be a passive scalar is for all species to have equal mass diffusion coefficients [29]. However, when differential diffusion exists in the combustion process, an additional term due to the differential diffusion of species appears in the governing equation for the mixture fraction. To ensure the accuracy of the results of numerical simulations, the influence of differential diffusion needs to be considered. Wang [30] introduced a differential diffusion term due to fuel with a non-unity Lewis number in the filtered governing equation of the mixture fraction. Wen et al. [31–33] performed an LES by using the flamelet method and compared the LES results with the experimental data of a Darmstadt flame. The coupling effect of mixture stratification and differential diffusion on the flame structure was analyzed. Gierth et al. [34] and Han et al. [35] chose Bilger's mixture fraction [36] as one of the flamelet parameters to take into account when studying the effect of differential diffusion. The transport equations for major species were solved for the calculation of the Bilger mixture fraction.

Under high-pressure conditions, some numerical studies were also carried out to understand the effects of differential diffusion in laminar flame configurations. Gao et al. [37] discussed the real-fluid effects of laminar premixed and non-premixed hydrothermal flames, and they pointed out the sensitivity of laminar hydrothermal flames to the Lewis number assumption. Kim et al. [38] extended the flamelet equation in the mixture fraction space to calculate gaseous hydrogen and cryogenic liquid oxygen laminar flames. The effects of real-fluid and differential diffusion on the local flame structures in liquid rocket engines were explored. Guven et al. [39] emphasized the importance of non-ideal transport under supercritical conditions, and the diffusion velocities of species were described by using the gradient of the chemical potential coupled with the Peng-Robinson equation of state. Yao et al. [40] used different mass diffusion models to calculate laminar counterflow flames for typical rocket propellants under transcritical and supercritical conditions. It was shown that the effects of differential diffusion on phase stability and pseudo-boiling were limited, even in high-pressure states. Despite the above-mentioned studies, the effect of differential diffusion on complex non-premixed turbulent flames under transcritical conditions when using LESs is still an open question.

In this context, LESs of turbulent non-premixed  $LO_2/CH_4$  combustion under transcritical conditions, which was reported in previous experiments [6], were performed in the present work to explore the effects of differential diffusion. The objectives are as follows. First, the real-fluid models are validated, and the LES results are compared with those of the experiments. Second, two LES cases with different Lewis number assumptions are carried out, and their results are compared in the physical space. Finally, the flame structures are further discussed in the mixture fraction space to understand the influence of differential diffusion.

## 2. Numerical Methods

#### 2.1. Real-Fluid Model

When the pressure of a fluid exceeds its critical value, the estimation of the fluid's properties by using the ideal gas assumption would cause deviations, so corrections are needed to account for the real-fluid effects. Based on previous studies, the Peng–Robinson equation of state (P-R EoS) [41] was employed in this work to replace the ideal gas EoS, which can be expressed as:

$$p = \frac{RT}{V_m - b_m} - \frac{a_m}{V_m^2 + 2V_m b_m - b_m^2}$$
(1)

where  $V_m$  is defined as  $V_m = M_m / \rho$ ,  $M_m$  is the mean molecular weight of the mixture,  $\rho$  is the density, T is the temperature, and R is the ideal gas constant. The coefficients  $a_m$  and  $b_m$  represent the influences of attractive and repulsive forces on the mixture, respectively, and they are calculated by using the mixing rules according to Poling et al. [42].

The departure function [42] of the P-R EoS is used to correct the thermodynamic properties, such as the specific heat capacity  $c_p$  and enthalpy h under supercritical conditions, which are determined as:

$$h = G - T\left(\frac{\partial G}{\partial T}\right)_{p,X} = h^0 + pv - RT + K_1\left(a_m - T\frac{\partial a_m}{\partial T}\right)$$
(2)

$$c_p = \left(\frac{\partial h}{\partial T}\right)_{p,X} = c_p^0 - T \frac{(\partial p/\partial T)^2_{V_{m,X}}}{(\partial p/\partial V_m)_T} - R - T \frac{\partial^2 a_m}{\partial T^2} K_1$$
(3)

$$K_1 = \frac{1}{2\sqrt{2}b_m} ln[\frac{V_m + (1 - \sqrt{2})b_m}{V_m + (1 + \sqrt{2})b_m}]$$
(4)

where  $h^0$  and  $c_p^0$  are the enthalpy and specific heat capacity of the ideal gas obtained through polynomial fitting from the NIST database [43].

