Methane Combustion in MILD Oxyfuel Regime: Influences of Dilution Atmosphere in Co-flow Configuration

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Abstract: MILD (moderate or intense low oxygen dilution) oxyfuel combustion is a recently proposed clean combustion mode which can remedy the shortcomings of the standard oxyfuel combustion technology. Nowadays most available studies on MILD oxyfuel combustion focus on how to realize this new combustion regime in O_2/CO_2 atmosphere. The open research on methane MILD oxyfuel combustion in O_2/H_2O atmosphere is quite sparse. In the present work, we carry out a comprehensive comparison study on methane MILD oxyfuel combustion in different dilution atmosphere for the first time.

The JHC (jet in hot co-flow) burner is adopted as a research prototype. The investigation is based on numerical simulation, so firstly the adopted numerical approach is validated by some experimental data in open literature. The numerical comparison is conducted by varying the mass fraction of oxygen in the co-flow and the temperature of the hot co-flow, two key parameters affecting fine reaction structures in JHC. Through the present investigation, a number of findings are reported for the first time and some conclusions presented in previous publications are checked with analyses, especially on some conflicted claims between the previous publications. In addition, several new questions are raised, which may inspire further research activities in future.

Keywords: MILD combustion; Oxyfuel combustion; Methane; CO_2-dilution; H_2O-dilution; oxy-steam

1 Introduction

MILD oxyfuel combustion [1,2] is a recently emerging term which can be regarded as an organic combination of two promising clean combustion technologies, MILD (moderate or intense low oxygen dilution) combustion and oxyfuel combustion. Originally, some of the present authors proposed this new idea in order to utilize biogas with a higher efficiency [3]. Soon after, it was extended to various fuels [2,4–8]. Through these preliminary studies, it was found that

the MILD combustion regime could be established more easily in oxyfuel condition [1,3,7] and meanwhile a number of shortcomings of the standard oxyfuel combustion technology could be remedied straightforwardly by the introduction of MILD combustion regime [4]. Especially, the experimental efforts [4,7] further demonstrated there was no obvious technical difficulty to establish and to sustain MILD oxyfuel combustion in industrial furnaces. Consequently, MILD oxyfuel combustion may become one of the next generation clean combustion technologies for carbon capture which is crucial to the sustainable development of human society [9]. For this purpose, consecutive research on MILD oxyfuel combustion is essential as our knowledge, as well as available open literature, on it is quite limited [1,2].

Originally, the research on MILD oxyfuel combustion focused on how to realize this new combustion regime in O_2/CO_2 atmosphere, namely oxygen in oxidant flow being diluted by carbon dioxide rather than nitrogen in conventional air-firing mode [2,4–8]. Recently, the present authors discussed the possibility to establish and to sustain MILD oxyfuel combustion in O_2/H_2O atmosphere where oxygen in oxidant flow is diluted by steam rather than carbon dioxide [1]. As shown in Ref.[1], compared with its O_2/CO_2 counterpart, there are at least three advantages to realize MILD oxyfuel combustion in O_2/H_2O atmosphere, such as simpler plant configuration, lower operation cost and high power-generation efficiency. In the oxyfuel combustion research community, the approach to realize oxyfuel combustion in O_2/H_2O atmosphere is named

as steam-moderated oxyfuel combustion or oxy-steam combustion [1,10]. As the chemical and physical properties of steam are quite different from those of CO_2, inevitably, compared with its O_2/CO_2 counterpart, combustion behavior may be significantly altered in the steam-moderated oxyfuel scenario. Consequently, comprehensive comparison of combustion characteristics between in O_2/CO_2 and in O_2/H_2O atmosphere is necessary, as it has done between in O_2/CO_2 (standard oxyfuel combustion) and in O_2/N_2 (air-firing mode) condition [11]. Unfortunately, nowadays the essential studies on this critical topic are extremely sparse. Some of the present authors compared the effects of CO_2- and H_2O-dilution on combustion temperature and reaction kinetics of methane [12]. It was observed that the chemical and thermal effects of CO_2 and of H_2O on combustion behavior of methane are quite different and consequently they will alter combustion temperature and reaction paths of methane in the oxyfuel combustion regime by different ways. Zou et al. investigated steam's effect on temperature distribution in methane oxy-steam combustion [13]. With the aid of numerical simulation, they found out the key elementary reaction step which determined the combustion temperature. In Refs.[14–16], wet recycle of oxy-coal combustion was investigated, not only by numerical simulation but also by experimental approaches. As steam is rich in wet recycle of oxyfuel combustion, it was observed that high concentration H_2O in recycled flue gas could influence combustion characteristics of pulverized coal significantly [14–16]. However, these studies [12–16] all are limited in the so-called "feed-back" combustion regime rather than MILD combustion regime

[1], so whether the conclusions made in these studies are tenable in the MILD oxyfuel combustion regime is still an open question. To the best knowledge of the present authors, on comparison study between CO_2 and H_2O on establishing and sustaining MILD oxyfuel regime, until now perhaps there are only three open publications [1,16,17]. In Ref.[1], the present authors compared the effects of CO_2 and of H_2O on establishing biogas MILD oxyfuel combustion with the aid of a counter-flow configuration. It was found that biogas MILD oxy-fuel combustion would be established more easily in O_2/H_2O atmosphere but meanwhile the reaction zone would become more complicated. Sabia et al. discussed propane auto-ignition delay time in MILD combustion regime, where reactants were diluted by CO_2 and H_2O, respectively [16]. In Ref.[16], a cross-flow configuration was adopted. The authors claimed that in the O_2/H_2O option the auto-ignition delay time would be a little shorter than its O_2/CO_2 counterpart. Recently, some of the present authors conducted a numerical investigation about the influence of H_2O addition on MILD oxy-coal combustion [17]. The concentration of H_2O in oxidant flow varied from 0% (standard O_2/CO_2 condition) to 70% (oxy-steam atmosphere). It was observed that NO emission could be suppressed and heat transfer would be enhanced in O_2/H_2O atmosphere. As the IFRF (International Flame Research Foundation) semi-industrial scale co-flow furnace adopted in Ref.[17] is not an ideal MILD oxyfuel combustion research prototype and the extreme complication of coal combustion, Ref.[17] failed to reveal the influence of different types of dilution gases (H_2O or CO_2) on fine reaction structures. In our latest work

[1], it has been underlined that further research on this topic is necessary as co-flow is more popularly found in practical combustion systems. Especially, through our recent research [12,19], it was observed that the effect of dilution gas on combustion performance in a co-flow configuration may differ from its counter-flow counterpart because flow-reaction interaction, which is excluded in a one-dimension model (e.g. a counter-flow configuration), will play an important role in a co-flow configuration. Consequently, in order to deepen our knowledge in this emerging area so to advance its application in energy industry, a systematic comparison between the performance of co-flow MILD oxyfuel combustion in O_2/H_2O condition and that in O_2/CO_2 atmosphere, is essential.

In order to bridge the aforementioned gap, in this work we numerically investigate methane combustion in MILD oxyfuel regime, diluted by carbon dioxide and steam, respectively. The JHC (jet in hot co-flow) burner developed in Ref.[20] is adopted in the present study as the research prototype. Besides the JHC burner proposed by Dally's group[20], there is another popularly used JHC burner developed by the researchers in Delft[21,22]. Within a JHC burner the influence of surrounding atmosphere on fine reaction structures can be prevented, so it is an ideal benchmark for a comparison study on MILD oxyfuel combustion in various dilution gases. The investigation is based on numerical simulation, so firstly the adopted numerical approach is validated by the experimental data [20]. In the present work, besides the influences of

various dilution atmospheres, the effects of temperature of co-flow on MILD oxyfuel combustion are also investigated as until now no open effort reported on this important issue. Through the present study, a number of findings are reported for the first time and some conclusions presented in previous publications are checked with analyses on the differences, especially on some conflicted claims. In addition, several new questions are raised, which may inspire future research activities.

2 Computational Details

2.1 Configuration of the JHC burner and numerical conditions

The configuration of the JHC burner is illustrated by Fig.1 and the detailed description on it please refer to Ref.[20]. As the JHC burner is axisymmetric, in order to reduce numerical simulation cost, the investigated domain can be simplified as a two-dimensional case, as shown by Fig. 2. In the JHC burner, fuel is injected through the central jet pipe whose inner diameter reads 4.25 mm. The fuel jet pipe is surrounded by an annulus oxidant co-flow pipe with an inner diameter 77.75 mm. The whole JHC burner is operated inside a wind tunnel filled by environmental gas. The velocity boundary condition is adopted for all jet flows and at the downstream exit the pressure outlet boundary condition is assumed. In addition, a zero-shear stress wall boundary condition is employed as the tunnel flow is much wider than the jet flows. Because the

JHC burner is originally designed for air MILD combustion research rather than MILD oxyfuel combustion, in the present simulation, we replace air in tunnel flow by steam or carbon dioxide, respectively. Furthermore, the temperature of tunnel flow is set as 400 K to guarantee H_2O at its steam status in tunnel flow. Finally, to reduce the complication induced by variation of fuel mixture, in the present work it is assumed that the fuel jet flow consists of pure methane, instead of the mixture of methane and hydrogen used in Ref.[20]. Table 1 lists the detailed information of investigated cases covered by the present simulation. In Table 1, **u** and T represent jet flow velocity and temperature, respectively. The mass fraction of reactants is also listed in the Table. As the present study aims at the effects of temperature and oxygen concentration of co-flow on methane MILD oxyfuel combustion in different dilution atmosphere, these two parameters vary over a wider range (the temperature of the co-flow $1500 \leq T_{cof} \leq 2100$ and the oxygen mass fraction in the co-flow $6\% \leq f_{o2} \leq 18\%$). Through a numerical test, it is found that reactants can not be ignited successfully if oxygen mass fraction in the co-flow is lower than 6% or the temperature of the co-flow is less than 1500 K. It is an obvious difference from the air MILD combustion [20] and it may result from two aspects: (1) there is no hydrogen addition in the present fuel flow while hydrogen is more active than methane to establish and to sustain combustion; (2) the specific heat capacity of H_2O, as well as that of CO_2, is larger than air.

2.2 Numerical methods and validation

The present numerical simulation is conducted with the aid of the commercial CFD software FLUENT (version 6.3) to solve the Reynolds Averaged Navier-Stokes (RANS) equations for turbulence [23]. For heat radiation calculation, the discrete ordinate (DO) model is used [23]. In addition, a modified weighted sum of gray gas (WSGG) model is adopted to calculate the gas mixture total emissivity [19]. Finally, the eddy dissipation concept (EDC) model [23] with detailed chemical kinetic mechanisms (GRI-Mech 3.0, excluding the reactions relevant to Nitrogen) [24] is employed for turbulence-reaction interaction and reaction kinetics. In the present numerical research, 46560 cells are employed, as illustrated by Fig.2, which is the same as that used in Ref.[19]. As demonstrated by our recent work [19], such grid resolution is fine enough to obtain grid-independent numerical prediction. The detailed information about grid discretization and numerical convergence please refer to our previous work [19].

