Study on Pure Oxygen Exhaust Gas Combustion: Key Technology of CO2 Capture for High Temperature Fuel Cell With Coal Syngas

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Abstract

IGFC based on high temperature SOFC coupled with CO₂ capture process provides a new technology route of high efficiency and nearly zero CO₂ emission power system. Flame burning is an ideal method for tail gas treatment. In this paper, an oxy-combustor for a 10kW IGFC system anode exhaust gas is experimentally and numerically studied. Simulation method is verified by experiments. Key performances of the combustor are studied under different system process design conditions. 315K might be an ideal condensation temperature before burning for flame stability. CO could be almost fully converted under flame burning condition. CO₂ concentration after burning is over 0.958 when excess O₂ is less than 5%. Overall, 5% excess O₂ could be recommended for environment consideration. An optimal tangential angle exists around 25° for liner temperature controlling. Systems with anode cycling might release less CO pollutant in theory for no addition of extra H₂O. Total fuel utilization percent had better be high enough to make oxygen flame temperature of anode exhaust gas lower than 1800K to make systems environment friendly. The results would be of great value to IGFC and CO₂ capture combined system designing.

1. Introduction

Traditional coal utilization power generation systems release amounts of CO₂ which causes greenhouse effect. Integrated gasification fuel cell (IGFC) based on high temperature solid oxide fuel cells (SOFC) coupled with CO₂ capture process provides a new technology route of high efficiency and nearly zero CO₂ emission power system (Thallam Thattai et al. 2017 and Al-Khori et al. 2020). Carbon element in anode syngas should be gathered to gain a CO₂ high capture rate. The fore-gathering technology such as water-gas-conversion (WGS) could be taken before SOFC (Wu et al. 2020). SOFC with high fuel utilization percent (Pirasaci, 2019) accomplishes inner-gathering. However, there still exists considerable combustible component mainly consisting CO and H₂ in exhaust gas. Thus, further measures are needed.

In some commercial SOFC systems like Bloom Energy, anode looping is taken into consideration to increase fuel utilization. Generally, exhaust gas needs to be completely oxidized to CO₂ and H₂O (Spallina et al. 2018) for either energy utilization improving or high CO₂ concentration. For CO₂ capture, oxy-combustion has been proved to be an effective method for CO₂ concentrating in power system (Francisco et al. 2016 and Chen et al. 2019). Catalytic combustion is reported in some process design study for lean CO fraction in anode tail gas (Wang et al. 2020). However, it is shown in recent reports that CO conversion of catalytic combustion is not high enough to reduce CO pollutant emission into ppm level especially in lean CO condition because of low reaction temperature, which may not meet the environment protecting requirements (Chen et al. 2019). Catalyst for CO converting may not show a stable performance under long term and high temperature (Liu et al. 2018). Therefore, it may be complex to put catalytic combustion into system design.

Direct flame burning shows another way to deal with the anode exhaust gas. It is indicated in some study that flame burning can almost fully convert CO to CO₂ even in low calorific gas, though may cause
problems on NO\textsubscript{x} emissions (Ilbas et al. 2019). Moreover, flame could be steady under relatively wide range of equivalent ratio, which means a better performance on reacting stability (Rashwan et al. 2017, Imteyaz et al. 2018 and Li et al. 2018). These might make flame burning an ideal method for tail gas treatment. However, calorific value of exhaust gas, which may be 2–3 MJ/Nm\textsuperscript{3}, is rather low for flame burner. Meanwhile, water vapor and carbon dioxide are main components in anode exhaust gas. Therefore, flame stability for exhaust gas needs further discussion not only because the low calorific value, but also both steam and carbon dioxide show some negative effects on it (Grosseuvres et al. 2018 and Ning et al. 2018). Approaching high CO\textsubscript{2} concentration means that flame should be burnt around equivalent ratio which leads to high flame temperature. For long-life systems, it is important control creep deformation of devices, which make liner temperature worthy to investigate. Moreover, high reacting temperature of oxy-combustion leads to high level NO\textsubscript{x} emission possibility since N\textsubscript{2} still exists in syngas (Schluckner et al. 2020). It should be taken into consideration because acidic gas may be harmful to systems and environment.

