- Methane Combustion in MILD Oxyfuel Regime: Influences of Dilu-
- 2 tion Atmosphere in Co-flow Configuration
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- 15 Abstract: MILD (moderate or intense low oxygen dilution) oxyfuel com-
- bustion is a recently proposed clean combustion mode which can remedy
- 17 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
- 22 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.

33 Keywords: MILD combustion; Oxyfuel combustion; Methane; CO₂-dilution;

 $_{34}$ H_2O -dilution; oxy-steam

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36 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 O₂/CO₂ 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 O₂/H₂O atmosphere
where oxygen in oxidant flow is diluted by steam rather than carbon dioxide
[1]. As shown in Ref.[1], compared with its O₂/CO₂ counterpart, there are
at least three advantages to realize MILD oxyfuel combustion in O₂/H₂O 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 O₂/H₂O 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 CO₂- and H₂O-dilution on combustion temperature and reaction kinetics of methane [12]. It was observed that the chemical and thermal effects of CO_2 and of H₂O 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 H₂O in recycled flue gas could influence combustion characteristics of pulverized coal significantly [14–16]. However, these studies [12–16] all are limited in the socalled "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 CO₂ and H₂O 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 CO₂ and of H₂O 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 O₂/H₂O 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 O₂/H₂O option the auto-ignition delay time would be a little shorter than its O₂/CO₂ counterpart. Recently, some of the present authors conducted a 101 numerical investigation about the influence of H₂O addition on MILD oxy-coal 102 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 104 that NO emission could be suppressed and heat transfer would be enhanced 105 in O₂/H₂O atmosphere. As the IFRF (International Flame Research Foundation) semi-industrial scale co-flow furnace adopted in Ref. [17] is not an ideal 107 MILD oxyfuel combustion research prototype and the extreme complication of coal combustion, Ref. [17] failed to reveal the influence of different types of 109 dilution gases (H₂O or CO₂) 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 114 counter-flow counterpart because flow-reaction interaction, which is exclud-115 ed 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 117 our knowledge in this emerging area so to advance its application in energy 118 industry, a systematic comparison between the performance of co-flow MILD 119 oxyfuel combustion in O_2/H_2O condition and that in O_2/CO_2 atmosphere, is essential. 121

In order to bridge the aforementioned gap, in this work we numerically investigate methane combustion in MILD oxyfuel regime, diluted by carbon dioxide 123 and steam, respectively. The JHC (jet in hot co-flow) burner developed in 124 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 burn-127 er 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 130 numerical simulation, so firstly the adopted numerical approach is validated 131 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.

140 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 tem-155 perature of tunnel flow is set as 400 K to guarantee H₂O at its steam status in tunnel flow. Finally, to reduce the complication induced by variation of fuel 157 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 Re-159 f.[20]. Table 1 lists the detailed information of investigated cases covered by 160 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 163 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 167 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 170 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. 174

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 180 emissivity [19]. Finally, the eddy dissipation concept (EDC) model [23] with 181 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, 184 as illustrated by Fig.2, which is the same as that used in Ref.[19]. As demon-185 strated by our recent work [19], such grid resolution is fine enough to obtain 186 grid-independent numerical prediction. The detailed information about grid discretization and numerical convergence please refer to our previous work 188 [19].189

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.

