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 environmental 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.

Introduction

With consumption of non-renewable resource increasing, higher efficiency of power system is required. Fossil fuel combustion causes greenhouse effects. Integrated gasification fuel cell (IGFC) (Al-Khori et al. 2020) provides a new technology route power system. Compared to traditional coal utilization power generation systems, it has higher efficiency in theory since less power converting steps are needed. For IGFC, solid oxide fuel cell (SOFC) could be suitable as it has wider fuel flexibility and could be directly fed by syngas through WGS pretreatment process (Wu et al. 2020). Moreover, coupled with some capturing process, a nearly zero greenhouse gas of CO₂ releasing system could be gained through IGFC system based on SOFC (Thallam Thattai et al. 2017). Carbon element in anode syngas should be gathered to gain a CO₂ high capture rate. SOFC with high fuel utilization percent (Pirasaci, 2019) accomplishes CO₂ inner-gathering. Besides, CO₂ could be gathered by specific system design. In some SOFC systems, anode looping through ejectors is taken into consideration to increase fuel utilization, which would certainly increase exhaust CO₂ density (Wang et al. 2020). Different processing design may lead to different exhaust gas component since anode looping gas has provided certain amount of steam for WGS already.

However, there may still exist about 10%-15% combustible component mainly consisting CO and H₂ in exhaust gas with calorific value about 2-3 MJ/Nm³. Thus, further measures are needed not only for CO₂ capturing efficiency but also for waste heat utilization and environment requirements. Generally, exhaust gas needs to be completely oxidized to CO₂ and H₂O. Spallina et al. proposed chemical looping combustion to deal with exhaust gas. Anode off-gas was imported to a fuel reactor and looping in it through ejectors. Off-gas reacted with the cathode inlet air through oxidizer carrier. The inlet air was heated up to 730°C before entering anode by the exothermic oxidation reaction (Spallina et al. 2018). However, application of ejectors increased the difficulty of system controlling.

Moreover, oxy-combustion has been proved to be an effective method for CO₂ concentrating in some traditional power system (Francisco et al. 2016). Compared to air combustion, no extra impurity like N₂ is imported to the production, which is very suitable for CO₂ capturing. As for IGFC, pure O₂ could be easily got from air separators of coal gasification process, which made oxy-combustion convenient to accomplish. To realize the exhaust gas oxy-combustion, a detailed combustor structure and exhaust gas dealing process design are needed.

Catalytic oxy-combustion is often used to deal with CO pollutant in power system exhaust gas. Okonkwo et al. shown that over 90% CO in coal oxy-combustion exhaust gas could convert to CO₂ through catalyst oxygen tri-reforming (Okonkwo et al. 2020). However, certain ratio of fresh CH₄ needs to be involved for tri-reforming reactions. Chen et al. showed that TiO₂-CuO was an effective catalyst for exhaust gas with 1.5% CO. If specific surface area was sufficient, the conversion could be over 90% in 7 hours (Chen et al. 2019). Han et al. It is shown that nickel oxide catalysts could deal with exhaust gas with 1% CO by vol at different temperature. The flow velocity was 10 ml/min and space velocity was 60000 ml/hg_{cat}. The characteristic temperature was up to 600°C and no deactivation of catalysts for CO oxidation for 720 min (Han et al. 2016). Dey et al. proposed a detailed catalytic combustor for CO when testing Pt catalytic activity, which had a representative porous catalyst bed to increase specific surface (Dey et al. 2020). For practical engineering system, fresh fuel supplement would decrease heat efficiency. Most catalyst could deal exhaust gas with extremely low CO density. However, SOFC exhaust gas contains far more CO and H₂. This may lead to high local combustion temperature which is harmful to catalyst activity. Sufficient specific surface area is required for catalytic combustion which made burner difficult to enlarge. Besides, porous catalyst bed increased back pressure of the system, which was to the disadvantage of fuel cells. Therefore, it might be unsuitable to apply catalytic oxy-combustion into practical system.