In this paper, an oxy-combustor for a 10 kW IGFC system anode exhaust gas is experimentally and numerically studied. CFD and chemical reactor network (CRN) method are used to simulate the flame and emissions of combustor. Detailed chemical kinetic scheme is taken to increase simulation accuracy. Flame stability of exhaust gas and pure oxygen with high steam fraction is discussed. Performance of the combustion on conversion rate and liner temperature is investigated. Pollutant emissions under different conditions are also studied. The results on the above key performance would be of great value to IGFC and CO\textsubscript{2} capture combined system designing.

2. Experimental Setup

As mentioned above, the exhaust gas components vary with the system design. Usually, fuel cells may not utilize CO directly. CO in syngas should be converted to H\textsubscript{2} through WGS reactions. As shown in Fig. 1, two different systems are compared in this work. Syngas with steam blended in gets through anode in a single pass, which is regarded as single case. Inlet syngas mixed with a part of humid anode exhaust gas before enter anode, which is known as cycling case. Exhaust gas in single case contains higher ratio of H\textsubscript{2} and H\textsubscript{2}O since extra steam is bring in for WSG process before. Figure 2 shows the exhaust gas oxy-combustor which mainly consists of a flame chamber, a flame stabilizator, oxygen swirler and fuel swirler. Swirlers and stabilizator provide a flow map with swirling and back flowing which could improve mixing process and be beneficial to stability of flame root. Cooling air enter cooling chamber to cool flame chamber liner.

The test rig is shown in Fig. 3. CO The modeled exhaust gas composed of H\textsubscript{2}, CO and CO\textsubscript{2} is taken as fuel. Gas is firstly mixed in a mixer with heating function, and then enters a steam evaporator from the bottom. At top of the evaporator, saturated vapor fills the room. Fuel together with vapor goes through the evaporator and gets into the combustor. Mole fraction of steam in the fuel could be adjusted by temperature of the evaporator. Operating conditions of the experiment are shown in Table 1. Exhaust gas
component of two systems under different total fuel utilization is shown. Other slight inert components are replaced by CO\(_2\) in this test. Cooling air through cooling chamber makes forced convective cooling on the liner. Convective coefficient is controlled by flow rate of air. Exhaust gas is burned with pure oxygen inside the combustor. An industrial gas analyzer with drying function is used to measure CO\(_2\), O\(_2\) and CO content in flue gas. To burn CO out, O\(_2\) is excessively supplied by 5% of equivalent. The temperature of the steam evaporator is kept at 315K.

**Table 1**

| Case     | Flow rate (kg/h) | Total Fuel Utilization (%) | O\(_2\) Flow rate (kg/h) | Exhaust Gas Mole Fraction |
|----------|------------------|----------------------------|---------------------------|---------------------------|
| Single1  | 3.48             | 86                         | 0.47                      | CO 0.069, H\(_2\) 0.197, H\(_2\)O 0.061, CO\(_2\) 0.673 |
| Single2  | 3.48             | 80                         | 0.69                      | CO 0.091, H\(_2\) 0.261, H\(_2\)O 0.061, CO\(_2\) 0.587 |
| Cycling1 | 3.45             | 86                         | 0.45                      | CO 0.101, H\(_2\) 0.169, H\(_2\)O 0.061, CO\(_2\) 0.669 |

3. **Computational Method**

3.1 **CFD method**

A CFD method is used to simulate flame inside the swirling combustor, which has shown good performance (Wang et al. 2015 and Wang et al. 2016). A modified realizable k-\(\varepsilon\) turbulent model and the eddy-dissipate concept (EDC) combustion model coupled with detailed chemical mechanism are used. The physical property is calculated by the transport and thermal package in the chemical mechanism. Ideal gas law is adopted to compute the density of gas mixture, and the specific heat is calculated by the mixing-law. The thermal conductivity and the kinematic viscosity are computed through the ideal-gas mixing-law. Kinetic theory is used to calculate the mass diffusivity and the heat diffusivity. The radiation is simulated by the discrete ordinate (DO) model. The liner wall is set as radiative and convective boundary. Commercial software is used to solve modeling equations.