The transport properties, such as the viscosity  $\mu$  and thermal conductivity  $\lambda$ , are obtained by using Chung's high-pressure calculation methods [44]:

$$\mu = 36.344 \eta^* \frac{\sqrt{M_m T_{c,m}}}{V_{c,m}^{2/3}} \tag{5}$$

$$\lambda = 31.2 \frac{\mu^0 \Psi}{M_m} (G_2^{-1} + B_6 y) + q B_7 y^2 G_2 \sqrt{T/T_{c,m}}$$
(6)

where  $T_{c,m}$  and  $V_{c,m}$  are the critical temperature and volume of the mixture, respectively,  $\mu^0$  represents the ideal gas reference viscosity, and  $\Psi$  is an empirical correlation. The parameters  $\eta^*$ ,  $G_2$ ,  $B_6$ ,  $B_7$ , q, and y are described in detail in [44] and are not repeated here for brevity.

Using the real-fluid models described above, two LES cases were considered to explore the effects of differential diffusion under transcritical conditions. Flamelet tables were required in the LES, as will be explained in Section 2.2. Non-unity Lewis numbers were adopted to generate the flamelet table in Case 1. The Lewis number is defined as  $Le_k = \lambda/(c_p D_k)$ , where  $D_k$  is the diffusion coefficient of species k. The Lewis number of each species was obtained by calculating the one-dimensional laminar premixed flame with Flamemaster [45] with detailed transport models. The code of Flamemaster used in this work was extended to coupling real-fluid effects [46]. The Lewis numbers of all species were set to unity to generate the flamelet table in Case 2 to eliminate the effects of differential diffusion.

In order to clarify the importance of Lewis number schemes for the flame structure, a comparison of laminar counterflow flames in the mixture fraction space with different Lewis number schemes was carried out and is displayed in Figure 1. The solid lines correspond to the results with non-unity Lewis numbers, and the dashed lines correspond to the results with unity Lewis numbers. Both calculations were carried out under a pressure of 5.59 MPa. The inlet of the oxidizer side consisted of pure oxygen and the temperature was 85 K, while the inlet of the fuel side consisted of pure methane and the temperature was 288 K, which was consistent with the experimental conditions [6]. The scalar dissipation rate at the flame location  $\chi_{st}$  was set to 2 s<sup>-1</sup>. GRI-Mech 3.0 was used as the reaction mechanism for the two cases. The temperature profiles showed that the maximum temperatures of the two flames were close to each other. However, careful discrimination indicates that the mixture fraction corresponding to the peak temperature was slightly biased towards the oxidizer side for the case with non-unity Lewis numbers. The trend of the OH mass fraction distribution was consistent with that of the temperature, as expected, while the peak value of the OH mass fraction with unity Lewis numbers was a little bit higher than that with non-unity Lewis numbers. Meanwhile, it was observed that the peak value of the  $H_2O$  mass fraction was larger when unity Lewis numbers were applied. As for the H mass fraction curve, it was seen that its maximum value in the flame with unity Lewis numbers was almost twice as large as that in the flame with non-unity Lewis numbers. The above results indicate that the Lewis numbers considerably influenced the structures of transcritical LO<sub>2</sub>/CH<sub>4</sub> laminar counterflow flames.



**Figure 1.** The contrast of laminar counterflow flames in the mixture fraction space. Solid lines: non-unity Lewis numbers; dashed lines: unity Lewis numbers.