Direct pure oxygen flame burning shows another way to deal with the anode exhaust gas, which could almost fully convert CO to CO₂. The main characters of exhaust gas were extremely high level moisture content and low calorific value. These characters may respectively challenge the flame stability and ignition. Ning et al. reported that water vapor and carbon dioxide increased the ignition temperature of H₂/O₂ flame, which both steam and carbon dioxide show some negative effects on it (Ning et al. 2018). It means that a cooling dewatering process of anode off-gas should be carried out before flame combustion. Recent researches proved that pure oxygen flame could be steady under extremely low equivalent ratio especially for H₂ enriched fuel, which means it might have good performance for low-calorific exhaust gas. Rashwan et al. showed that oxygen partially premixed flame of natural gas could be steady in equivalent ratio range from 0.7-1.1 (Rashwan et al. 2017). Imteyaz et al. showed similar results on H₂ enriched methane oxygen premixed combustion. Besides, it was shown that H₂ could decrease the lower limited equivalent ratio down to 0.2 (Imteyaz et al. 2018). Li et al. gained a stable oxy-methane premixed flame with O₂ mole fraction from 0.25 to 1.0. And stability increased when O₂ mole fraction increased (Li et al. 2018). Too much excessive O₂ would decrease CO₂ concentration in production although it improves flame stability. Ilbas et al. studied oxy-combustion of low-calorific

syngas numerically and experimentally. It is indicated that oxygen flame could almost burn out all the CO content even in low-calorific syngas even in low calorific gas. But flame burning may cause problems on NO_x emissions. Pollutant emission was related to flame temperature and burner design (Ilbas et al. 2019). Pollutant should be taken into consideration during system design because acidic gas may be harmful to systems and environment.

According to the above, for integrated system design, condensation process of anode off-gas would surely lead to heat loss. This makes it meaningful to discuss a proper condensation temperature for exhaust gas oxy-combustion to avoid undercooling. A reliable burner, which could reach a satisfactory CO converting rate, is needed. Amount of excessive O₂ is worth to study to balance the flame stability, converting rate and CO₂ concentration. Exhaust gas should be burnt around equivalent ratio which leads to high local flame temperature. For long-life systems, it is important control thermal creep deformation of combustor liner which is influenced by the inner jet flow and cooling strength. Differing from air combustor, oxidizer gas could not be directly used as liner cooling medium through method such as film cooling (Topal et al. 2019) since quantity of O₂ for exhaust gas burning is rather small. Therefore, liner forced cooling should be involved. It decreases flame temperature and then CO converting rate. And drivers of forced cooling will increase inner consumption of the energy system. Moreover, high reacting temperature of oxy-combustion leads to high level NO_x emission possibility since N₂ still exists both in industry syngas (Schluckner et al. 2020) and oxygen (Khallaghi et al. 2020).

In this paper, an oxy-combustor for a 10kW IGFC system anode exhaust gas is experimentally and numerically studied. CFD and chemical reactor network (CRN) method are verified and used to simulate the flame and emissions of combustor. Flame ignition of exhaust gas and pure oxygen under different condensation temperature is discussed. Then effects of excessive O₂ and liner cooling on CO conversion rate are investigated. And influences of jet flow and outside forced cooling operating conditions on liner temperature are analyzed. Pollutant emissions of different anode processing under different excess O₂ amount are also studied. The results could be very helpful for the integrated IGFC coupled with CO₂ capturing system design.

Experimental Setup

As mentioned above, the exhaust gas components vary with the system design. Usually, fuel cells may not utilize CO directly. CO will firstly react with H₂O to form CO₂ and H₂ 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 H₂ and H₂O since extra steam is bring in for WSG process before. Fig. 2 shows the exhaust gas oxy-combustor which mainly consists of a flame chamber, a flame stabilizer, oxygen swirler and fuel swirler. Swirlers and stabilizer 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_2 , CO and CO_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.

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). A modified realizable k- ϵ 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. 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 radiant and convective boundary. (Wang et al. 2016) Commercial software is used to solve modeling equations.