3.2. **Chemical reactor network method**

To simulate the emissions, a CRN model for the combustor is established according to CFD results obtained above. The modeling process was reported, verified and used in previous reports (Wang et al. 2016 and Shao et al. 2017). The model is shown in Fig. 4. PSRs upstream are used to simulate the fuel/oxygen unmixedness in primary zone. Reactors downstream simulate the developing zone. A detailed chemical mechanism consisting of a syngas combustion scheme and a NO\(_x\) chemistry scheme is adopted in this study, which was previously verified (Wang et al. 2016). The CRN is solved in the commercial software. CO emission simulation results of experiment cases are shown in Fig. 5 compared
4. Results And Discussion

4.1. Vapor effects on flame stability

In anode exhaust gas, moisture level could be extremely high which makes it difficult to gain a steady flame in the combustor. It is mainly because some radical decomposed from H$_2$O suppresses burning reactions of syngas. Therefore, in IGFC systems, anode exhaust gas should be condensed before oxy-combustor to remove some water. In that case, steam partial pressure at entrance of combustor is determined by the saturated vapor pressure. A single PSR model is used to study the relationship between the condensation temperature and flame temperature, which is of great importance to flame stability. Ignite temperature is set to 1100K. Resident time is 1 s. Results are shown in Fig. 6.

It is shown in Fig. 6 that the equivalent adiabatic flame temperature T$_{ad}$ decreases with combustor inlet temperature increasing. It is because moisture content is lower under lower condensation temperature. If the condensation temperature is higher than 350K, flame temperature is lower than ignition temperature which means excess steam existing and failing to ignite. In practical systems, steam could not be wiped out completely which make T$_{ad}$ always lower than it without vapor. For cycling1 case, T$_{ad}$ under dry condition is around 1700K. In view of heat loss during burning process, it might be proper to have a T$_{ad}$ 500K higher than ignition temperature to gain a steady flame. Based on above, 315K might be an ideal condensation temperature for system process design where Tad is about 1680K.

4.2. CO conversion

For the exhaust gas treating device aiming at CO$_2$ capture, the most important indicator is the processing capacity. For oxy-combustor, it could be represented by the dry CO$_2$ mole fraction in exhaust gas after burning. CO conversion after combustor could be determined by the flame temperature and oxidant quantity. Beside equivalent ratio, heat loss affects flame temperature directly. As mentioned, forced convection is set outside flame chamber to control the liner temperature which would certainly bring heat releasing problem. CFD method is used to calculate the CO$_2$ mole fraction after treatment under different equivalent ratio and liner convective coefficient conditions. Simulation results are shown in Fig. 7. Besides, data of experiment above is shown in Table 2.

It shows in Fig. 7 that CO$_2$ mole fraction after burning under dry condition is higher than 0.958 under every calculating cases. It indicates that the existing combustor performs well in tail gas treating. Convective coefficient seems to show slight effects on CO conversion, which may due to tiny influence to flame temperature caused by liner heat loss. CO mole fraction varies obviously with excess O$_2$ quantity.
relatively. It should be noticed that CO₂ content decreases when excess O₂ quantity rises. This confirms that CO could be almost fully converted under flame burning condition (Ilbas et al. 2019). Unreactive O₂ decreases the CO₂ mole fraction. For practical device, excess O₂ helps CO being burned out which reduces CO emission in consideration of unexpected unmixedness at jet outlets. Therefore, 5% excess O₂ could be recommended since an acceptable CO₂ capture rate is gained. The liner convection could be enhanced as far as possible for it shows little influence on CO conversion. Table 2 shows that CO₂ concentration of dry flue gas after burning is over 0.958, which could be considered as an ideal rate for capture. Single cases have a slightly higher concentration percent due to more H₂ and H₂O content. They would be dried out before capture process. It is experimentally proved that CO can be converted to CO₂ efficiently by fully oxy-combustion.