## 2.2. FPV Model

The flamelet/progress variable (FPV) model [47] was used here as the combustion model. Compared with the steady laminar flamelet (SLF) model [48], the FPV model introduces a progress variable *C* so that all of the state information on the S-curve can be used when building tables. In previous studies [49], it was considered reasonable that the progress variable was defined by linear combinations of the main products' mass fractions. Here, the progress variable *C* is defined as:

$$C = Y_{CO_2} + Y_{H_2O} + Y_{CO} + Y_{H_2}$$
(7)

By solving the steady flamelet equations, any thermo-chemical variable  $\phi$  can be written as:

$$\phi = \phi(Z, \Lambda) \tag{8}$$

where  $\Lambda$  is the sequence number of flamelet solutions along the S-curve, and Z is the mixture fraction based on Bilger's definition [36]. Since the progress variable C can also be considered a specific thermo-chemical variable, it is expressed as:

$$C = C(Z, \Lambda) \tag{9}$$

Assuming that a unique inversion exists in Equation (9) [47], then  $\Lambda$  can be determined with a given *Z* and *C*:

$$\Lambda = C^{-1}(Z, C) \tag{10}$$

Therefore, any thermo-chemical variable  $\phi$  can be regarded as a function of the mixture fraction *Z* and progress variable *C*:

$$\phi = \phi(Z, C^{-1}(Z, C)) = \Phi(Z, C)$$
(11)

In the framework of an LES, the filtered variable needs to be integrated by sub-filter probability density functions (PDFs). Here, the  $\beta$ -PDF is used for *Z* and the  $\delta$ -PDF is used for *C*. Therefore, the filtered variable can be written as:

$$\tilde{\Phi} = \iint \Phi(Z,C) P(Z;\tilde{Z},Z^{\tilde{\prime}\prime 2}) P(C;\tilde{C}) dZ dC = \tilde{\Phi}(\tilde{Z},Z^{\tilde{\prime}\prime 2},\tilde{C})$$
(12)

In the FPV model, the transport equations of  $\tilde{Z}$ ,  $Z^{\tilde{\prime}\prime_2}$ , and  $\tilde{C}$  are solved [29] in the following form:

$$\frac{\partial(\bar{\rho}\bar{Z})}{\partial t} + \frac{\partial\bar{\rho}\tilde{u}_i\bar{Z}}{\partial x_i} = \frac{\partial}{\partial x_i} [(\bar{\rho}\tilde{D} + \frac{\mu_t}{Sc_t})\frac{\partial\bar{Z}}{\partial x_i}]$$
(13)

$$\frac{\partial(\bar{\rho}Z^{\tilde{\prime}'2})}{\partial t} + \frac{\partial(\bar{\rho}\tilde{u}_iZ^{\tilde{\prime}'2})}{\partial x_i} = \frac{\partial}{\partial x_i}[(\bar{\rho}\tilde{D} + \frac{\mu_t}{Sc_t})\frac{\partial Z^{\tilde{\prime}'2}}{\partial x_i}] + 2(\frac{\mu_t}{Sc_t})(\frac{\partial\tilde{Z}}{\partial x_i}) - \bar{\rho}\tilde{\chi}$$
(14)

$$\frac{\partial(\bar{\rho}\tilde{C})}{\partial t} + \frac{\partial\bar{\rho}\tilde{u}_{i}\tilde{C}}{\partial x_{i}} = \frac{\partial}{\partial x_{i}}[(\bar{\rho}\tilde{D} + \frac{\mu_{t}}{Sc_{t}})\frac{\partial\tilde{C}}{\partial x_{i}}] + \bar{\omega_{C}}$$
(15)

where  $\bar{\rho}$  and  $\tilde{u}_i$  denote the filtered density and Favre filtered velocity in the *i*th direction, respectively.  $\mu_t$  is the subgrid eddy viscosity calculated with the Smagorinsky model [50].  $Sc_t$  is the turbulent Schmidt number and was set to 0.7.  $\bar{\omega}_C$  is the source term of the progress variable.  $\tilde{D}$  is the filtered molecular diffusivity. In an LES, the calculation of  $\tilde{\chi}$  is decomposed into the solved large-scale part and the unsolved subgrid part as follows:

$$\tilde{\chi} = 2\tilde{D}(\nabla\tilde{Z})^2 + 2\frac{\mu_t Z^{\tilde{\prime}'2}}{Sc_t \Delta^2}$$
(16)

In the above equations,  $\bar{\rho}$ ,  $\tilde{D}$ , and  $\bar{\omega}_C$  are obtained from the flamelet table. Note that the differential diffusion term [51] in the governing equation of the mixture fraction is neglected in the present work, and this has been implemented in the same way in previous studies [52,53]. For filtered variables, a three-dimensional table indexed by  $\tilde{Z}$ ,  $Z^{\tilde{I}'2}$ , and  $\tilde{C}$  can be generated. Note that all thermodynamic and transport properties used in the FPV model were predicted with the real-fluid models discussed in Section 2.1.

