A REVIEW ON FUNDAMENTAL RESEARCH OF OXY-COAL COMBUSTION TECHNOLOGY

by

Shiquan SHAN[a*]***, Binghong CHEN***[b]***, Zhijun ZHOU***[a]***, and Yanwei ZHANG***[a]

[a] State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou, China
[b] School of Energy and Power Engineering, University of Shanghai for Science and Technology, Shanghai, China

Review paper
https://doi.org/10.2298/TSCI210329238S

Oxy-fuel combustion is a key technology to realize CO_2 capture and storage in coal-fired power boilers in the future. In recent years, there have been extensive studies on it. This review summarizes some key results of fundamental research on oxy-coal combustion. By comparing with traditional coal-fired boiler with air combustion in power station, the typical characteristics of oxy-coal combustion are introduced from four aspects: combustion, heat transfer, pollutant emission, and numerical simulation; so that readers have a relatively comprehensive understanding of fundamental research on oxy-fuel combustion. Furthermore, some scientific issues in future research are summarized. The paper is both academic and popular, providing basic knowledge and academic direction inspiration for scholars or graduate students who are about to engage in oxy-fuel combustion research.

Key words: *oxy-coal combustion, coal reactivity, heat transfer, pollutant emission, numerical simulation*

Introduction

Greenhouse effect is a significant environmental problem in 21st century. The CO_2 greenhouse gas and other pollutant caused by coal-fired boilers in power plants are a major reason. [1]. There are many CO_2 emission control technologies for coal-fired power plants, where oxy-fuel combustion is a promising one. Proposed first by Abraham *et al.* [2], oxy-coal combustion uses pure oxygen rather than air to support coal combustion in boiler. Since N_2 is almost removed, the flue gas produced in oxy-fuel condition has a high CO_2 concentration more than 90%, and a part of it is recycled into the furnace to suppress excessive flame temperature. Furthermore, the remaining flue gas is cooled, compressed and separated for CO_2 capture and storage. Oxy-fuel combustion is suitable for both construction of new power stations and reform of old coal-fired boilers. Figure 1 shows its schematic [3]. In addition to the advantage of CO_2 reduction, oxy-fuel boiler also has dramatic energy-saving effects, which can promote complete combustion, reduce boiler exhaust loss, and improve boiler efficiency. Thus, oxy-coal combustion is crucial for developing countries in the reform of coal-fired power stations.

Using CO_2 instead of N_2 as the dilution gas makes oxy-fuel combustion significantly different from air-fuel combustion in flame temperature, heat transfer, reaction kinetics, and pollutant emissions. This is caused by the different properties of CO_2 and N_2:

* Corresponding author, e-mails: shiquan1204@163.com; shiquan1204@zju.edu.cn

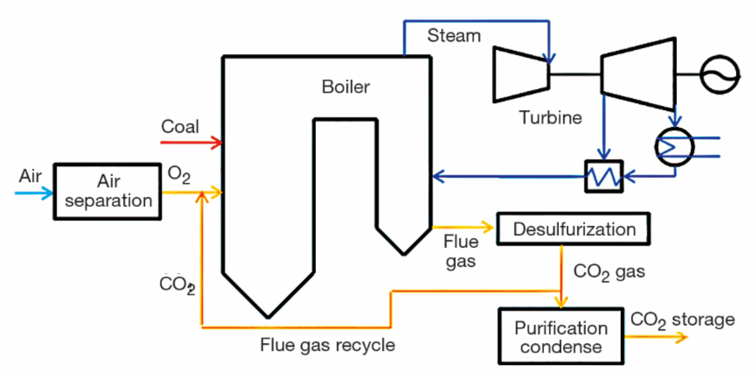

Figure 1. Schematic of oxy-coal combustion technology

– The CO_2 has greater density.
– The CO_2 has a larger specific heat than N_2.
– The O_2 diffusion rate is smaller in CO_2 atmosphere, which is only 0.8 times the size of N_2.
– Oxy-fuel combustion generates much triatomic gases CO_2 and H_2O with high emissivity.