Because there is no open experimental data on the JHC oxyfuel MILD combustion cases investigated in the present work, we validate the reliability and accuracy of the present numerical approach by the JHC air MILD combustion experiments conducted in Ref.[20]. Fig. 3 illustrates the comparison of temperature and species, along the radial direction, between the present numerical prediction and the experimental measurements of JHC air MILD combustion

with 3%, 6%, and 9% oxygen mass fraction in the co-flow at the axial location $x = 30$ mm [20]. For the measured CO hump in the co-flow stream, it was explained to be the result of cooling and extinction of the secondary flame near the burner outer wall [19,20]. The present prediction agrees well with the experimental data for these three JHC MILD flames, which demonstrates the present numerical approach is adequate for modeling JHC combustion.

3 Results and Discussion

As shown in previous research[2,6,19], for JHC combustion, the temperature of the co-flow (T_{cof}) and the oxygen mass fraction in the co-flow (f_{o2}) are the key parameters that affect fine reaction structures. Therefore, in the present work we compare the MILD oxyfuel combustion characteristics in different dilution atmosphere by adjusting these two parameters, respectively. Firstly we try to reveal the MILD oxyfuel combustion characteristics in different dilution atmosphere with a changeable f_{o2} and a fixed T_{cof}. In succession, T_{cof} varies with a constant f_{o2}.

3.1 Comparison against various oxygen concentration f_{o2} in co-flow

In order to compare the effects of oxygen mass fraction in the co-flow (f_{o2}) on combustion behavior in different dilution conditions, the cases at $T_{cof} = 1800$ K and $6\% \leq f_{o2} \leq 18\%$ are chosen as the representatives in this section.

Figure 4 illustrates the temperature distribution in O_2/H_2O or O_2/CO_2 atmosphere, respectively. According to this figure, it can be observed that there are two common features between H_2O- and CO_2-dilution condition: (1) the maximum temperature of the reactants will increase when more oxygen is added into the co-flow; (2) the zone with high temperature will expand towards the exit as f_{o2} increases. These phenomena are expected as combustion will be enhanced with more oxygen. Meanwhile, the differences between them are also obvious: (1) their maximum temperatures are not identical; and (2) their temperature profiles are quite different. The details are discussed below.

Figure 5 plots the maximum temperature (T_{max}) and temperature rise (ΔT) of the reactants in O_2/H_2O or O_2/CO_2 condition, respectively. In both dilution atmosphere, T_{max}, as well as ΔT, is almost a linear increasing function of f_{o2}. As shown by Fig. 5(b), the temperature rise of the reactants is lower than the ignition temperature of methane and the temperature of co-flow is above the ignition temperature of methane, so the reactants react in the MILD oxyfuel regime [1,25]. The peak temperature in O_2/CO_2 condition is always higher than its steam counterpart. In addition, the increasing rate of T_{max} in O_2/CO_2 condition is faster, so the gap between T_{max} in different dilution atmosphere becomes wider with f_{o2} growing up. It mainly results from that the mass specific heat capacity of H_2O is larger than CO_2. Furthermore, the dilution gas may alter reaction paths by different ways, especially in relation to dissociation reactions, which also will influence heat release in combustion,

as discussed in our previous studies [1,12,19]. As shown in our latest work[1], the exothermic reaction chain of methane will be suppressed in oxy-steam atmosphere, depending on local temperature.

It has been reported that compared with its air-firing counterpart, in MILD oxyfuel combustion oxidization of fuels will take place within a larger area [2,6]. As shown in Refs. [2,6], in O_2/CO_2 condition, the zone with intensive heat release will expand toward the exit of JHC. In our simulation, this phenomenon is observed, too. According to Fig. 4, it can be observed that the zone with high temperature will expand towards the exit in both O_2/CO_2 and O_2/H_2O condition. Especially, we find that in O_2/CO_2 atmosphere the zone will expand more quickly. However, in its steam dilution counterpart, the zone with high temperature will expand obviously not only axially but also radially. In other words, in oxy-steam condition, most heat is released in the area closer to the fuel jet nozzle. It is another discovery reported by the present work for the first time. In all available open literature on MILD oxyfuel combustion [1,2,4–8,16,17], few pay attention to compare reaction structure alteration between in CO_2- and in H_2O-dilution condition until the present work. This new finding is very crucial for burner and chamber design as they both depend closely on temperature distribution. It is clearer with the aid of the distribution of hydroxyl radial (OH), as depicted by Fig. 6. Usually in the MILD combustion research community OH is used as a kind of marker for "flame" front region as MILD combustion is flameless [26]. As illustrated by

Fig. 6, in both dilution atmosphere the OH contours will expand with more oxygen being added, but their shape are completely different. The shape of OH contours in O_2/CO_2 condition looks like dragonflies' wings (namely long and slender), which is similar with its air MILD combustion counterpart [27]. However, the shape of OH contours in O_2/H_2O atmosphere looks like butterflies' wings (namely relative wider but shorter), quite different from its air MILD combustion [27] and CO_2-dilution counterpart [6]. In the "feedback" oxy-steam combustion [14–16], whether there exists a similar feature is still an open question and we will try to answer it in our future work. As in the oxy-steam condition the "flame" front is closer to the fuel jet nozzle, it is easier to establish the MILD oxyfuel regime in H_2O-dilution atmosphere. This conclusion agrees with that drawn from its counterflow counterpart [1]. Figure 7 plots the maximum of OH concentration at various f_{o2}. In both dilution atmosphere, the maximum of OH concentration will ascend nearly linearly with f_{o2}. It is in the expectation that the maximum of OH in O_2/H_2O condition is larger than its O_2/CO_2 counterpart since the production of OH will be enhanced by H_2O addition, as explained in detail in our previous work [12]. In Ref. [12], a counter-flow combustion prototype was adopted and the research objective is "feed-back" oxyfuel combustion. The present work demonstrates that the conclusion for "feed-back" oxyfuel combustion can be extended to MILD oxyfuel combustion. With relative lower OH concentration, as well as dragonfly-wing-like OH distribution, in O_2/CO_2 condition it is easier to sustain the MILD oxyfuel combustion regime across the whole domain, which is

consistent with the conclusion claimed in Ref.[1].

Figure 8 illustrates CO distribution at various f_{o2}. In Ref.[6], Mei et al. discussed the dimension of CH_4 JHC flame in O_2/CO_2 condition and they suggested to use the contour of CO mass fraction $f_{co} = 0.01$ as an indicator to visualize "flame" size. In this work we follow their suggestion. Through Fig. 6, one can observe the "flame" size in oxy-steam condition will grow dramatically with more oxygen addition. However, in its O_2/CO_2 counterpart, the size of "flame" will nearly not change with f_{o2}. In Ref.[6], Mei et al. claimed that "flame" size would decreased against f_{o2} in O_2/CO_2 co-flow. Our observation is different from their claim but similar with that reported in Ref.[5] in which oxyfuel combustion in the IFRF semi-industrial scale furnace was investigated. The difference between the present work and Ref.[6] results from that Mei et al. adopted a modified JHC configuration in their research. In Ref.[6], the cold tunnel flow in original JHC burner scheme was removed and replaced by hot co-flow. Consequently, the diameter of the hot co-flow jet in Ref.[6] is so wide that there is sufficient oxygen for combustion anywhere in the whole investigated domain. Accordingly, the consumption speed of CH_4 is mainly determined by reaction rate. It can be looked as a kinetic-controlled combustion. However, in the present work, the consumption speed of CH_4 depends not only on reaction rate but also on local available oxygen concentration since in the present JHC configuration there is no oxygen in the tunnel flow. In other words, the combustion in the present work is diffusion-kinetic-controlled

where chemical kinetics and aerodynamics (turbulent mixing) compete with each other. No doubt, the combustion style investigated in the present work is much closer to real combustion situation than the modified JHC scheme in Ref.[6]. It also can explain why the result obtain in the present study is consistent with that from the semi-industrial scale furnace [5]. Nowadays there appear a number of studies (please see [19] and references therein) in which a modified JHC burner like that used in Ref.[6] was adopted as a co-flow research prototype. Through the present work, it is indicated that we should check carefully before extending the conclusions claimed in these studies to realistic co-flow combustion systems. To mimic a realistic MILD oxyfuel combustion system, the present settings may be better. In addition, through Fig. 8 it can be observed that the "flame" size in oxy-steam condition is generally smaller and closer to the fuel jet nozzle, in comparison with its O_2/CO_2 counterpart. This observation is consistent with the above conclusion made from temperature and OH distribution. In Ref. [6], it was observed that with more oxygen addition in the O_2/CO_2 co-flow, the peak value of CO concentration within the reaction zone would increase, similar with its air-firing MILD combustion counterpart [20]. A comprehensive explanation on this phenomenon has been presented in Ref.[12]. Through the present work, we find such conclusion is also true in O_2/H_2O condition. In addition, as Ref.[12] focuses on "feed-back" oxyfuel combustion, through the present work it can be proved the above phenomenon is a common feature in oxyfuel combustion, regardless of dilution gases. In Ref.[2], it was reported that CO concentration would decrease slight-

ly against f_{o2}. At first glance it seems that this conclusion is opposed to that in Refs. [6,20] and the present study. In fact, the conclusion in Ref.[2] was tenable within the mix layer (namely the network reactor illustrated by Fig. 11 in Ref.[2]) rather than the whole domain of JHC [6,20]. As shown by Fig. 9, the CO maximum in both dilution atmosphere will grow almost linearly with f_{o2}. Whatever f_{o2} is, the CO maximum in O_2/CO_2 is always bigger than that in its oxy-steam counterpart. Especially, their gap will be enlarged with f_{o2} increasing. In our previous work [12], it was observed a similar phenomenon in counter-flow "feed-back" oxyfuel combustion. Consequently, it is also a common feature of oxyfuel combustion. The detailed explanation on how H_2O addition will suppress CO generation please refer to Ref.[12]. Figures 10-11 plot the CO profiles at different axial positions $x = 90$ and $x = 120$ mm. The profiles in both O_2/CO_2 and O_2/H_2O condition are similar with their air-firing counterpart: the gradient of CO concentration along radial direction becomes sharp with more oxygen addition [6,20]. Furthermore, it can be observed in O_2/H_2O condition the gradient of CO concentration along radial direction is more gentle than its O_2/CO_2 counterpart. This observation implies that in oxy-steam co-flow condition the MILD combustion regime can be established more easily, agreeing with the conclusion from the counter-flow configuration [1]. Moreover, in our previous study [28], it was found that the co-flow methane MILD combustion would be influenced significantly by the shape of furnace chamber. According to Figs.8, 10 and 11, one may conclude MILD combustion in oxy-steam condition is more flexible as the size of reaction zone in O_2/H_2O

condition is smaller (especially at low oxygen concentration) and within the near-field of the fuel jet nozzle. Accordingly it will receive less effect than its O_2/CO_2 counterpart.