### 3. Experimental and Numerical Setup

The LES configuration of the present work was based on G2 of the Mascotte test rig from ONERA [8], which has been studied both experimentally and numerically. In the experiment, a dense oxygen stream was injected by a single coaxial injector with an inner diameter of  $d_0 = 5$  mm at a low velocity, and this was surrounded by an annular high-velocity methane stream with an outer diameter of  $d_1 = 10$  mm. The pressure in the chamber was set to 5.59 MPa, which was above the critical values for both streams. The inlet temperatures were 85 and 288 K for the oxygen and methane stream, respectively. Therefore, the injected oxygen was transcritical and liquid-like, while the injected methane is supercritical. The operating parameters are listed in Table 1, where  $p_R$  and  $T_R$  are the reduced temperature and pressure, respectively, which were obtained through normalization by using their respective critical values.

Table 1. Operating parameters.

Species	<i>ṁ</i> (g/s)	$T_R$	$P_R$
O <sub>2</sub>	44.4	0.55	1.11
CH4	143.1	1.51	1.22

Figure 2 shows the geometry of the computational domain for the LES, which had a square cross-section of  $50 \times 50 \text{ mm}^2$ . The length of the computational domain was 100 mm. The whole domain was discretized with 4,090,000 points and 4,060,000 hexahedrons. An additional simulation with only half of the grid points was also performed, and the results were similar to those presented in this paper. The cell was stretched downstream, where the flame and flow structures were larger. The smallest cell was located at the inlets between the oxygen and methane streams. A turbulent intensity of 3% was added to the mean velocity profiles of the oxygen and methane streams. A non-slip iso-thermal boundary condition was applied to the walls, and the wall temperature was the same as that of the methane stream, i.e., 288 K. The imposed pressure condition was applied at the outlet. In order to capture the detailed reaction process, a detailed methane mechanism, i.e., GRI-Mech 3.0, which contained 53 species components and 325 elementary reactions, was used to generate the flamelet table [45].



Figure 2. Schematic of the computational domain and the boundary conditions.

In order to completely describe the chemical state of the combustion process, about 200 one-dimensional laminar counterflow flames with different stoichiometric scalar dissipation rates were calculated with extended Flamemaster [46], which included the whole real-fluid models. The flamelet table was generated through coordinate transformation and integration of these flamelet calculation results. The size of the flamelet table for each variable was  $101 \times 10 \times 401$  grids for  $\tilde{Z} \times Z^{\tilde{\prime}'2} \times \tilde{C}$ .  $Z^{\tilde{\prime}'2}$  was normalized and uniformly distributed on a scale from 0 to 1. To improve the accuracy of the simulations,  $\tilde{Z}$  and  $\tilde{C}$ were unevenly distributed and encrypted in the reaction zone. The maximum value of C was 0.94. The LESs were carried out by using a solver called scFPVFoam [46], which was developed from the standard solver reactingFoam in the open-source software OpenFoam-2.3.1 [54]. In addition to the governing equations of  $\tilde{Z}$ ,  $Z''^2$ , and  $\tilde{C}$  that were mentioned above, the mass conservation and momentum conservation equations also needed to be solved with scFPVFoam to obtain the velocity and pressure. The variables needed could be obtained from the flamelet table with the results of the governing equations. Therefore, the real-fluid corrections were not coupled in scFPVFoam. The PIMPLE algorithm was used to solve the coupling of the velocity and pressure. The unsteady term was solved by using the implicit Euler scheme, while the convective and diffusion terms were solved by using the second-order central difference scheme. Two LES cases were carried out, as mentioned earlier. In Case 1, non-unity Lewis numbers were used to generate the flamelet table, while in Case 2, unity Lewis numbers were employed. Otherwise, the numerical details of the two cases were the same. For both cases, the simulations ran for 40 ms to reach a statistically steady state, corresponding to about four convective times (based on the oxygen injection velocity and the flame length). Then, the simulations ran another 40 ms to collect the statistics that are presented in the paper. As the flamelet table had the same size in two conditions, each case in the stage of LES used 128 processors for approximately 10 days, requiring 30,000 CPU hours.