2 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 $6\% \le f_{o2} \le 18\%$ 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 242 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 246 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 CO₂- and in H₂O-dilution condition until the present 253 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 261 and slender), which is similar with its air MILD combustion counterpart [27]. 262 However, the shape of OH contours in O₂/H₂O atmosphere looks like but-263 terflies' wings (namely relative wider but shorter), quite different from its air MILD combustion [27] and CO₂-dilution counterpart [6]. In the "feedback" 265 oxy-steam combustion [14–16], whether there exists a similar feature is still 266 an open question and we will try to answer it in our future work. As in the 267 oxy-steam condition the "flame" front is closer to the fuel jet nozzle, it is easier to establish the MILD oxyfuel regime in H₂O-dilution atmosphere. This 269 conclusion agrees with that drawn from its counterflow counterpart [1]. Figure 270 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 $\mathrm{O}_2/\mathrm{CO}_2$ counterpart since the production of OH will be enhanced by H₂O addition, as explained in detail in our previous work [12]. In Ref. [12], a counter-flow combustion prototype was adopted and the research 276 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 279 dragonfly-wing-like OH distribution, in O₂/CO₂ condition it is easier to sus-280 tain 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 sug-284 gested to use the contour of CO mass fraction $f_{co}=0.01$ as an indicator to 285 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 dramatical-287 ly with more oxygen addition. However, in its O_2/CO_2 counterpart, the size 288 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 291 oxyfuel combustion in the IFRF semi-industrial scale furnace was investigat-292 ed. 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 CH₄ is mainly 298 determined by reaction rate. It can be looked as a kinetic-controlled combustion. However, in the present work, the consumption speed of CH₄ depends 300 not only on reaction rate but also on local available oxygen concentration s-301 ince in the present JHC configuration there is no oxygen in the tunnel flow. In 302 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 307 with that from the semi-industrial scale furnace [5]. Nowadays there appear 308 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 311 carefully before extending the conclusions claimed in these studies to realistic 312 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 314 be observed that the "flame" size in oxy-steam condition is generally smaller 315 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 tempera-317 ture and OH distribution. In Ref. [6], it was observed that with more oxygen 318 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 321 presented in Ref.[12]. Through the present work, we find such conclusion is 322 also true in O₂/H₂O 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 325 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, 330 the CO maximum in both dilution atmosphere will grow almost linearly with 331 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 334 counter-flow "feed-back" oxyfuel combustion. Consequently, it is also a common feature of oxyfuel combustion. The detailed explanation on how H₂O addition will suppress CO generation please refer to Ref. [12]. Figures 10-11 337 plot the CO profiles at different axial positions x = 90 and x = 120 mm. The 338 profiles in both O₂/CO₂ and O₂/H₂O condition are similar with their air-firing counterpart: the gradient of CO concentration along radial direction becomes 340 sharp with more oxygen addition [6,20]. Furthermore, it can be observed in 341 O₂/H₂O 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, O₂ and OH in O₂/N₂ atmosphere and those in O₂/CO₂ condition are very small. However, through the present work, it is observed that except O₂, there are obvious differences in most scalar distributions between in O₂/H₂O and in O₂/CO₂ 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 373 peak value of HCO would grow up with f_{o2} . The present results are consistent 374 with it and prove this conclusion also can hold water in its oxy-steam counter-375 part. 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, 377 HCO concentration in O_2/CO_2 condition is always higher than its O_2/H_2O 378 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 381 to be sustained in O_2/CO_2 condition. In our previous work [12], it was also 382 observed that HCO concentration in O₂/H₂O condition was lower than its 383 O₂/CO₂ counterpart, which resulted from that H₂O addition would modify the chemical equilibrium of the reaction step R46. Through the present work, 385 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 (CH₂O) which can serve as an indicator for ignition [2,26]. Since CH₂O predominantly exists in low temperature condition, therefore the concentration of CH₂O 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 H₂O addition will suppress CH₂O 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 O₂/CO₂ condition, the profiles of CH₂O alter sensitively to 396 the variation of f_{o2} , while in O_2/H_2O atmosphere the change is relatively 397 smaller. It implies the establishment of MILD combustion in O₂/H₂O condition receives less influence by oxygen fluctuation. Consequently, it is easier 399 to establish MILD combustion regime in oxy-steam condition. It is consistent 400 with the above analysis and the conclusion in Ref.