| Case       | CO₂ mole fraction |
|------------|-------------------|
| Single1    | 0.962             |
| Single2    | 0.962             |
| Cycling1   | 0.958             |

### 4.3. Liner temperature

Liner temperature of combustor should be paid attention to since high temperature leads to creep deformation which may damage the combustor especially for long term running systems. That might be another key indicator for oxy-combustor evaluating. Mostly, the liner temperature is determined by flame temperature inside. However, for an oxy-combustor aiming at CO₂ capturing, equivalent ratio might varies in a narrow range. It makes total flame temperature range relatively small especially for diffusion flame. In that case, liner temperature is influenced by heat transfer inside and outside. Heat transfers in from high temperature burning gas and then out to cooling air outside chamber. For anode exhaust gas characterized by extremely low calorific value, strong swirling might be bring in to the flow field organizing inside to stabilize the flame. Tangential angle of fuel jet determines centrifugal force of flow which would affect convective heat transfer inside liner for fuel flow rate is times to oxygen flow rate. Moreover, tangential angle of jet influences mixedness of fuel and oxygen which may affect local flame temperature. Maximum liner temperature is calculated by CFD model under different outside convective efficient and fuel jet tangential angle. Results are shown in Fig. 8.

Max liner temperature decreases with the outside convective coefficient, which proves that force convection outside flame chamber is an effective way to protect the combustor. It can be noticed that max liner temperature has an optimal value at tangential angle around 25°. Main reason for this might be as following. Stronger swirling makes a better mixing condition. When jet tangential angle is around 20°, mixing between fuel and oxygen is not ideal. High level of unmixedness leads flame to burning under low
equivalent ratio, which causes high local temperature. Liner temperature increases when local temperature is high. When jet tangential angle is around 30°, larger centrifugal force makes high temperature gas scour liner more fiercely which may cause high liner temperature, although mixing level is more ideal. It can be inferred that an optimal tangential angle, which could vary with dimensions of combustor, exists for liner temperature controlling.

4.4. Pollutant emissions

Pollutant emission is always an important indicator for different kinds of power plants. For SOFC, electrochemical reactions take place inside fuel cells. Differing from traditional plants, no chemical energy converts to heat which might cut pollutant formation process. However, oxy-combustor adds burning step into systems which may cause some emission problem. In practical IGFC system, syngas contains amount N₂ even O₂ gasification technology is involved. So combustor pollutant is mainly consisting of CO and NOₓ. CRN model is used to compute pollutant emissions of the oxy-combustor. Cycling1 case with a total fuel utilization rate of 86% is computed compared with Single2 case whose total fuel utilization rate is 80%. As shown above, this leads to a difference between components of exhaust gas. In this period, exhaust gas contains 3% N₂ by volume in simulation case. Ratio of other component is kept as before. Emissions under different excess O₂ quantity are computed. Results are shown in Fig. 9 and Fig. 10.

It is shown that both CO and NOₓ emissions decrease with rising of excess O₂ percent. It is known that NOₓ formation is directly related to flame temperature. When excess O₂ percent is low, local flame temperature could be higher which leads to higher NOₓ emissions. On the other side, more excess O₂ supply makes CO burnt more completely which means lower CO emissions. Therefore, excess O₂ supply is in favor of pollutant reducing. Combined with mentioned, if CO₂ concentration could meet the requirement of CO₂ capturing, O₂ should be supplied as more as possible. Barely influence of excess O₂ percent is shown on combustor exit temperature since O₂ flow rate varies slightly. Single2 case releases more CO and NOₓ than Cycling1. It is because that Single2 case has a lower total utilization rate of 80% in fuel cells which means exhaust gas contains more combustible component. Flame temperature of Single2 case is about 2000K whereas it is 1650K for Cycling1 case. High temperature leads to high NOₓ emission. Besides, Single2 case contains more H₂ so that H₂O mole fraction is high after burning. Water vapor prevents CO converting to CO₂ especially under high temperature, which leads to high CO emission level (Wang et al. 2016). The experiment data above also shows some agreement to that. At this perspective, anode cycling design seems to have a better performance on emission reducing in theory compared with single pass design for no extra steam addition and less H₂, though more experiments are needed further. As known, NOₓ formation increases extremely when temperature exceeds 1800K. It could be concluded that for system design, total fuel utilization percent had better be high enough to make oxygen flame temperature of anode exhaust gas lower than 1800K, which make systems environment friendly.
4. Conclusions