### 4. Results and Discussion

In this section, the real-fluid models described in Section 2.1 are first verified, and the LES results are compared with those of the experiment. Then, the flame and flow structures of different LES cases are both qualitatively and quantitatively analyzed. Finally, the flame structure is examined in the mixture fraction space to explore the influence of differential diffusion.

#### 4.1. Model Validation

The accuracy of the real-fluid properties is verified first, as reliable fluid properties are essential for transcritical non-premixed flame simulations. Figure 3 shows the profiles of the densities, constant-pressure heat capacities, viscosities, and thermal conductivities

of the fuel and oxidizer at a supercritical pressure of 5.59 MPa over a certain range of temperatures. The results of the ideal gas and real-fluid models are compared with the values in the NIST database [43]. It can be seen that the predictions of the real-fluid models were much better than that of the ideal-gas model. However, some differences between the real-fluid model and the NIST results were observed as the temperature approached the critical temperature for oxygen (T < 154.6 K), consistently with what was reported in [37]. Methane was injected above its critical temperature (T > 190.6 K), and the real-fluid model was also in good agreement with the values in the NIST database.



**Figure 3.** Comparisons of the real-fluid model, ideal gas model, and NIST data for physical properties of the reactants at 5.59 MPa. Symbol: NIST data; red line: real-fluid model; blue line: ideal gas model. (a) Physical properties of O<sub>2</sub>. (b) Physical properties of CH<sub>4</sub>. For each subgraph, top left: density; top right: constant-pressure heat capacity; bottom left: viscosity; bottom right: thermal conductivity.

The real-fluid models were further validated in laminar flames coupled with the transport properties and chemical reactions, and the results are shown in Figure 4. The non-premixed  $O_2/CH_4$  laminar counterflow flame under supercritical pressure was calculated and compared with existing data from the literature [55]. The operating pressure was 100 bar, which was above the critical value for both  $O_2$  and  $CH_4$ . The temperature of  $CH_4$  was set to 300 K. The temperature of  $O_2$  was set to 120 K, which was below the critical temperature of oxygen, corresponding to the transcritical condition. Non-unity Lewis numbers were used for the simulation. In the calculation, the strain rate was a = 140 s<sup>-1</sup>, which was defined as the velocity gradient in the oxidizer stream [45]. The profiles of the temperature and various species' mass fractions from the present work were compared with those from Wang et al. [55]. It was seen that very good agreement was obtained for the distributions of various quantities, thus validating the real-fluid model and transport properties that were employed in the present work.



**Figure 4.** Validations of laminar counterflow diffusion flames at 100 bar (the solid lines indicate the present work and the symbols represent the data from Wang et al. [55]).

In the following, the LES results are validated against the experimental measurements. As mentioned in Section 3, Case 1 included the real-fluid model with non-unity Lewis numbers, which was more realistic than Case 2, so the predictions from Case 1 for G2 of the Mascotte test rig were compared with the experimental results. Note that under the very-high-pressure cryogenic condition, only a few experimental data are available. The most quantitative data were the signals of OH<sup>\*</sup>, which characterized the flame shape [5]. In the present work, the OH mass fraction was used to approximate the OH<sup>\*</sup> signal, which was proved to be a reasonable approximation [9]. In the previous work of Schmitt [13], the chemical species were considered to have equal diffusivities, and the transport equation for each species needed to be solved. The flame structure was characterized by the distribution of the mean heat release. It can be seen from the simulation results that although the flame length was consistent with the experimental data, the expansion of the flame tail was overestimated. The upper panel of Figure 5a shows the distributions of the mean OH mass fraction from Case 1, while the lower panel shows the Abel transform of the OH<sup>\*</sup> signal from the experiment [13]. It was seen that the reaction layer was thin near the nozzle, and it was progressively broadened with increasing downstream distance. The expansion angle of the flame was large close to the flame tip at around x/d = 10. After expanding to its maximum, the flame terminated abruptly. Overall, the flame structures of the experiment were well captured by using the LES.