Figure 12 depicts the distribution of O_2 with various f_{o2}. One can observe that in the vicinity of the jet, the profiles of O_2 in O_2/H_2O and O_2/CO_2 condition are very similar, however their discrepancies become obvious in the far-field. It is more clear with the aid of Fig.13, where the profiles of O_2 at $x = 30$ mm (near-field) and $x = 90$ mm (far-field) are illustrated. In the near-field, the profiles of O_2 in both dilution atmosphere nearly overlap with each other. It agrees with the observation in Ref.[2] where only O_2/CO_2 condition was considered. In the far-field, O_2 concentration in O_2/H_2O condition is always lower than its O_2/CO_2 counterpart, which is consistent with Fig.10 and indicates oxidants are consumed faster in oxy-steam condition.

In Ref.[2], it was reported that in the near field ($x = 30$ mm), the differences between the profiles of CO, O_2 and OH in O_2/N_2 atmosphere and those in O_2/CO_2 condition are very small. However, through the present work, it is observed that except O_2, there are obvious differences in most scalar distributions between in O_2/H_2O and in O_2/CO_2 condition, even in the near field. Consequently, one should pay great attention on burner design for oxy-steam combustion due to its complicated reaction structures. This conclusion is consistent with that in Ref.[1].

The distribution of formyl (HCO) is shown by Fig.14. In the near-field of the fuel jet nozzle, HCO concentration will increase with more oxygen addition in both dilution atmosphere. In Ref.[6] it was reported in O_2/CO_2 condition the peak value of HCO would grow up with f_{o2}. The present results are consistent with it and prove this conclusion also can hold water in its oxy-steam counterpart. As HCO is an indicator for heat release during combustion [2,26], it can be concluded that heat release will be enhanced by increasing f_{o2}. Moreover, HCO concentration in O_2/CO_2 condition is always higher than its O_2/H_2O counterpart, as illustrated by Figs.14 and 16 (a), so in O_2/CO_2 condition heat release intensity is higher than in oxy-steam atmosphere, which is consistent with Fig. 4. It can answer why MILD oxyfuel combustion is easier to be sustained in O_2/CO_2 condition. In our previous work [12], it was also observed that HCO concentration in O_2/H_2O condition was lower than its O_2/CO_2 counterpart, which resulted from that H_2O addition would modify the chemical equilibrium of the reaction step R46. Through the present work, it can be proved the analysis in Ref.[12] where counter-flow prototype adopted still works well for JHC configuration. And it is a common feature between "feed-back" oxyfuel combustion and MILD oxyfuel combustion.

Figure 15 plots the distribution of formaldehyde (CH_2O) which can serve as an indicator for ignition [2,26]. Since CH_2O predominantly exists in low temperature condition, therefore the concentration of CH_2O will decrease against f_{o2} increasing [6,26]. The present results agree with the conclusion in [6,26].

As shown by Fig.4, a higher f_{o2} implies a higher combustion temperature. Because H_2O addition will suppress CH_2O generation [12], in oxy-steam atmosphere CH_2O concentration in the near-field is a slightly lower than its O_2/CO_2 counterpart. In O_2/CO_2 condition, the profiles of CH_2O alter sensitively to the variation of f_{o2} , while in O_2/H_2O atmosphere the change is relatively smaller. It implies the establishment of MILD combustion in O_2/H_2O condition receives less influence by oxygen fluctuation. Consequently, it is easier to establish MILD combustion regime in oxy-steam condition. It is consistent with the above analysis and the conclusion in Ref.[1]. Fig. 16 (b) depicts the maximums of CH_2O in both dilution atmosphere. The peak value of CH_2O in CO_2-dilution atmosphere is always larger than its H_2O-dilution counterpart, which is consistent with its counter-flow "feed-back" counterpart [12]. Together with Fig.5, Fig. 16 (b) indicates that over the whole domain the uniformity of ignition in oxy-steam is better than its CO_2-dilution counterpart.

3.2 Comparison against various temperature T_{cof} of co-flow

In order to compare the effects of the temperature of the hot co-flow (T_{cof}) on combustion behavior in O_2/H_2O and O_2/CO_2 condition, the cases at $f_{o2} = 9\%$ and $1500K \leq T_{cof} \leq 2100K$ are chosen as the representatives in this section.

Figure 17 illustrates the temperature distribution in O_2/H_2O and O_2/CO_2 condition at various T_{cof}. In CO_2-dilution condition, the maximum temper-

ature of the reactants will climb up with a higher T_{cof} and the zone with high temperature will expand towards the exit as T_{cof} increases. The former phenomenon has been reported in Ref.[6] and the latter one was also observed in Ref.[2]. Refs.[2,6] just focused on O_2/CO_2 atmosphere. Through the present study, we can confirm these phenomena exist in oxy-steam condition, too. However, the influences of variation of T_{cof} on the temperature field in O_2/H_2O and O_2/CO_2 condition are quite different. The isotherms in these two types of dilution atmosphere differ with each other obviously, especially in the vicinity of the fuel jet nozzle. In addition, the high temperature zone expands more quickly in CO_2-dilution atmosphere. The maximum temperature of the reactants is illustrated by Fig.18. T_{max} is a monotonic increasing function of T_{cof} in both dilution conditions and since $T_{cof} \geq 1600$ K T_{max} grows up almost linearly. T_{max} in O_2/H_2O atmosphere is always smaller than its CO_2-dilution counterpart. As mentioned above, it results from that the mass specific heat capacity of H_2O is larger than CO_2. However, their gap will decrease against T_{cof} increasing, which implies a higher T_{cof} will improve the uniformity of temperature field of MILD oxyfuel combustion in either dilution atmosphere. This observation agrees with that presented in Ref.[6]. Moreover, making a comparison between Figs. 4-5 and Figs.17-18, one may conclude the influence of variation of f_{o2} on the temperature field is more significant than T_{cof}.

The distribution of OH with various T_{cof} is depicted by Fig. 19. It can be observed that the "flame" front region in oxy-steam atmosphere is more sen-

sitive to T_{cof}, in comparison with its O_2/CO_2 counterpart. With a relative low co-flow temperature, such as T_{cof} = 1500 K, the "flame" front region in oxy-steam atmosphere is much smaller than its O_2/CO_2 counterpart. The shape of OH contours in H_2O-dilution condition looks like a dragonfly's wing, similar with its CO_2-dilution counterpart although the former is shorter. Since $T_{cof} \geq$ 1600 K, the "flame" front region in oxy-steam condition expands substantially along the radial direction and now the shape of OH contours in H_2O-dilution condition looks like a butterfly's wing, not resembling that in O_2/CO_2 atmosphere any longer. And now the "flame" front region in the former is much larger than the latter. The sensitivity of OH generation to T_{cof} in oxy-steam condition is also reflected by Fig.20. The maximum of OH concentration in O_2/H_2O condition ascends much faster than in O_2/CO_2 atmosphere. In our previous study on MILD oxyfuel counterflow combustion [1], it was found that the reaction structures in steam-dilution condition would be more complex than in O_2/CO_2 atmosphere. The present work demonstrates such conclusion can apply to the co-flow scenario. Fig.20 illustrates the variation of maximum of OH concentration at various T_{cof}. The maximum of OH concentration in either dilution atmosphere will grow up with T_{cof}, which is consistent with the result reported in Ref.[6]. As mentioned above, as the peak temperature in O_2/H_2O condition is lower than its O_2/CO_2 counterpart, the maximum of OH concentration in the former is always higher than the latter. The increasing rate of the maximum of OH concentration in oxy-steam atmosphere is much faster than its CO_2-dilution counterpart, which also implies

the fine reaction structures in O_2/H_2O condition are more sensitive to T_{cof}, in comparison with its O_2/CO_2 counterpart. In our previous work [1], it was claimed that the MILD oxyfuel combustion regime was established more easily in oxy-steam condition. Through Figs. 19-20, we find this conclusion may depend on T_{cof} in the present co-flow configuration. Only since $T_{cof} > 1500$ K, in O_2/H_2O condition, the peak value of OH concentration is significantly larger than its O_2/CO_2 counterpart and the "flame" front region is substantially wider than that in CO_2-dilution atmosphere. Consequently, in the present investigated cases, only since $T_{cof} > 1500$ K, it is sure that the MILD oxy-fuel combustion regime can be established more easily in O_2/H_2O atmosphere.

Figure 21 plots the distribution of CO with various T_{cof}. The iso-concentration lines of CO are affected significantly by the variation of T_{cof}, especially in oxy-steam condition. If taking the contour of CO mass fraction $f_{co} = 0.01$ as an indicator to visualize the "flame" size, as mentioned above, one can observe that the "flame" size in O_2/H_2O atmosphere changes substantially with T_{cof}. When $T_{cof} = 1500$ K, the "flame" size in H_2O-dilution condtion is much smaller than its CO_2-dilution counterpart. Then the "flame" size in the former atmosphere grows quickly with T_{cof} increasing. While $T_{cof} = 2100$ K, the "flame" size in both dilution conditions is almost the same. On the contrary, although the CO iso-concentration lines in O_2/CO_2 condition will alter obviously with T_{cof}, the "flame" size in CO_2-dilution atmosphere grows slightly. The maximum of CO is depicted by Fig. 22. It is clear that the maximum

of CO will ascend with a higher T_{cof}, which is consistent with the conclusion given in Ref.[6]. In Ref.[6] only O_2/CO_2 atmosphere was investigated. The present work shows there is a similar trend in oxy-steam condition. However, the increasing rate of the peak value of CO in O_2/H_2O condition is much slower than its O_2/CO_2 counterpart. Taking Figs. 9 and 22 together, one can conclude that a crucial issue to guarantee the performance of MILD oxyfuel combustion in O_2/CO_2 atmosphere is to ensure fuel to burn out in a finite room as the maximum of CO concentration at the outlet of the investigated domain will jump up quickly, exceeding 10%, with the fluctuation of either T_{cof} or f_{o2}. Such high value of CO concentration at the outlet implies an extremely low combustion efficiency. On the contrary, in the oxy-steam condition, the maximum of CO concentration at the outlet of the investigated domain is always less than 4%, no matter whatever T_{cof} and f_{o2} are. From this viewpoint, burner and chamber design, which can improve aerodynamics in furnace and accordingly improve combustion efficiency, is more critical for operation in CO_2-dilution condition.

Figure 23 shows the CO radial profiles at $x = 90$ mm. In Ref.[29], it was reported that, in air MILD condition, the peak value of CO concentration along the radial direction would grow up with T_{cof} increasing. The present work proves such conclusion can be extended to MILD oxyfuel regime. In addition, a higher T_{cof} will sharpen the gradient of CO concentration in both dilution conditions. A similar observation was reported in Ref.[6] where only

O_2/CO_2 atmosphere was investigated. Through the present work, it is found the radial gradient of CO concentration in O_2/H_2O is always much smaller than its CO_2-dilution counterpart, no matter whatever T_{cof} is. The results reveal that the potential performance of MILD oxyfuel combustion in O_2/H_2O condition may be better than its O_2/CO_2 counterpart not only along the axial direction but also along the radial direction of a chamber as in oxy-steam condition most fuel can be burnt out in a relative small zone, in comparison with its CO_2-dilution counterpart.