[1]. Fig. 16 (b) depicts the maximums of CH₂O in both dilution atmosphere. The peak value of CH₂O in CO₂-dilution atmosphere is always larger than its H₂O-dilution counterpart, 403 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 CO₂-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 $1500K \le T_{cof} \le 2100K$ 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 ob-415 served in Ref. [2]. Refs. [2,6] just focused on O_2/CO_2 atmosphere. Through the 416 present study, we can confirm these phenomena exist in oxy-steam condition, 417 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 419 types of dilution atmosphere differ with each other obviously, especially in the 420 vicinity of the fuel jet nozzle. In addition, the high temperature zone expands more quickly in CO₂-dilution atmosphere. The maximum temperature of the reactants is illustrated by Fig.18. T_{max} is a monotonic increasing function of 423 T_{cof} in both dilution conditions and since $T_{cof} \ge 1600 \text{ K } T_{max}$ 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 H₂O is larger than CO₂. However, their gap will decrease against 427 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 430 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 H₂O-dilution condition looks like a dragonfly's wing, 438 similar with its CO₂-dilution counterpart although the former is shorter. S-439 ince $T_{cof} \geq 1600$ K, the "flame" front region in oxy-steam condition expands substantially along the radial direction and now the shape of OH contours in H₂O-dilution condition looks like a butterfly's wing, not resembling that in $\mathrm{O}_2/\mathrm{CO}_2$ 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 O₂/H₂O condition ascends much faster than in O₂/CO₂ 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 449 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 451 concentration in either dilution atmosphere will grow up with T_{cof} , which is 452 consistent with the result reported in Ref. [6]. As mentioned above, as the peak 453 temperature in O_2/H_2O condition is lower than its O_2/CO_2 counterpart, the 454 maximum of OH concentration in the former is always higher than the latter. 455 The increasing rate of the maximum of OH concentration in oxy-steam atmo-456 sphere is much faster than its CO₂-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-469 steam condition. If taking the contour of CO mass fraction $f_{co} = 0.01$ as an 470 indicator to visualize the "flame" size, as mentioned above, one can observe 471 that the "flame" size in O_2/H_2O atmosphere changes substantially with T_{cof} . When $T_{cof} = 1500$ K, the "flame" size in H₂O-dilution condition is much smaller than its CO₂-dilution counterpart. Then the "flame" size in the former 474 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 CO₂-dilution atmosphere grows slightly. 478 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 483 slower than its O_2/CO_2 counterpart. Taking Figs. 9 and 22 together, one can 484 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 486 room as the maximum of CO concentration at the outlet of the investigated 487 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 494 operation in CO₂-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 atmo-511 sphere, especially in the far-field. Against T_{cof} increasing, O_2 concentration 512 near the exit will decrease as a higher T_{cof} will intensify chemical reactions. 513 Near the fuel jet nozzle, the influence of variation of T_{cof} on O_2 distribution is 514 slight, as illustrated by Fig.25. In Ref. [29], it was also found that, in methane-515 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 517 methane MILD JHC combustion, regardless of dilution atmosphere. Taking 518 Figs. 13 and 24 together, it can be observed that for any T_{cof} and f_{o2} , in 519 the far-field the O_2 radial concentration in O_2/H_2O condition is always low-520 er than its O_2/CO_2 counterpart. Such observation demonstrates once again that in oxy-steam condition the "flame" size is smaller than its CO₂-dilution 522 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 527 whatever T_{cof} is, HCO concentration in O_2/CO_2 condition is always higher 528 than its O_2/H_2O counterpart, as shown by Fig.27 (a). At a relative low coflow temperature $(T_{cof} = 1500 \text{ K})$, one can observer HCO concentration in 530 oxy-steam atmosphere is rarefied. According to Fig. 27, it can be observed 531 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 534 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 538 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 CO₂-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 CO₂dilution 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 CH₂O concentration along the radial direction while in Ref. [29] only one was 549 observed. In O₂/CO₂ condition, the second peak of CH₂O distribution along 550 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 552 will take place over a wider range than in O_2/CO_2 or O_2/N_2 condition. Fur-553 thermore, it also indicates the effect of T_{cof} on ignition is more complicated in H₂O-dilution condition. In addition, in Ref. [29], it was observed that the "sharp angle" of the CH₂O 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 557 in methane-air MILD combustion, the present work reveals that this conclusion is also tenable in the MILD oxyfuel combustion regime, either diluted by 559 CO₂ or by H₂O. 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].