**Figure 5.** Flame and flow structures for Case 1. (a) Upper panel: mean distribution of the OH mass fraction for Case 1; lower panel: Abel transform of OH\* emissions from the experiments [13]. (b) Mean distribution of the axial velocity. The white line shows the iso-line of zero axial velocity.

The abrupt termination of the flame was a result of the coupling of combustion and turbulent flows, which was further examined. The mean axial velocity distribution from Case 1 is shown in Figure 5b. Two recirculation zones were observed in the velocity contour, i.e., one near the wall and the other one near the centerline. The recirculation zone near the wall was formed due to the confinement by the walls, which promoted the movement of the methane jet toward the wall; thus, the expansion angle of the flame rapidly increased. The recirculation zone near the centerline was formed due to the large velocity difference resulting from the low velocity of the oxygen stream and the high velocity of the methane stream. The recirculation zone near the centerline facilitated the abrupt termination of the flame.

#### 4.2. Flame Structure and Flow Dynamics

In this section, the flame and flow structures of two LES cases are presented and compared to explore the effects of differential diffusion. Figure 6 displays the instantaneous LES results of the two cases. Figure 6 shows two iso-surfaces for both LES cases. One is the iso-surface of the Q-vortex, which is colored by the axial velocity. The other is the

iso-surface of the density, which is shown in a gray color. The Q-vortex is defined as  $Q = 0.5(|\Omega|^2 - |S|^2)$ , where  $\Omega$  is the vorticity tensor and *S* is the strain rate tensor. The iso-surface of the Q-vortex characterized the dynamic structure in the flow and the formation of coherent structures near the nozzle, and a rapid transition to a disrupted flow could be observed. This highly turbulent structure wrinkled the flame front, as will be discussed shortly.



**Figure 6.** Instantaneous calculation results in the LES. The iso-surface of the Q-vortex  $(4 \times 10^8 \text{ s}^{-1})$  colored by the axial velocity, and the iso-surface of the density (100 kg/m<sup>3</sup>) for both LES cases: (a) Case 1 and (b) Case 2.

Figure 7 shows the instantaneous distributions of the axial velocity U, temperature T, and mass fractions of OH, O<sub>2</sub>, and H from the LES cases. In the first column, the solid white lines represent the contour lines with an axial velocity of zero (U = 0). It was seen that the instantaneous distributions of the axial velocities for the two cases were very similar, and recirculation zones were observed near the wall and near the centerline. There was a strong shear turbulence layer between the outer methane jet and the oxygen jet core due to the existence of a large velocity gradient. In the temperature contour, the solid white lines indicate the isolines of the stoichiometric mixture fraction ( $Z = Z_{st}$ ). It was found that the high-temperature region was near the stoichiometric mixture fraction, and the front of the flame was wrinkled due to the shear between the methane and oxygen streams. The flame expanded in the downstream region due to the interaction between the flame and the central recirculation zone. As an intermediate species of methane/oxygen combustion, the distribution of OH was similar to that of the temperature. Overall, the reaction zones became thicker with increasing downstream distance. It was noted that the magnitude of the OH mass fraction of Case 2 was slightly larger than that of Case 1, and the expansion of the flame after x > 8d was more obvious. Compared with that in Case 1, the oxygen jet length in Case 2 was slightly shorter, indicating that the reaction rate of oxygen was faster under the conditions with unity Lewis numbers. The difference in the H distribution between the two cases is significant. In particular, the concentration of H in Case 2 was much higher than that in Case 1. This highlights the role of differential diffusion in influencing the flame structure.