The O_2 distribution with various T_{cof} is plotted by Fig. 24. Generally, the variation of T_{cof} will alter O_2 distribution significantly in both dilution atmosphere, especially in the far-field. Against T_{cof} increasing, O_2 concentration near the exit will decrease as a higher T_{cof} will intensify chemical reactions. Near the fuel jet nozzle, the influence of variation of T_{cof} on O_2 distribution is slight, as illustrated by Fig.25. In Ref.[29], it was also found that, in methane-air MILD combustion, O_2 distribution in the near-field is insensitive to T_{cof}. Through the present work, one may conclude that it is a common feature of methane MILD JHC combustion, regardless of dilution atmosphere. Taking Figs. 13 and 24 together, it can be observed that for any T_{cof} and f_{o2}, in the far-field the O_2 radial concentration in O_2/H_2O condition is always lower than its O_2/CO_2 counterpart. Such observation demonstrates once again that in oxy-steam condition the "flame" size is smaller than its CO_2-dilution counterpart.

Figure 26 illustrates HCO profiles in O_2/H_2O and O_2/CO_2 atmosphere, respectively. HCO concentration will increase with a higher T_{cof} in both dilution atmosphere as heat release will be enhanced by a hotter co-flow. A similar phenomenon was also observed in methane-air MILD combustion [29]. No matter whatever T_{cof} is, HCO concentration in O_2/CO_2 condition is always higher than its O_2/H_2O counterpart, as shown by Fig.27 (a). At a relative low co-flow temperature (T_{cof} = 1500 K), one can observer HCO concentration in oxy-steam atmosphere is rarefied. According to Fig. 27, it can be observed that in oxy-steam condition the variation of OH concentration versus T_{cof} is "smooth", which also can be reflected by Fig. 26 (a). However, it is not true for its O_2/CO_2 counterpart. As shown by Fig. 26 (b), when $T_{cof} < 1700$, the peak value of HCO at x = 30 mm will increase quickly with T_{cof}, but since $T_{cof} \geq 1700$, the change becomes slow. It agrees with the results depicted by Figs.19 and 21. Through these figures, one can observe that in O_2/CO_2 condition the shapes of OH, CO and HCO contours in the near-field change significantly when T_{cof} rises from below 1700 K to above 1700 K. It implies there appears a substantial change of the reaction structure in CO_2-dilution atmosphere. In other words, the MILD oxyfuel combustion performance in O_2/CO_2 condition is more sensitive to T_{cof}.

The radial distribution of CH_2O in the near-field is plotted by Fig.28. In Ref.[29], it was found in the near-field of air MILD combustion the maximum of CH_2O concentration along the radial direction would decrease against T_{cof}

growing up . The present results show this conclusion can be extended to CO_2-dilution MILD oxyfuel combustion. However, it is not true in MILD oxy-steam condition. In O_2/H_2O atmosphere, there will appear two obvious peak values of CH_2O concentration along the radial direction while in Ref.[29] only one was observed. In O_2/CO_2 condition, the second peak of CH_2O distribution along the radial direction is not as obvious as that in its H_2O-dilution counterpart. This phenomenon implies in oxy-steam atmosphere the ignition of reactants will take place over a wider range than in O_2/CO_2 or O_2/N_2 condition. Furthermore, it also indicates the effect of T_{cof} on ignition is more complicated in H_2O-dilution condition. In addition, in Ref.[29], it was observed that the "sharp angle" of the CH_2O profile at a low T_{cof} (e.g. $T_{cof} = 1500$ K) will be flattened by a high T_{cof} (e.g. $T_{cof} = 1800$ K). Although it was observed firstly in methane-air MILD combustion, the present work reveals that this conclusion is also tenable in the MILD oxyfuel combustion regime, either diluted by CO_2 or by H_2O. Such phenomenon indicates chemical reaction will become to vary mildly as T_{cof} increasing, which is consistent with the available research on high temperature air combustion [30].

4 Conclusion

In order to deepen our insight into MILD oxyfuel combustion, a recently emerging idea for next generation clean combustion technology, in the present

work we carry out a comprehensive comparison study on methane MILD oxy-fuel combustion in different dilution atmosphere (O_2/H_2O and O_2/CO_2). The JHC burner is adopted as a research prototype. The comparison is conducted by varying the mass fraction of oxygen in the co-flow (f_{o2}) and the temperature of the hot co-flow (T_{cof}), two key parameters affecting fine reaction structures in JHC. The literature survey demonstrates the present work is a pioneering effort in this field.

Through the present study, a number of findings are reported for the first time and it is found the combustion characteristics in various dilution atmosphere are obviously different:

(1) In oxy-steam condition, the CO contours are affected more significantly by the variation of T_{cof} and f_{o2}. But generally speaking, the "flame" size in CO_2-dilution atmosphere is much larger than its O_2/H_2O counterpart. The maximum concentration of CO in O_2/CO_2 atmosphere is about ten times large than its steam-dilution counterpart.

(2) In oxy-steam atmosphere the ignition of reactants will take place over a wider range than its O_2/CO_2 or O_2/N_2 counterpart. Especially, the effect of T_{cof} on ignition is more complicated in H_2O-dilution condition.

(3) In general, it is easier to establish the MILD oxyfuel regime in H_2O-dilution atmosphere and in O_2/CO_2 condition it is easier to sustain the MILD oxyfuel combustion regime across the whole domain. This conclusion agrees with that

drawn from its counter-flow counterpart investigated in our previous work [1].

(4) In Ref.[2], it was reported that in the near field of the JHC, the differences between the profiles of CO, O_2 and OH in O_2/N_2 and in O_2/CO_2 atmosphere are very small. However, through the present work, it is observed that except O_2, there are obvious differences in most scalar distributions between in O_2/H_2O and in O_2/CO_2 condition, even in the near field. Consequently, one should pay great attention on burner design for MILD oxy-steam combustion due to its complicated reaction structures.

Finally, several new questions are raised by the present study. For example, whether the shape of OH contours in "feed-back" oxyfuel combustion diluted by H_2O will change from the dragon-wing-style to butterfly-wing-style? It is an important question as "feed-back" oxyfuel combustion diluted by H_2O already appeared in industrial-scale furnaces [14–16] but until now nobody is aware of this issue. We will try to answer it in our future work since it will influence combustion performance, as shown by the present study.

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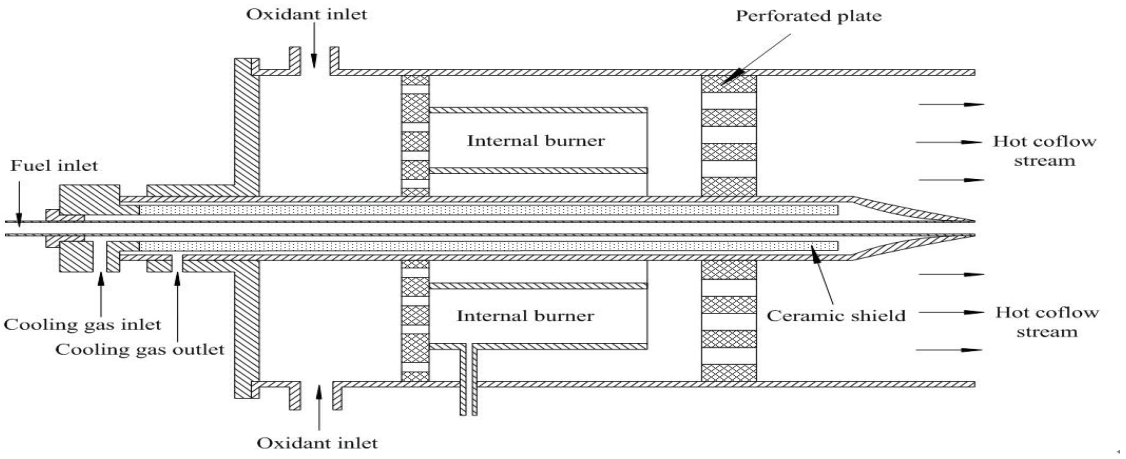


Fig. 1. Configuration of the JHC burner proposed in Ref. [20].

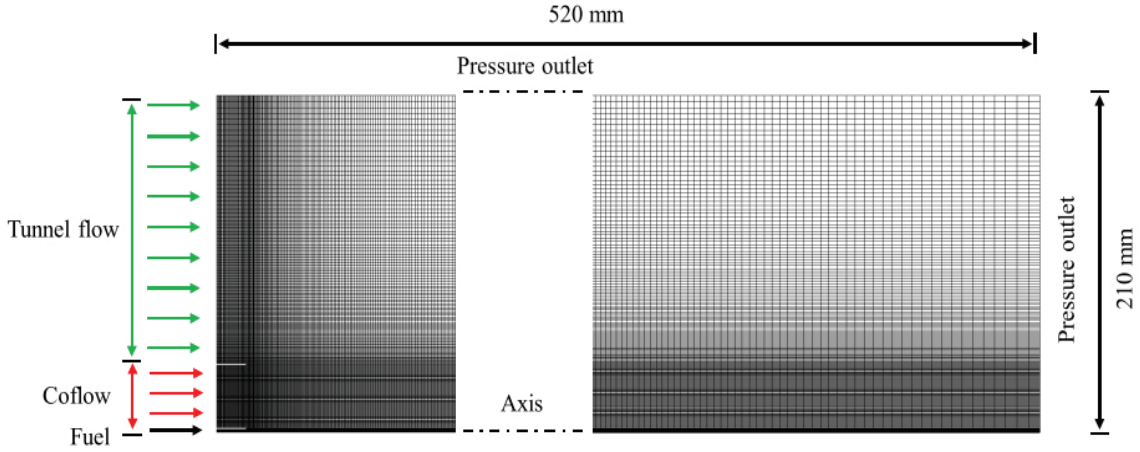


Fig. 2. Schematic configuration and coordinate system of the computational domain.