563 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 oxyfuel 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:
- 576 (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 578 CO₂-dilution atmosphere is much larger than its O_2/H_2O counterpart. The 579 maximum concentration of CO in O_2/CO_2 atmosphere is about ten times 580 large than its steam-dilution counterpart.
- T_{cof} on ignition is more complicated in H₂O-dilution condition.
- $_{584}$ (3) In general, it is easier to establish the MILD oxyfuel regime in $_{2}$ O-dilution atmosphere and in $_{2}$ /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].

588 (4) In Ref.[2], it was reported that in the near field of the JHC, the differences 589 between the profiles of CO, O₂ and OH in O₂/N₂ and in O₂/CO₂ atmosphere 590 are very small. However, through the present work, it is observed that ex-591 cept O₂, there are obvious differences in most scalar distributions between in 592 O₂/H₂O and in O₂/CO₂ condition, even in the near field. Consequently, one 593 should pay great attention on burner design for MILD oxy-steam combustion 594 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 H₂O will change from the dragon-wing-style to butterfly-wing-style? It is an
important question as "feed-back" oxyfuel combustion diluted by H₂O 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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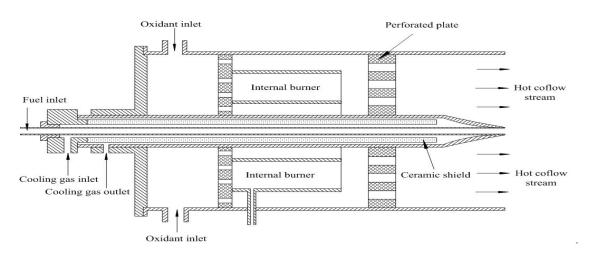


Fig. 1. Configuration of the JHC burner proposed in Ref. [20].

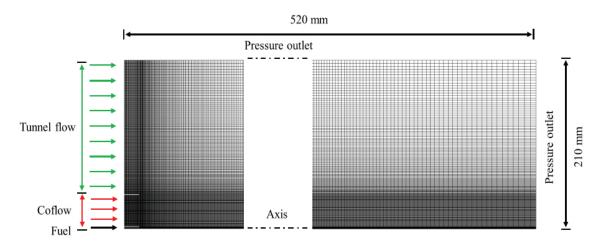


Fig. 2. Schematic configuration and coordinate system of the computational domain.

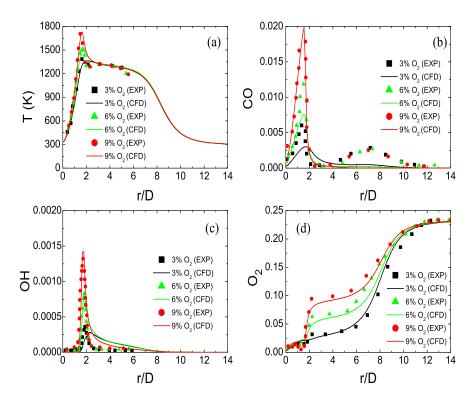


Fig. 3. comparison of (a) temperature, (b) CO mass fraction, (c) OH mass fraction, (d) O_2 mass fraction profiles between the present numerical prediction (CFD) and the experimental measurements (EXP) for JHC air MILD combustion with 3%, 6%, and 9% oxygen mass fraction in the co-flow at the axial location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

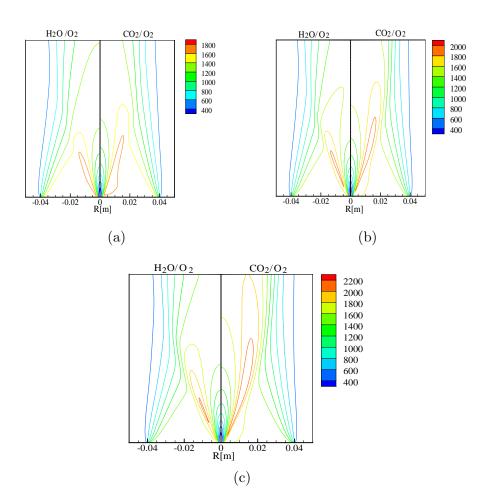


Fig. 4. Temperature distribution in O_2/H_2O or O_2/CO_2 condition at (a) $f_{o2}=6\%$ (b) $f_{o2}=12\%$ and (c) $f_{o2}=18\%$: $T_{cof}=1800$ K.