In this work, an oxy-combustor, which could be regard as a key technology for CO\(_2\) capturing, for a 10 kW IGFC system anode exhaust gas is designed aiming at gaining high CO\(_2\) concentration. It is studied through experiment, CFD and CRN simulation method under different system processing conditions, which affects components in exhaust gas. The simulation method could be verified by experiment data. Then some key indicators of the combustor including flame stability, CO conversion, and liner temperature and pollutant emissions are discussed. Results might be meaningful for long term IGFC with CO\(_2\) capturing system design. Main conclusions are as follow:

Anode exhaust gas should be condensed before oxy-combustor to remove some water. Moisture content is lower under lower condensation temperature. Steam could not be wiped out completely which make T\(_{ad}\) always lower than it without vapor. For cycling1 case which contains least H\(_2\), T\(_{ad}\) under dry condition is around 1700K. Considering heat loss, 315K might be an ideal condensation temperature for system process design where T\(_{ad}\) is about 1680K.

CO\(_2\) mole fraction after burning under dry condition is higher than 0.958 with excess O\(_2\) less than 5%. Convective coefficient seems to show slight effects on CO conversion. It is confirmed that CO could be almost fully converted under flame burning condition and unreactive O\(_2\) existing decreases the CO\(_2\) mole fraction. For practical device, excess O\(_2\) helps CO being burned out. 5% excess O\(_2\) could be recommended since an acceptable CO\(_2\) capture rate is gained. The liner convection could be enhanced as far as possible for it shows little influence on CO conversion.

For an oxy-combustor aiming at CO\(_2\) capturing, total flame temperature range relatively small. Liner temperature is influenced by heat transfer inside and outside. Force convection outside flame chamber is an effective way to protect the combustor. Max liner temperature has an optimal value at tangential angle around 25°. When jet tangential angle is smaller, high level of unmixedness between fuel and oxygen leads to high temperature. While jet tangential angle is larger, centrifugal force makes high temperature gas scour liner more fiercely. It can be inferred that an optimal tangential angle, which could vary with dimensions of combustor, exists for liner temperature controlling.

Both CO and NO\(_x\) emissions decrease with rising of excess O\(_2\) percent. When excess O\(_2\) percent is low, more NO\(_x\) releases due to high flame temperature. While excess O\(_2\) supply makes CO burning more completely. If CO\(_2\) concentration could meet the requirement of CO\(_2\) capturing, O\(_2\) should be supplied as more as possible. Anode cycling design seems to have a better performance on CO emission reducing in theory compared with single pass design for no extra steam is needed and less H\(_2\) exists in exhaust gas, though further experiments are needed. For system design, total fuel utilization percent had better be high enough to make oxygen flame temperature of anode exhaust gas lower than 1800K, which make systems environment friendly.
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**Figures**

**Figure 1**

10kW IGFC system flow diagram

**Figure 2**

Exhaust gas oxy-combustor for 10kW IGFC system (1. flame chamber 2. flamestabilizer 3. oxygen swirler 4. fuel swirler 5. connector 6. fuel pipe 7. oxygen pipe 8. base plate 9-10. cooling air chamber 11-13. sealing material)
Figure 3

Exhaust gas combustion test rig

Figure 4

Chemical reactor network model for the combustor

Figure 5

Comparison between calculation and experiments.
Figure 6

Equivalent adiabatic flame temperature of exhaust gas under different condensation temperature.

Figure 7

CO2 mole fraction in dry exhaust gas after burning by CFD calculation.

Figure 8

Maximum temperature of combustor liner under different condition by CFD calculation.
Figure 9

Computed pollutant emissions of Cycling1 by CRN model.
Figure 10

Computed pollutant emissions of Single2 by CRN model.