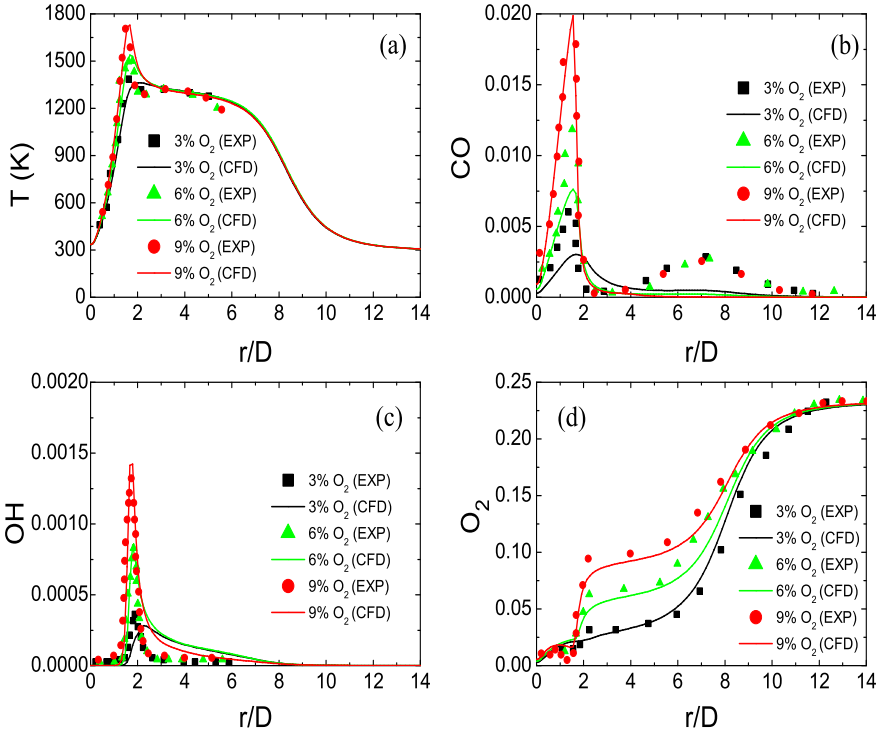


Fig. 3. comparison of (a) temperature, (b) CO mass fraction, (c) OH mass fraction, (d) O_2 mass fraction profiles between the present numerical prediction (CFD) and the experimental measurements (EXP) for JHC air MILD combustion with 3%, 6%, and 9% oxygen mass fraction in the co-flow at the axial location $x = 30$ mm [20].

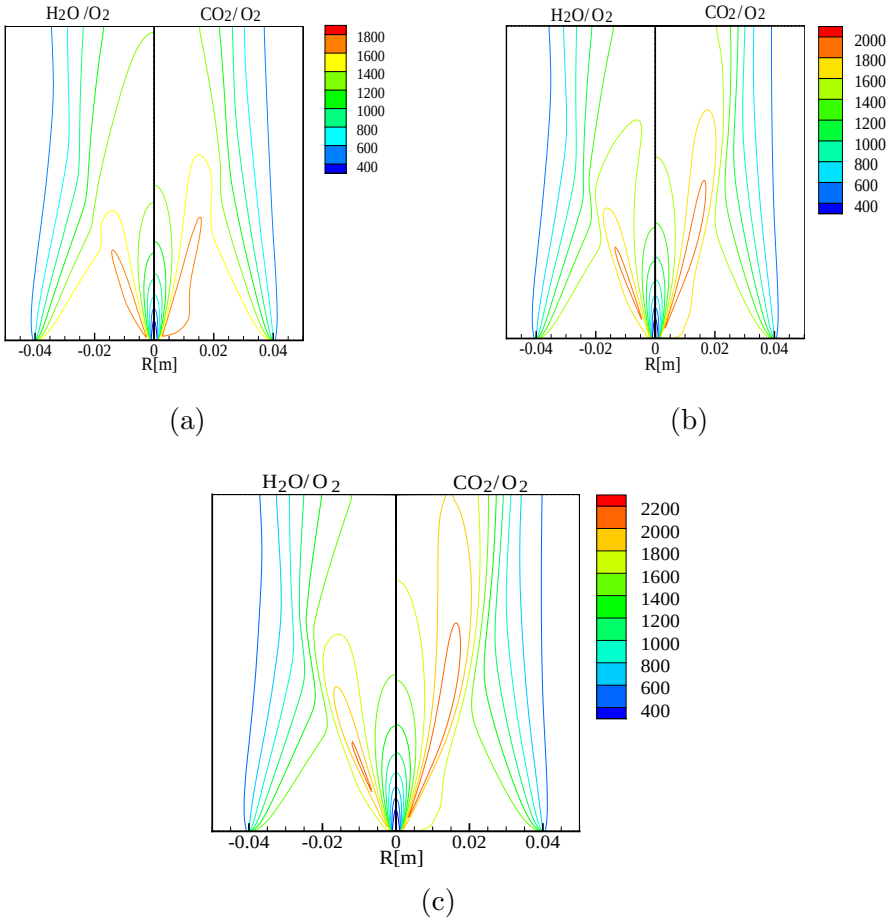
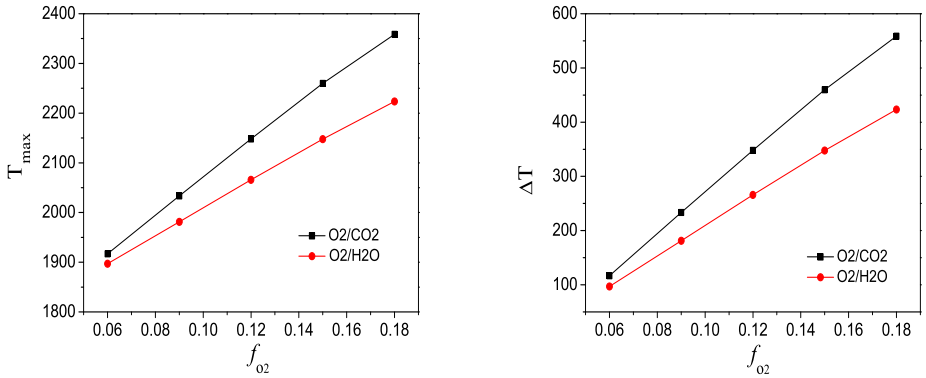
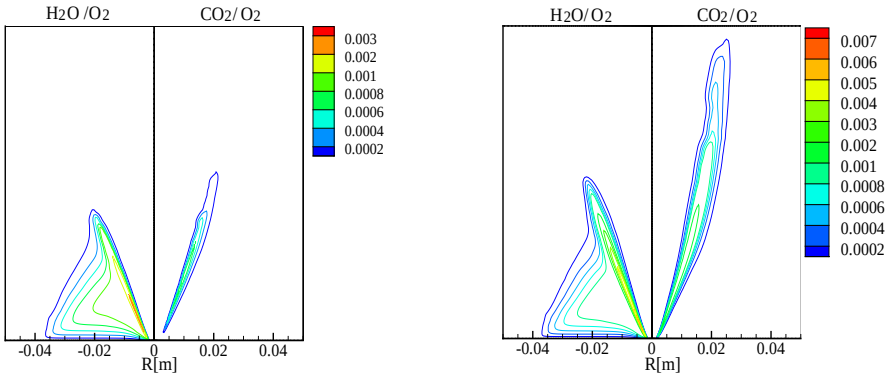


Fig. 4. Temperature distribution in O_2/H_2O or O_2/CO_2 condition at (a) $f_{o2} = 6\%$ (b) $f_{o2} = 12\%$ and (c) $f_{o2} = 18\%$: $T_{cof} = 1800$ K.



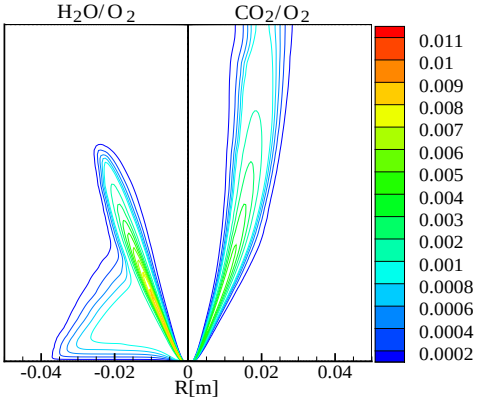
(a) (b)

Fig. 5. Maximum temperature (a) and temperature rise (b) at various f_{o2} and $T_{cof} = 1800$ K.



(a)

(b)



(c)

Fig. 6. OH distribution in O_2/H_2O or O_2/CO_2 condition at (a) $f_{o2} = 6\%$ (b) $f_{o2} = 12\%$ and (c) $f_{o2} = 18\%$ and $T_{cof} = 1800$ K.

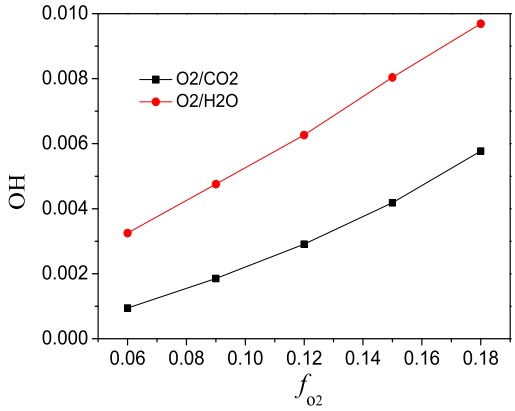


Fig. 7. The maximum of OH concentration at various f_{o2} and $T_{cof} = 1800$ K.

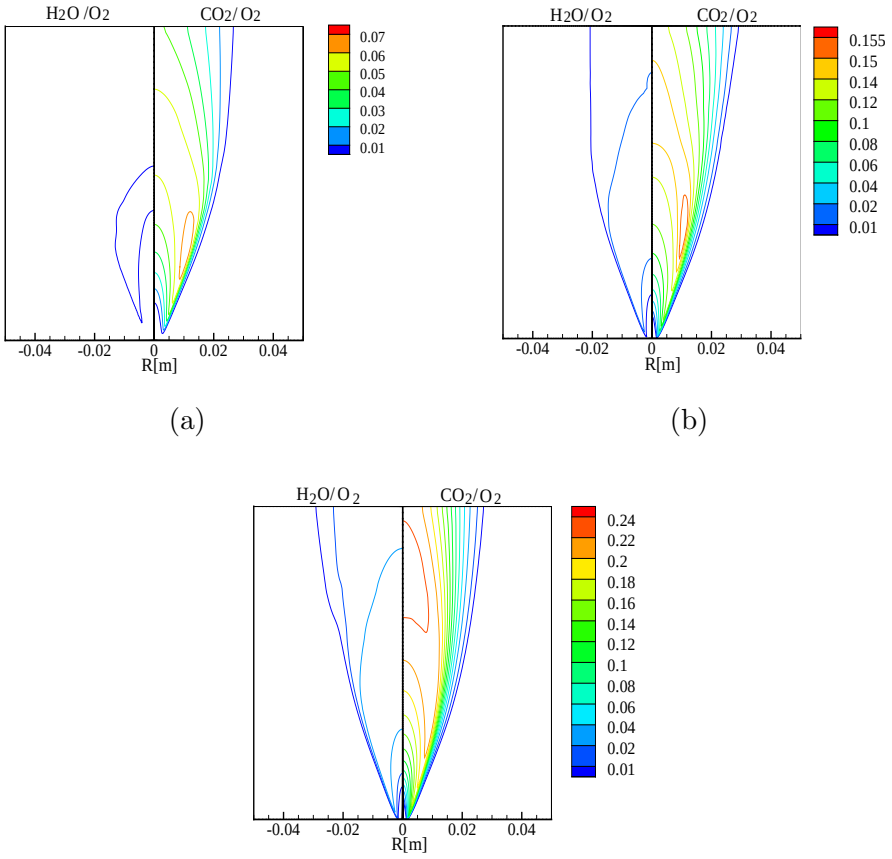


Fig. 8. CO distribution in O_2/H_2O or O_2/CO_2 condition at (a) $f_{o2} = 6\%$ (b) $f_{o2} = 12\%$ and (c) $f_{o2} = 18\%$ and $T_{cof} = 1800$ K.