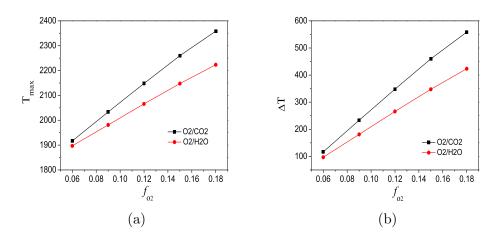


Fig. 5. Maximum temperature (a) and temperature rise (b) at various f_{o2} and $T_{cof}=1800~\mathrm{K}.$

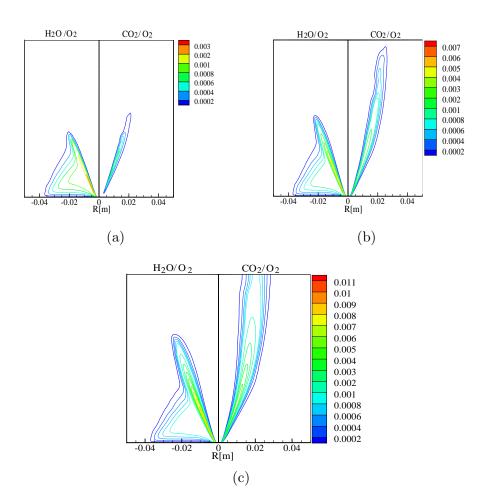


Fig. 6. OH distribution in O_2/H_2O or O_2/CO_2 condition at (a) $f_{o2}=6\%$ (b) $f_{o2}=12\%$ and (c) $f_{o2}=18\%$ and $T_{cof}=1800$ K.

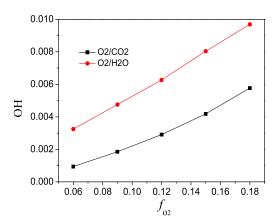


Fig. 7. The maximum of OH concentration at various f_{o2} and $T_{cof}=1800$ K.

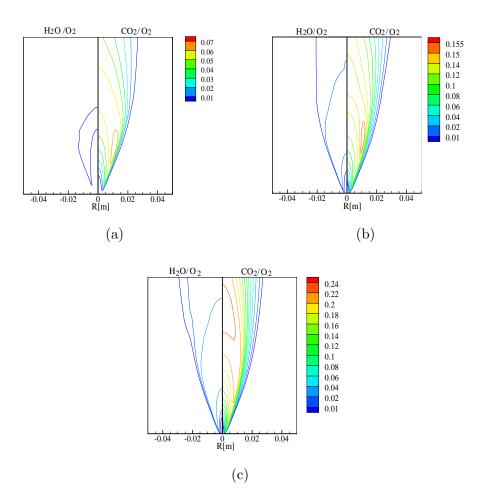


Fig. 8. CO distribution in O_2/H_2O or O_2/CO_2 condition at (a) $f_{o2}=6\%$ (b) $f_{o2}=12\%$ and (c) $f_{o2}=18\%$ and $T_{cof}=1800$ K.

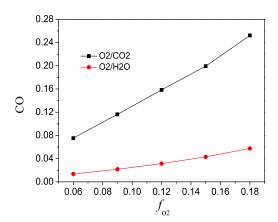


Fig. 9. The maximum of CO concentration at various f_{o2} and $T_{cof}=1800$ K.

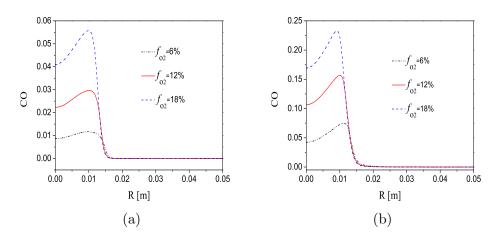


Fig. 10. CO profile in (a) ${\rm O_2/H_2O}$ and (b) ${\rm O_2/CO_2}$ condition at x=90 mm and $T_{cof}=1800$ K.