The independence verification of grids is shown in Fig.4. Two different series of mesh are used to calculate the flame. In case (a), 200,000 cells are taken for simulation, while 1,000,000 cells for case (b). No notable difference is shown for two different series of mesh. Thus, grids of 200,000 are taken for following calculations for computing resource saving consideration.

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 (Wang et al. 2016) and used (Shao et al. 2017) in previous reports. The model is shown in Fig.5. PSRs upstream are used to simulate the fuel/oxygen unmixedness in primary zone. Reactors downstream simulate the developing zone. The resident time was determined by the flow velocity. Mass transferring among the reactors was decided by

element conservation. 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). As shown, model B in Fig.5 (b) has more reactors to simulate each flame zones which mean the model is more detailed compared to model A in Fig.5 (a). The CRN is solved in the commercial software. CO emission simulation results of experiment cases are shown in Fig.6 compared with experiment data. Agreements between calculation and data are indicated which could prove reliability of the model. NO notable difference was shown between the results of model A and B. In following studies, model A was adopt to save some computing resource and species components of Cycling1 case are taken into consideration as examples unless special declaration is made.

Results And Discussion

4.1. Exhaust gas condensation temperature effects on ignition

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_2O 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. The condensation temperature T_{con} could be considered as the inlet temperature of combustor. 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 1s. Results are shown in Fig. 7.

It is shown in Fig. 7 that the equivalent adiabatic flame temperature T_{ad} decreases with condensation temperature increasing. It is because moisture content is low under low condensation temperature. If the condensation temperature is higher than 350K, flame temperature is lower than ignition temperature which means too much 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 T_{ad} 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 cooling convective coefficient conditions. Simulation results are shown in Fig. 8. Besides, data of experiment above is shown in Table 2.

It shows in Fig.8 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. Cooling 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 CO_2 content decreases when excess O_2 quantity rises. This confirms that CO could be almost fully converted under flame burning condition. Excess O_2 decreases the CO_2 mole fraction. For practical device, excess O_2 helps CO being burned out which reduces CO emission in consideration of unexpected unmixedness at jet outlets. Therefore, 5% excess O_2 could be recommended since a acceptable CO_2 capture rate is gained. The liner forced cooling strength shows little influence on CO conversion. Table 2 shows that CO_2 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 H_2 and H_2O content. They would be dried out before capture process. It is experimentally proved that CO can be converted to CO_2 efficiently by fully oxy-combustion.

4.3. Liner temperature

Liner temperature of combustor might be another key indicator for oxy-combustor evaluating. Mostly, the liner temperature is strongly influenced by flame temperature distribution. For an oxy-combustor aiming at CO_2 capturing, equivalent ratio might varies in a narrow range. It makes flame temperature range relatively small especially for diffusion flame. Thus, flame temperature distribution is mainly determined by jet form. As mentioned, forced air cooling should gets involved outside the liner. In that case, liner temperature is influenced by heat transfer and jet flow form. 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 mass and heat transfer inside liner. Because fuel flow rate could be several times more than 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.9.

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. Generally speaking, O_2 production from cryogenic air separation unit (ASU) may usually contain N_2 up to 5% by vol. Therefore, in practical IGFC system, some N_2 always exists in syngas even though O_2 gasification technology might be involved. So combustor pollutant is mainly consisting of CO and NO_x . 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% N_2 by volume in simulation case. Ratio of other component is kept as before. Emissions under different excess O_2 quantity are computed. Comparison of combustion with pure O_2 and industrial O_2 containing 5% N_2 was applied to figure out the oxygen purity impacts on emissions. Results are shown in Fig. 10 and Fig. 11.