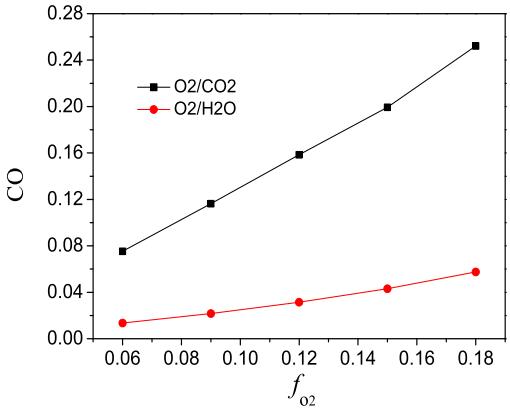


Fig. 9. The maximum of CO concentration at various f_{o2} and $T_{cof} = 1800$ K.

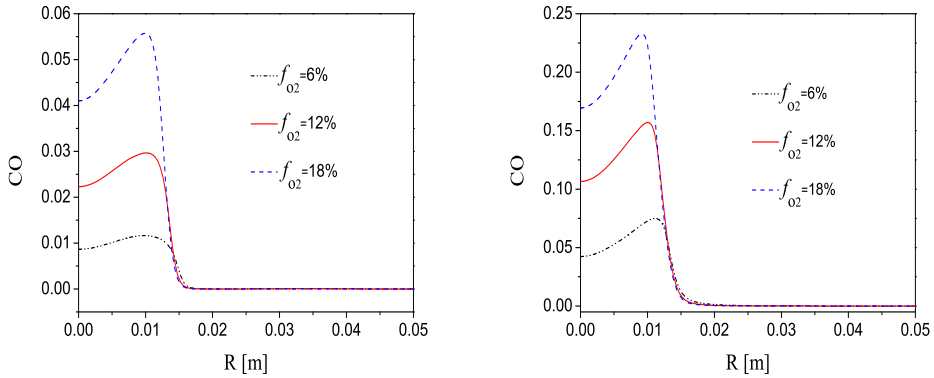


Fig. 10. CO profile in (a) O_2/H_2O and (b) O_2/CO_2 condition at $x = 90$ mm and $T_{cof} = 1800$ K.

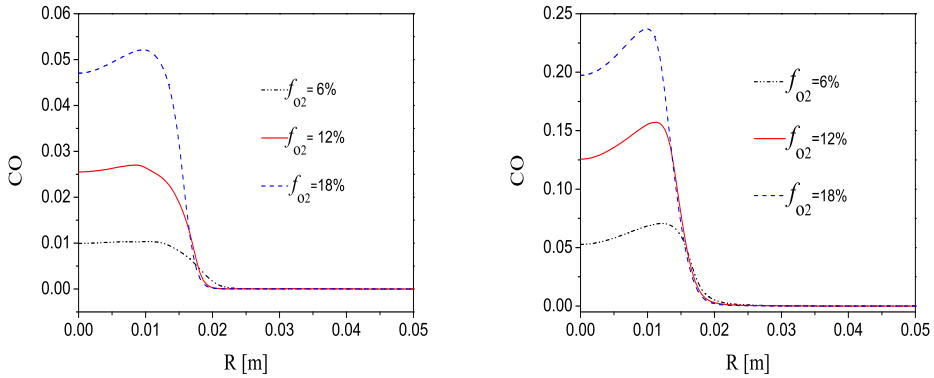


Fig. 11. CO profile in (a) O_2/H_2O and (b) O_2/CO_2 condition at $x = 120$ mm and $T_{cof} = 1800$ K.

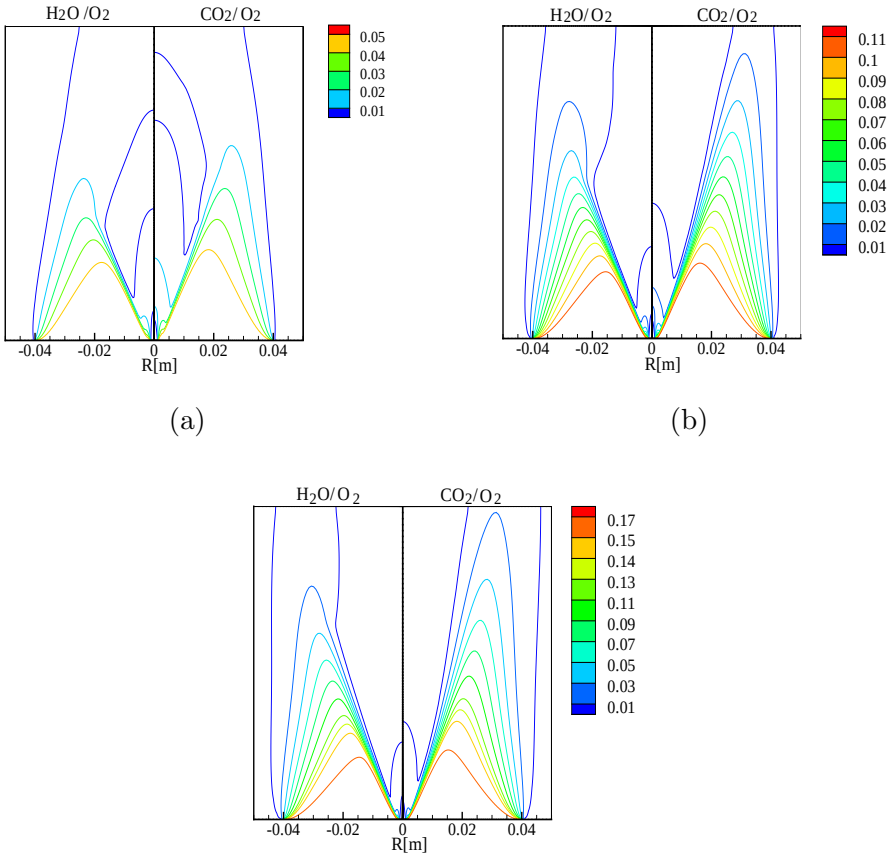
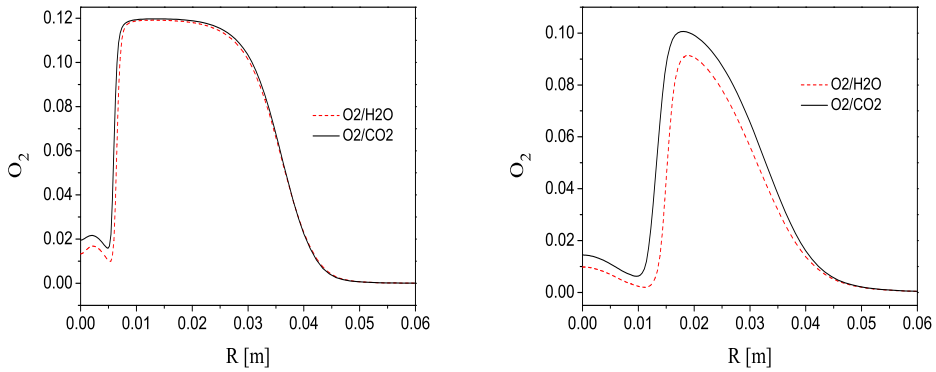


Fig. 12. O_2 distribution in O_2/H_2O or O_2/CO_2 condition at (a) $f_{o2} = 6\%$ (b) $f_{o2} = 12\%$ and (c) $f_{o2} = 18\%$ and $T_{cof} = 1800$ K.



(a) (b)

Fig. 13. O_2 profile at (a) $x = 30$ mm and (b) $x = 90$ mm : $f_{o2} = 12\%$ and $T_{cof} = 1800$ K.

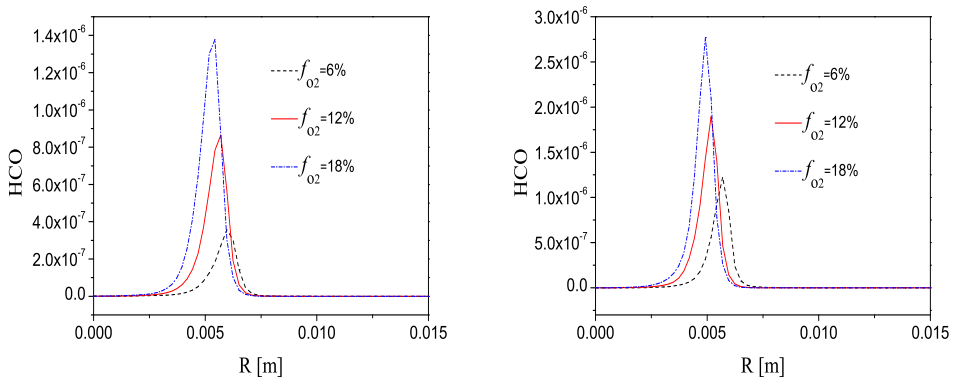
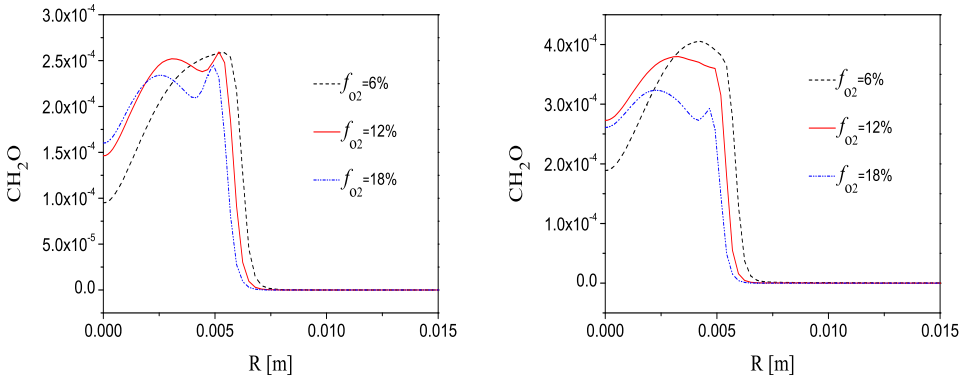


Fig. 14. HCO profile in (a) O_2/H_2O and (b) O_2/CO_2 condition at $x = 30$ mm and $T_{cof} = 1800$ K.