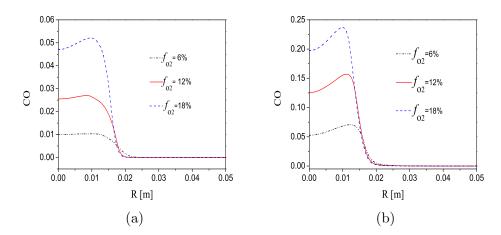


Fig. 11. CO profile in (a) $\rm O_2/H_2O$ and (b) $\rm O_2/CO_2$ condition at x=120 mm and $T_{cof}=1800$ K.

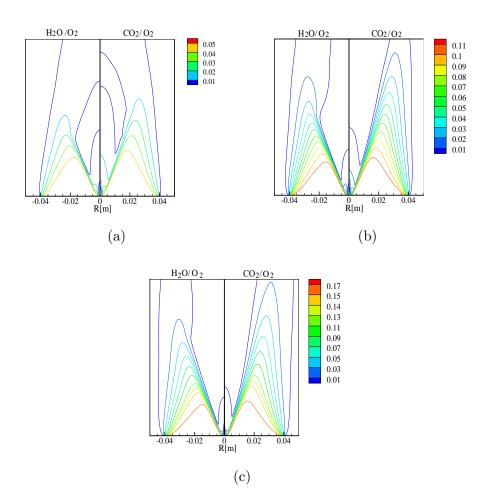


Fig. 12. O₂ distribution in O₂/H₂O or O₂/CO₂ condition at (a) $f_{o2}=6\%$ (b) $f_{o2}=12\%$ and (c) $f_{o2}=18\%$ and $T_{cof}=1800$ K.

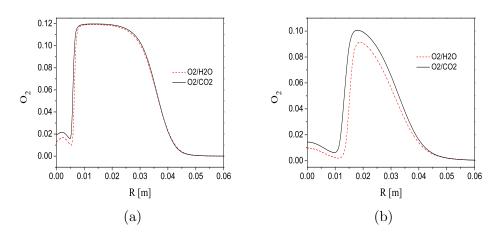


Fig. 13. O₂ profile at (a) x=30 mm and (b) x=90 mm : $f_{o2}=12\%$ and $T_{cof}=1800$ K.

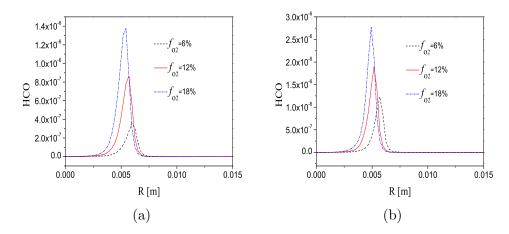


Fig. 14. HCO profile in (a) $\rm O_2/H_2O$ and (b) $\rm O_2/CO_2$ condition at x=30 mm and $T_{cof}=1800$ K.

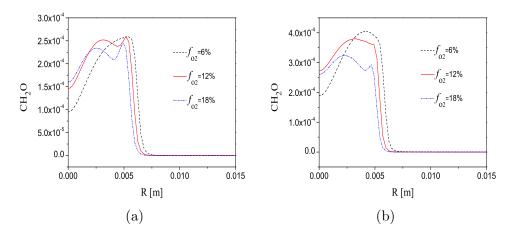


Fig. 15. CH₂O profile in (a) O₂/H₂O and (b) O₂/CO₂ condition at x=30 mm and $T_{cof}=1800$ K.

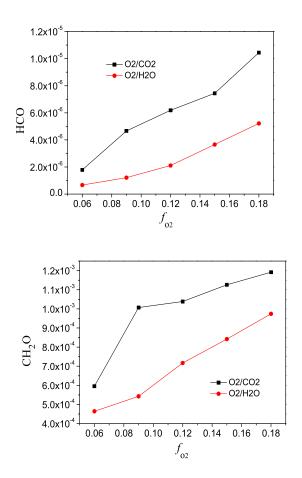


Fig. 16. The maximum of HCO (a) and CH₂O (b) concentration at various f_{o2} and $T_{cof}=1800$ K.