The flame structure and flow dynamics can be more quantitatively illustrated by using the mean field distribution. Figure 8 shows the averaged axial velocity and the species' mass fraction distributions for the two cases. The averaging was performed in the azimuthal direction and over time. In Figure 8a, it can be seen that the distributions of the mean axial velocity were very similar. However, the recirculation zone in Case 2 occurred slightly more upstream compared with that of Case 1. It can be seen from Figure 8b,c that the mean flame structures of the two cases were considerably different. At 6d < x < 10d, Case 1 had a smaller flame expansion angle and a thinner reaction region compared with those of Case 2, while the concentrations of OH and H were higher in Case 2 than in Case 1, as was also observed in the instantaneous distributions of species' mass fractions shown in



Figure 7. It can also be seen in the figure that the flame in Case 2 was shorter than that in Case 1.

**Figure 7.** Representative snapshots in a slice of the calculation domain of the axial velocity, temperature, and mass fractions of OH,  $O_2$ , and H in the 3D LES. Case 1 is listed in the first line, while Case 2 is in the second. The solid white lines in the first column indicate the isolines that the axial velocity is equal to 0, and the solid white lines in the second column indicate the isolines of the stoichiometric ratio mixture fraction.

Figure 9 compares the radial profiles of various quantities at different axial locations. The top row presents the results of the mean axial velocity. At x = 4d, the axial velocities of the two cases were almost the same, and the difference between the two cases increased along the flow direction. The recirculation zone near the wall in both cases was reflected in the profiles of large values of r. It was seen that the peak velocity of the two cases moved slowly in the radial direction with increasing axial distance, and that in Case 2 was further away from centerline compared with that in Case 1 in the downstream. The second row shows the mean OH mass fraction profiles at different axial locations. It was confirmed that, at the first three axial locations, the OH mass fraction in Case 2 is higher than that in Case 1, and it had a wider distribution, indicating a thicker reaction region. As a result, the flame was completed earlier in Case 2, and its length appeared to be shorter. The mean H mass fraction profiles are displayed in the third row of Figure 9. The trend of the H mass fraction distribution was similar to that of the OH mass fraction, and it was seen that the mean H mass fraction was higher in Case 2 than that in Case 1 at the first three axial locations. At x/d = 10, the peak H mass fraction in Case 1 was higher than that in Case 2. This indicated that the differential diffusion considerably affected the flame structure and flow structure under transcritical conditions.







**Figure 9.** Radial profiles of mean quantities for the three cases at different axial locations: x = 4d, x = 6d, x = 8d, x = 10d.

## 4.3. Flame Structure in the Mixture Fraction Space

In order to further explore the influence of differential diffusion, Figure 10 shows scatterplots of the OH mass fraction in the mixture fraction space for the two cases. The solid black lines represent the conditional means of the LES results, while the scatter points are colored according to the H mass fraction values. The solutions of three laminar counterflow flames with different stoichiometric scalar dissipation rate are also shown for reference. The stoichiometric scalar dissipation rates of the laminar flames were  $\chi_{st} = 2 \text{ s}^{-1}$ ,  $\chi_{st} = 500 \text{ s}^{-1}$ , and  $\chi_{st} = 6000 \text{ s}^{-1}$ . The results show that differential diffusion led to more

H<sub>2</sub>, H, and other species diffusing to the lean side, which affected the chemical reaction process and, thus, caused the flame to move to the oxidizer side. A similar phenomenon was also found in the studies of Kim [38] and Yao et al. [40]. This was also consistent with the observation that the mixture fractions corresponding to the peaks of the two curves in Figure 1 were different. Compared with the results of laminar counterflow flames, the OH mass fraction of the flames when using the LES had a wider range in the mixture fractions to occur in a wider range of mixture fractions. By comparing laminar counterflow flames with different Lewis number schemes, it was found that the OH mass fraction was more sensitive to scalar dissipation rates when differential diffusion was considered.