It is shown that both CO and NO_x emissions decrease with rising of excess O_2 percent. It is known that NO_x formation is directly related to flame temperature. When excess O_2 percent is low, local flame temperature could be higher which leads to higher NO_x emissions. On the other side, more excess O_2 supply makes CO burnt more completely which means lower CO emissions. Therefore, excess O_2 supply is in favor of pollutant reducing. Combined with mentioned, if CO_2 concentration could meet the requirement of CO_2 capturing, O_2 should be supplied as more as possible. Barely influence of excess O_2 percent is shown on combustor exit temperature since O_2 flow rate varies slightly. Single2 case releases more CO and NO_x 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 NO_x emission. Besides, Single2 case contains more H_2 so that H_2O mole fraction is high after burning. Water vapor prevents CO converting to CO_2 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 H_2 , though more experiments are needed further. As known, NO_x 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. NO_x emissions increase when oxygen purity decreases. And CO emissions showed slightly decline with oxygen purity decreasing. But the impacts of oxygen purity on NO_x and CO are not notable. This is mainly because flow rate of oxygen is relatively small compared to that of exhaust gas. Thus, N_2 importing with oxygen flow is rather small and could not influence total N_2 concentration obviously. As N_2 does not participate in CO oxidizing reactions, rarely difference is shown under different O_2 purity conditions.

Conclusions

In this work, an oxy-combustor is studied through experiment, CFD and CRN simulation method under different system processing conditions. The simulation method could be verified by experiment data. Then some key indicators of the combustor including exhaust gas condensation temperature impacts on ignition, CO conversion, and liner temperature and pollutant emissions are discussed. Main conclusions are as follow:

Anode exhaust gas should be condensed before oxy-combustor to remove some water. For cycling1 case which contains least H_2 , T_{ad} under dry condition is around 1700K. Considering heat loss in practical system, 315K might be an ideal condensation temperature for system process design where T_{ad} is about 1680K. It would be a key index for the IGFC with CO_2 capturing integrated system process designing.

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. 5% excess O_2 could be recommended since an acceptable CO_2 capture rate is gained. The liner forced cooling strength shows little influence on CO conversion. It is proved that direct flame burning is an effective and reliable way to accomplish CO_2 concentrating for IGFC system. Oxy-combustor could be used in industrial systems.

For an oxy-combustor aiming at CO_2 capturing, total flame temperature range relatively small. Liner temperature is influenced by heat transfer outside and jet form. Force convection outside flame chamber is an effective way to protect the combustor. 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. Max liner temperature has an optimal value at jet tangential angle around 25° . These conclusions might be of guiding significance for combustor design.

Both CO and NO_x emissions decrease with rising of excess O₂ percent. When excess O₂ percent is low, more NO_x releases due to high flame temperature. While excess O₂ supply makes CO burning more completely. If CO₂ concentration could meet the requirement of CO₂ capturing, O₂ should be supplied as more as possible. Purity of O₂ barely influenced the pollutant emissions. 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 H₂ 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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Tables

Table 1 Operating conditions of experiments for the exhaust gas combustor

	Flow rate (kg/h)	Total Fuel Utilization (%)	O ₂ Flow rate (kg/h)	Exhaust Gas Mole Fraction			
				CO	H ₂	H ₂ O	CO ₂
Single1	3.48	86	0.47	0.069	0.197	0.061	0.673
Single2	3.48	80	0.69	0.091	0.261	0.061	0.587
Cycling1	3.45	86	0.45	0.101	0.169	0.061	0.669

Table 2 Experiment data of CO₂ mole fraction in dry exhaust gas after burning.

Case	CO ₂ mole fraction
Single1	0.962
Single2	0.962
Cycling1	0.958

Figures

Figure 1

10kW IGFC system flow diagram

Figure 2

Exhaust gas oxy-combustor for 10kW IGFC system (1. flame chamber 2. flamestabilizator 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

Figure 4

Temperature distribution through CFD under different mesh density ((a) for 200,000 cells; (b) for 1,000,000 cells)

(a) model A

(b) model B

Figure 5

Chemical reactor network model for the combustor.

Figure 6

Comparison between calculation and experiments.

Figure 7

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

Figure 8

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

Figure 9

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

(a) NO_x emissions

(b) CO emissions

Figure 10

Computed pollutant emissions of Cycling1 by CRN

(b) CO emissions

Figure 11

Computed pollutant emissions of Single2 by CRN model.