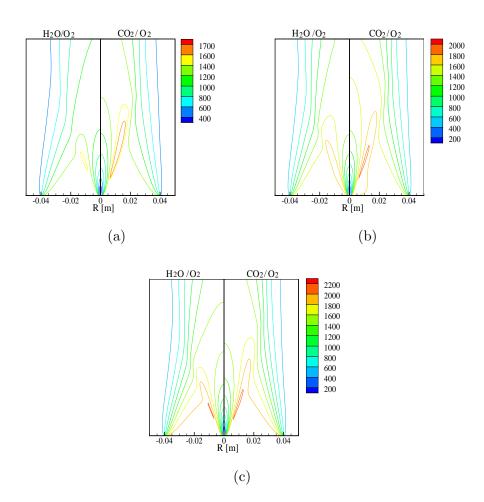


Fig. 17. Temperature distribution in O_2/H_2O or O_2/CO_2 condition at (a) $T_{cof}=1500~{\rm K}$ (b) $T_{cof}=1800~{\rm K}$ and (c) $T_{cof}=2100~{\rm K}$: $f_{o2}=9\%$.

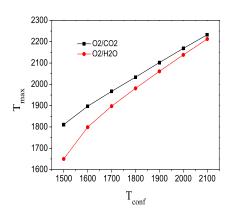


Fig. 18. Maximum temperature at various T_{cof} and $f_{o2} = 9\%$.

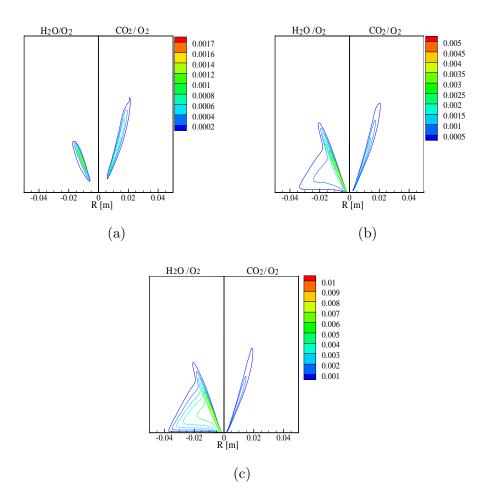


Fig. 19. OH distribution in O_2/H_2O or O_2/CO_2 condition at (a) $T_{cof}=1500$ K (b) $T_{cof}=1800$ K and (c) $T_{cof}=2100$ K: $f_{o2}=9\%$.

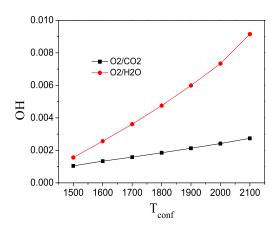


Fig. 20. The maximum of OH concentration at various T_{cof} and $f_{o2}=9\%$.

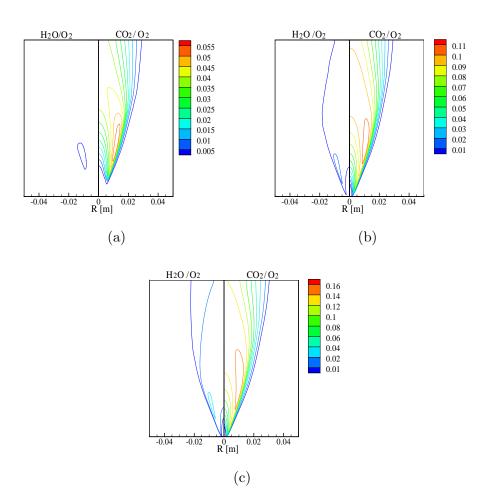


Fig. 21. CO distribution in O_2/H_2O or O_2/CO_2 condition at (a) $T_{cof}=1500$ K (b) $T_{cof}=1800$ K and (c) $T_{cof}=2100$ K: $f_{o2}=9\%$.