(a) (b)

Fig. 15. CH_2O profile in (a) O_2/H_2O and (b) O_2/CO_2 condition at $x = 30$ mm and $T_{cof} = 1800$ K.

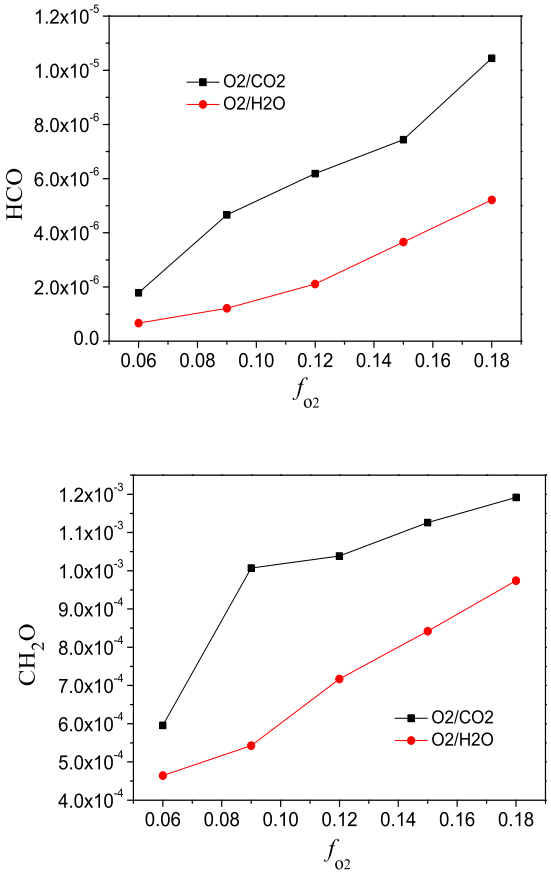


Fig. 16. The maximum of HCO (a) and CH_2O (b) concentration at various f_{o2} and $T_{cof} = 1800$ K.

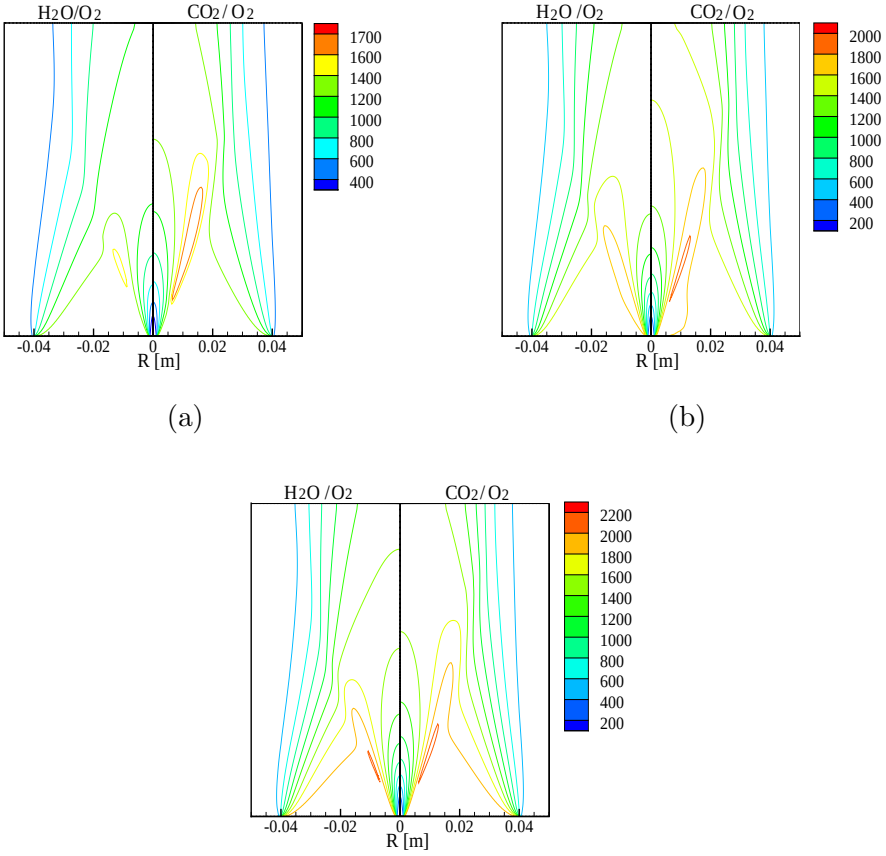


Fig. 17. Temperature distribution in O_2/H_2O or O_2/CO_2 condition at (a) $T_{cof} = 1500$ K (b) $T_{cof} = 1800$ K and (c) $T_{cof} = 2100$ K: $f_{o2} = 9\%$.

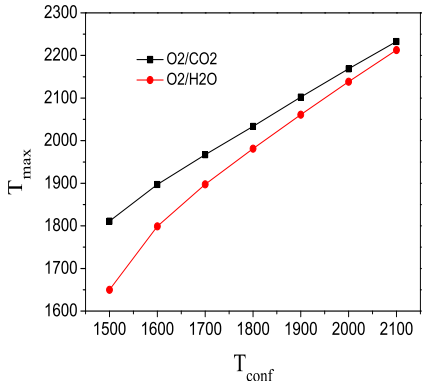


Fig. 18. Maximum temperature at various T_{cof} and $f_{o2} = 9\%$.

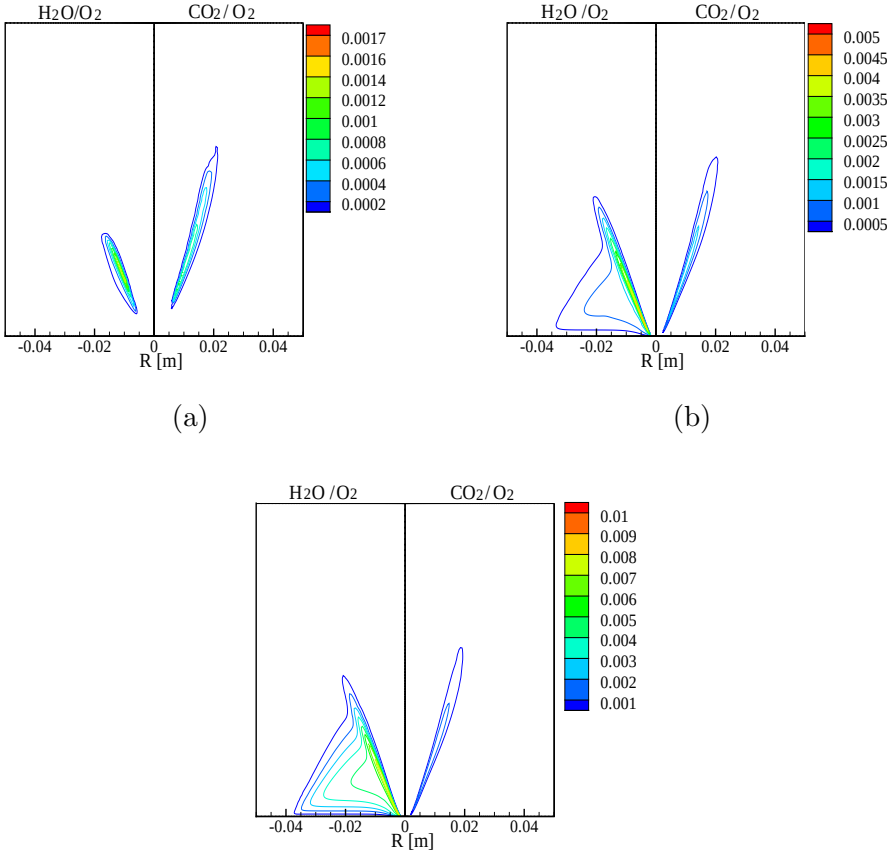


Fig. 19. OH distribution in O_2/H_2O or O_2/CO_2 condition at (a) $T_{cof} = 1500$ K (b) $T_{cof} = 1800$ K and (c) $T_{cof} = 2100$ K: $f_{o2} = 9\%$.

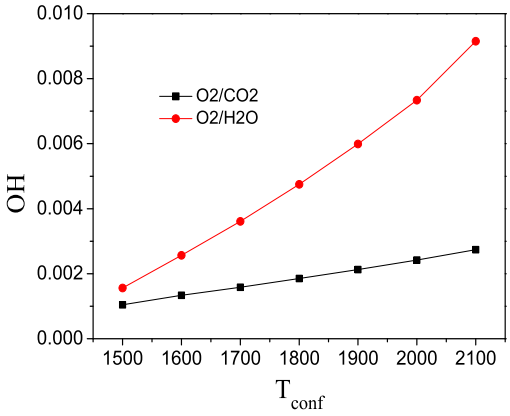


Fig. 20. The maximum of OH concentration at various T_{cof} and $f_{o2} = 9\%$.

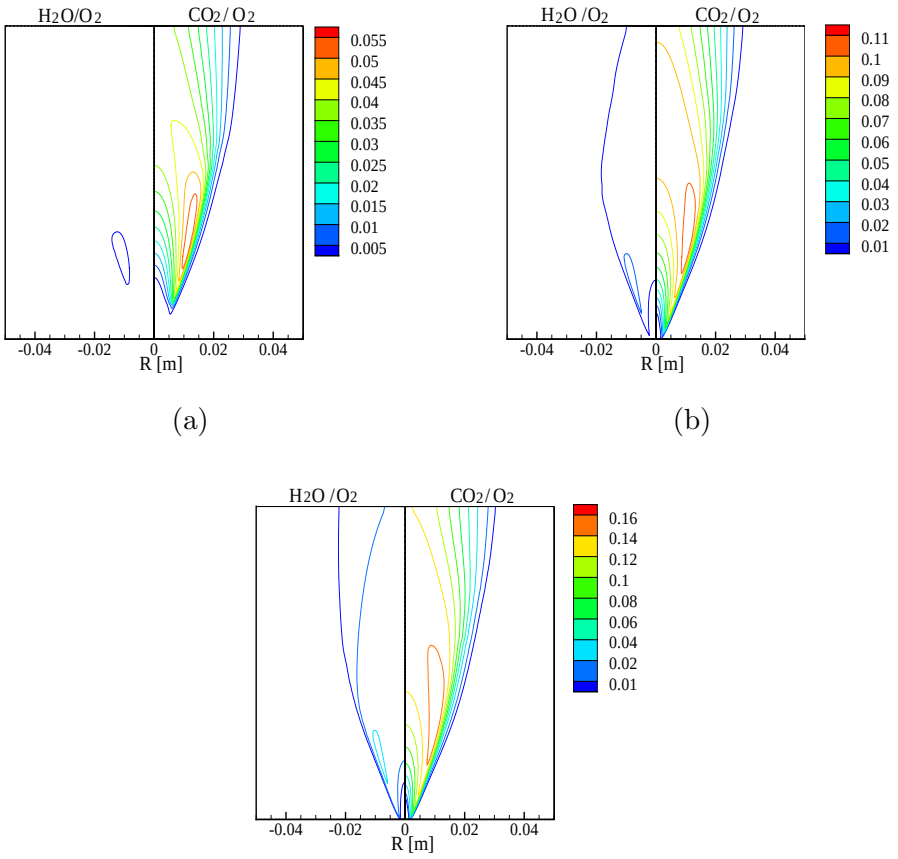


Fig. 21. CO distribution in O_2/H_2O or O_2/CO_2 condition at (a) $T_{cof} = 1500$ K (b) $T_{cof} = 1800$ K and (c) $T_{cof} = 2100$ K: $f_{o2} = 9\%$.