The impacts of differential diffusion can be quantitatively measured with the differential diffusion parameter  $Z_{HC}$  [56], which is defined as the difference between the elemental mixture fractions of hydrogen and carbon:

$$Z_{HC} = Z_H - Z_C \tag{17}$$

According to Barlow et al. [56], the mixture fractions for hydrogen and carbon are defined as:

$$Z_H = \frac{Y_H - Y_{H,1}}{Y_{H,2} - Y_{H,1}}, Z_C = \frac{Y_C - Y_{C,1}}{Y_{C,2} - Y_{C,1}}$$
(18)

where the subscripts 1 and 2 represent the fuel side and the oxidizer side, respectively. The variable  $Z_{HC}$  was computed, and the results are shown in Figure 11. In addition to the conditional statistics of the two LES cases, the laminar counterflow flame results when considering differential diffusion with various stoichiometric scalar dissipation rates are also presented. The distributions of  $Z_{HC}$  in Case 2 with unity Lewis numbers did not vary significantly in the mixture fraction space. It can be seen in the figure that all numerical simulation results with non-unity Lewis numbers had obvious effects of differential diffusion. The differential diffusion of  $H_2$  and H to the reaction zone led to a deficit in hydrogen relative to carbon on both the oxidizer side and the fuel side. A positive peak of  $Z_{HC}$  in the reaction zone on the rich side of the stoichiometric mixture fraction could be observed. By comparing the laminar flame results of different stoichiometric scalar dissipation rates, it was found that the differential diffusion effect gradually decreased with the increase in the scalar dissipation rate. This indicated that the differential diffusion effect was sensitive to the imposed scalar dissipation rate.



**Figure 10.** Scatter data colored according to the H mass fraction and the conditional means of the OH mass fraction in the mixture fraction space for (left) Case 1 and (right) Case 2. The solid black line represents the LES's conditional statistical results. The purple curves correspond to laminar counterflow flames with different stoichiometric scalar dissipation rates: (solid)  $2 \text{ s}^{-1}$ , (dashed) 500 s<sup>-1</sup>, and (dash-dotted) 6000 s<sup>-1</sup>. The vertical dashed lines mark the stoichiometric mixture fraction.



**Figure 11.** Differential diffusion parameter  $Z_{HC}$  plotted over the mixture fraction space for two LES cases and laminar counterflow flames with non-unity Lewis numbers. Along the arrow's direction, the dissipation rate of laminar counterflow flames increases, and the values of the scalar dissipation rate are the same as those in Figure 10.

In this work, oxygen was in a transcritical state when it was injected, so the phase transition in the system is an interesting topic. In transcritical flows, the location of the pseudo-boiling point is critical, as it can have significant implications for the flame structure and stability. The pseudo-boiling point can be characterized by a large drop in the density of the fluid, which is shown in Figure 12. It can be seen in the figure that the position where the maximum value of the density gradient appeared corresponded to the same mixture fraction in both cases. The results here show that the unity Lewis number assumption did not have significant impacts on the pseudo-boiling point of the system. This was consistent with the findings of Yao et al. [40]. Figure 12 shows that the positions of the pseudo-boiling points in the two cases were very close to the oxidizer side, while the differential diffusion occurred in the reaction zone near the stoichiometric mixture fraction, as shown in Figure 10. This indicates that the influence caused by differential diffusion mainly occurred in the high-temperature region, i.e., near the flame front, and the effect near the pseudo-boiling point was minor.



Figure 12. Density distribution in the mixture fraction space for the two cases.

### 5. Conclusions

In the present work, the FPV model was used for an LES of turbulent non-premixed  $LO_2/CH_4$  flames under transcritical conditions. Two cases with different Lewis numbers were considered. Because the sizes of the flamelet tables were the same, the two cases had similar computational costs. The influence of the differential diffusion on the flame structure and flow dynamics in G2 of the Mascotte test rig from ONERA was explored.

The main findings about the differential diffusion are summarized as follows. First, the accuracy of the real-fluid models used in this work was comprehensively verified for a single species and in laminar counterflow flames. The flame structure of G2 of the Mascotte test rig was also well captured by the LES. Second, it was shown that different Lewis number assumptions had evident impacts on both laminar counterflow flames and the LES results. The flame length was underestimated and the flame expansion was more significant downstream when using unity Lewis numbers. Finally, it was revealed that differential diffusion caused the chemical reaction to be biased toward the oxidizer side. Turbulence allowed combustion to take place over a more expansive mixture fraction space. Differential diffusion under transcritical conditions did not affect the pseudo-boiling point. In conclusion, it is essential to consider differential diffusion in order to accurately model transcritical turbulent flames using LESs, even though its influence on phase transitions at high pressures is limited.

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