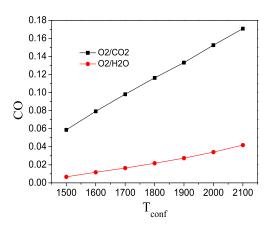


Fig. 22. The maximum of CO concentration at various T_{cof} and $f_{o2} = 9\%$.

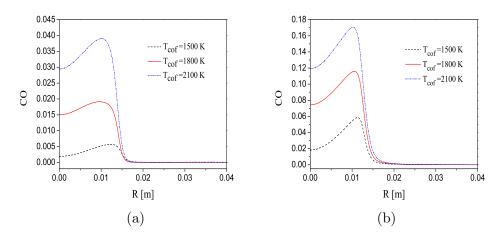


Fig. 23. CO profile in (a) $\rm O_2/H_2O$ and (b) $\rm O_2/CO_2$ condition at x=90 mm and $f_{o2}=9\%$.

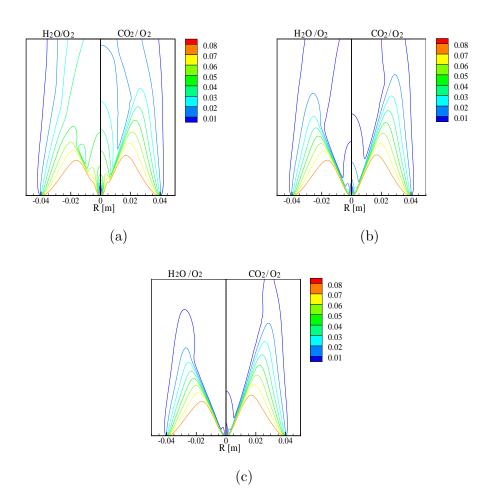


Fig. 24. O₂ distribution in O₂/H₂O or O₂/CO₂ condition at (a) $T_{cof}=1500$ K (b) $T_{cof}=1800$ K and (c) $T_{cof}=2100$ K: $f_{o2}=9\%$.

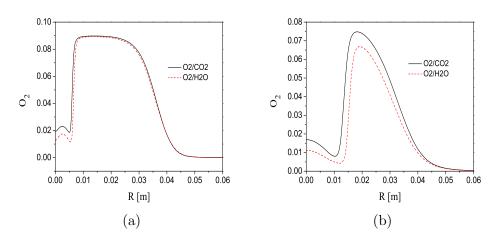


Fig. 25. O₂ profile at (a) x=30 mm and (b) x=90 mm : $f_{o2}=9\%$ and $T_{cof}=2100$ K

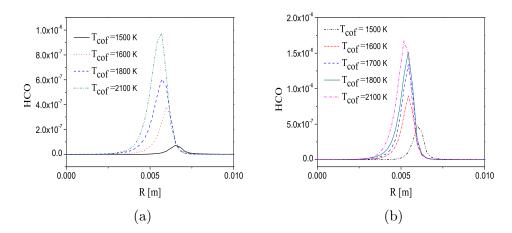


Fig. 26. HCO profile in (a) ${\rm O_2/H_2O}$ and (b) ${\rm O_2/CO_2}$ condition at x=30 mm and $f_{o2}=9\%$.

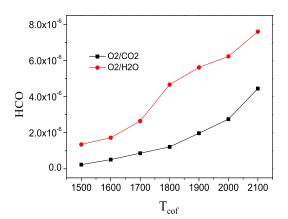


Fig. 27. The maximum of HCO concentration at various T_{cof} and $f_{o2}=9\%$.

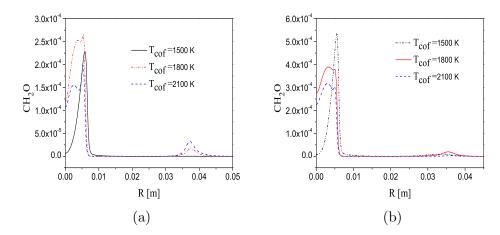


Fig. 28. CH₂O profile in (a) O₂/H₂O and (b) O₂/CO₂ condition at x=30 mm and $f_{o2}=9\%$.

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 ₂ O (%)	CO ₂ (%)	u (m/s)	T (K)	H ₂ O (%)	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