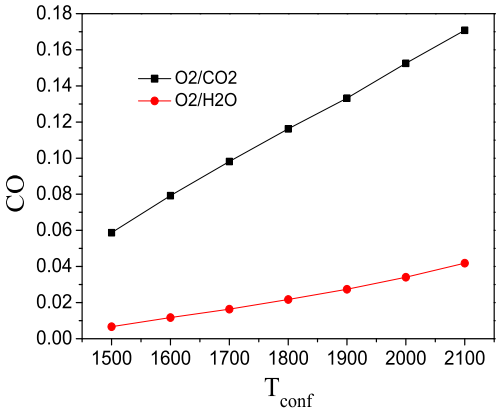


Fig. 22. The maximum of CO concentration at various T_{cof} and $f_{o2} = 9\%$.

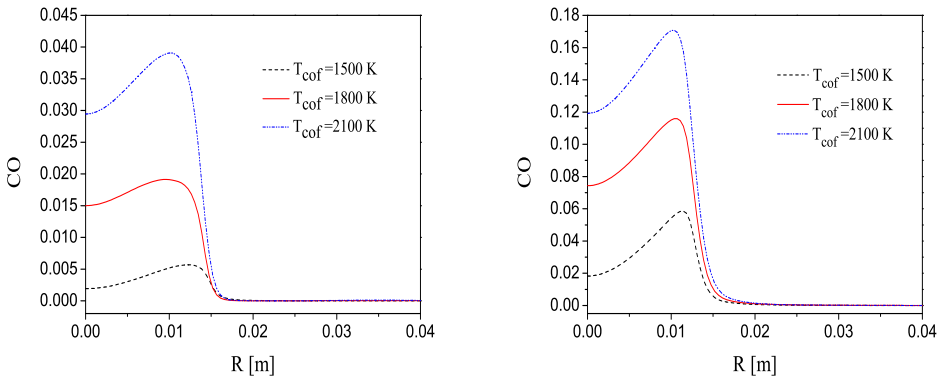


Fig. 23. CO profile in (a) O_2/H_2O and (b) O_2/CO_2 condition at $x = 90$ mm and $f_{o2} = 9\%$.

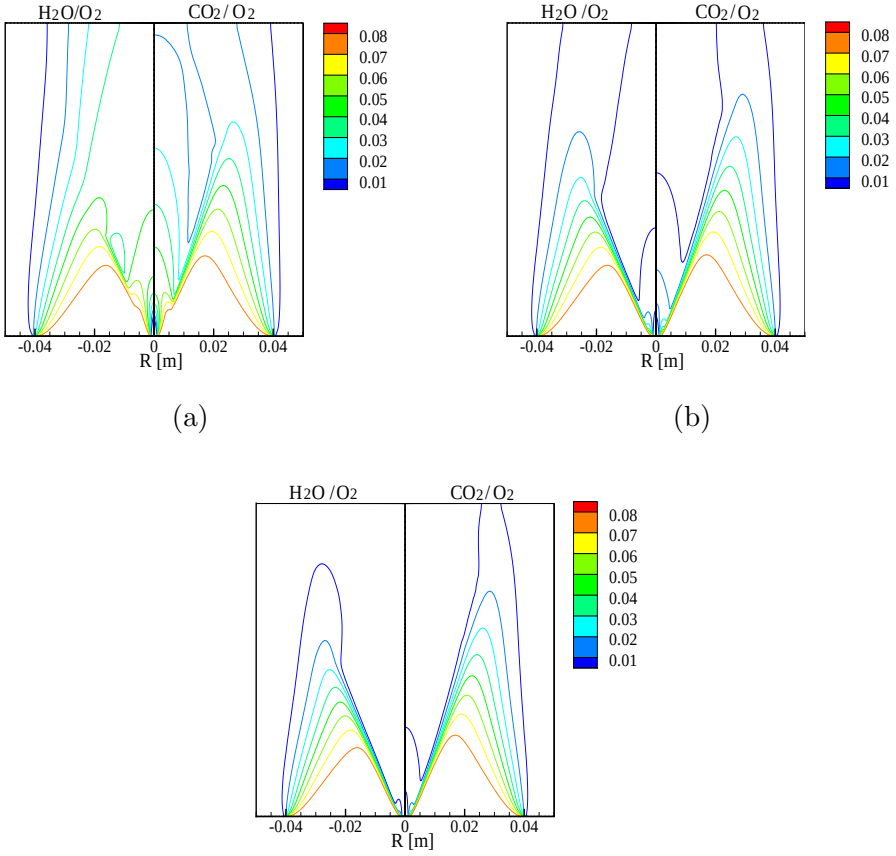
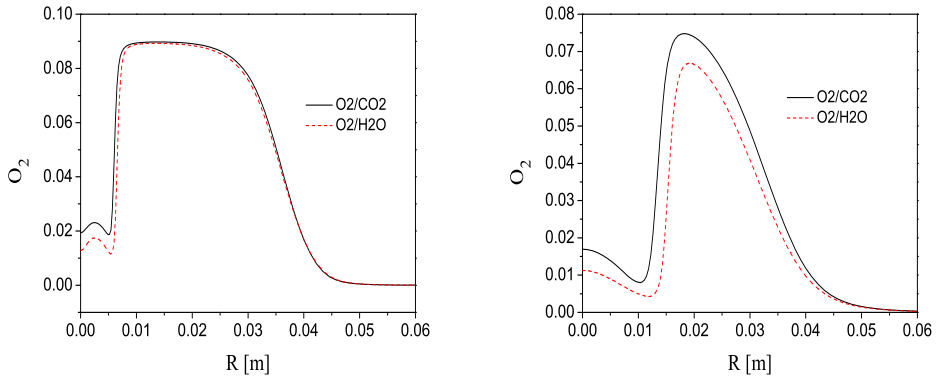
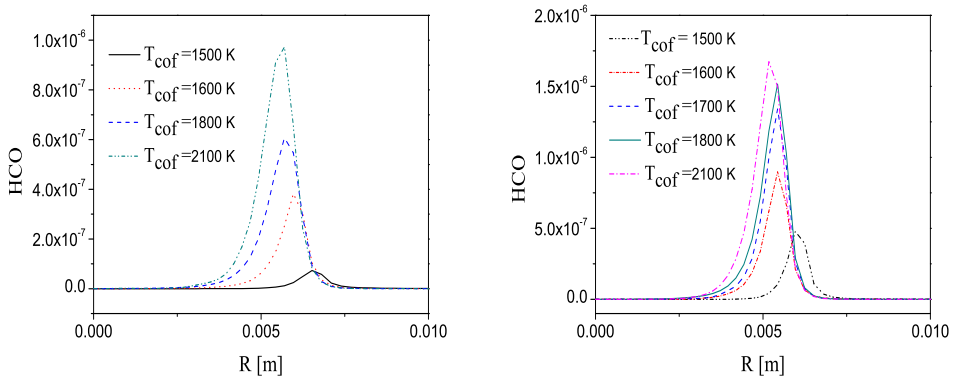


Fig. 24. O_2 distribution in O_2/H_2O or O_2/CO_2 condition at (a) $T_{cof} = 1500$ K (b) $T_{cof} = 1800$ K and (c) $T_{cof} = 2100$ K: $f_{o2} = 9\%$.



(a) (b)

Fig. 25. O_2 profile at (a) $x = 30$ mm and (b) $x = 90$ mm : $f_{o2} = 9\%$ and $T_{cof} = 2100$ K.



(a) (b)

Fig. 26. HCO profile in (a) O_2/H_2O and (b) O_2/CO_2 condition at $x = 30$ mm and $f_{o2} = 9\%$.

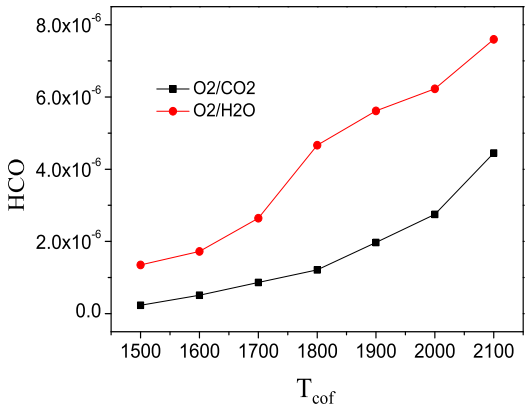
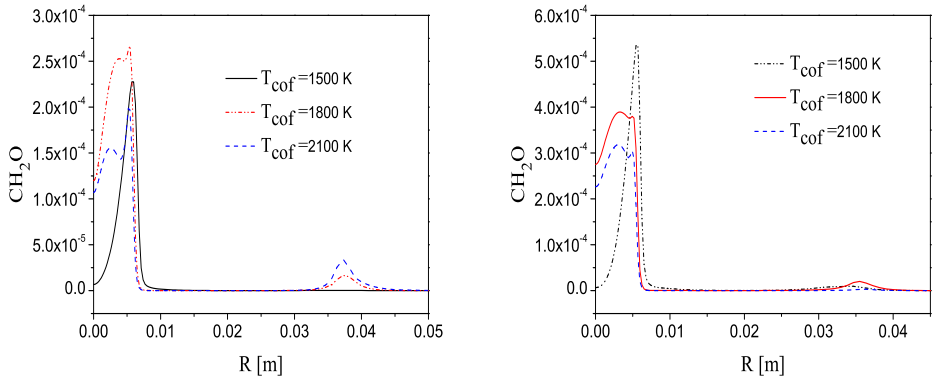


Fig. 27. The maximum of HCO concentration at various T_{cof} and $f_{o2} = 9\%$.



(a) (b)

Fig. 28. CH_2O profile in (a) O_2/H_2O and (b) O_2/CO_2 condition at $x = 30$ mm and $f_{o2} = 9\%$.

Table 1
Computational conditions of the present work.

Case	Fuel Flow			Co-flow					Tunnel Flow			
	u (m/s)	T (K)	CH_4 (%)	**u** (m/s)	T (K)	O_2 (%)	H_2O (%)	CO_2 (%)	**u** (m/s)	T (K)	H_2O (%)	CO_2 (%)
O_2/H_2O	60	305	100	3.2	1500 / 2100	6 / 18	82 / 94	0	3.2	400	100	0
O_2/CO_2	60	305	100	3.2	1500 / 2100	6 / 18	0	82 / 94	3.2	400	0	100