As a result, oxy-fuel combustion requires about 30% oxygen ratio to achieve a similar flame temperature as air-fuel combustion. In past decades, there have been many studies and some complicated reviews on oxy-fuel combustion. However, there is not a short review which summarizes classical results and makes readers quickly know the fundamental and research topic of oxy-fuel combustion. Therefore, in this paper, key results of fundamental researches on oxy-coal combustion are selected and organized as a mini review so that readers could have a relatively comprehensive understanding of the oxy-coal combustion within half an hour. Both basic knowledge and academic direction inspiration are provided for scholars or graduate students who are about to engage in oxy-fuel combustion research.

Combustion characteristic

Ignition and flame propagation

Ignition and flame propagation are the fundamental combustion characteristics. Based on self-built Hencken burner in Sandia Laboratories, Molina and Shaddix [4] studied the ignition characteristics of pulverized coal combustion in oxy-fuel condition and found that the higher CO_2 concentration and lower oxygen concentration increase the ignition delay time. This is because CO_2 has a higher molar specific heat and the oxygen concentration could affect the reaction activity of the mixture. The particles ignition and volatilization separation in 30% O_2/CO_2 atmosphere are similar to those in air-fuel condition. In addition to the high specific heat of CO_2, Glarborg and Bentzen [5] proposed that the competition of free radical in the reaction $CO_2 + H \rightleftharpoons CO + OH$ is also a reason. This reaction will suppress the combination of O_2 and H free radicals, and then causes the decrease in burning rate [6]. Moreover, Japan's IHI [7] studied the flame propagation speed of coal combustion on a self-built microgravity measurement device and found that the flame propagation is the slowest in oxy-fuel atmosphere. Moron' *et al.* [8] found that the temperature of laminar flame in oxy-coal combustion is about 1-15 K lower than that of air-coal combustion. These are mainly due to the high specific heat of CO_2. In conclusion, the influence of oxy-fuel atmosphere on coal ignition and flame characteristics is mainly caused by the high specific heat of CO_2 and free radical competition.

Coal reactivity

The influence of oxy-fuel atmosphere on the pulverized coal combustion also lies in the coal reactivity. It mainly affects the de-volatilization and the coal char combustion. A high CO_2 concentration delays the de-volatilization process [9], but this does not defer the complete combustion time of volatiles. Rathnm *et al.* [10] found that the amount of de-volatilization in O_2/CO_2 atmosphere is higher than that in O_2/N_2 atmosphere. This is mainly due to the CO_2-char gasification reaction. Besides, the pyrolysis components are less affected by CO_2. Thus the de-volatilization amount of pulverized coal under O_2/N_2 atmosphere should be close to the real volatile content.

Char combustion after de-volatilization is the second process of coal combustion, mainly affected by temperature and oxygen concentration. The influence of temperature on the reactivity of coal char combustion is generalized in fig. 2. Actually, it is also affected by various factors such as particle size and minerals. Coal char combustion is in the reaction-control zone at low temperature and the reaction rates of oxy-fuel combustion and air-fuel combustion are similar. As the temperature rises, coal char combustion enters the diffusion-reaction-control zone, where the reaction rate of oxy-fuel combustion exceeds that of air-fuel combustion since the effect of the CO_2 gasification reaction. The general pulverized coal boiler should be in this range. Moreover, the coal char combustion enters the diffusion-control zone when the temperature is very high, where the diffusion of O_2 is significantly affected by CO_2 and the oxidation reaction of coal char is restricted, thus, the reaction rate of oxy-fuel combustion is not as good as air-fuel combustion. In oxy-coal combustion, the reactivity of coal char combustion increases with the oxygen concentration, and the required temperature also decreases. At the combustion temperature in boiler, the coal char reactivity of oxy-fuel combustion is better than that of air-fuel combustion. For this phenomenon, Saastamoinen [11] believes although the coal char combustion reaction is suppressed by the decrease of O_2 diffusion rate under the CO_2 atmosphere, the CO_2-char gasification reaction rate still increases since the pulverized coal is in the diffusion-reaction control zone at the boiler temperature. Thus, more CO_2 reaches char surface and enters the particles to promote gasification reaction, and the overall reactivity is enhanced.

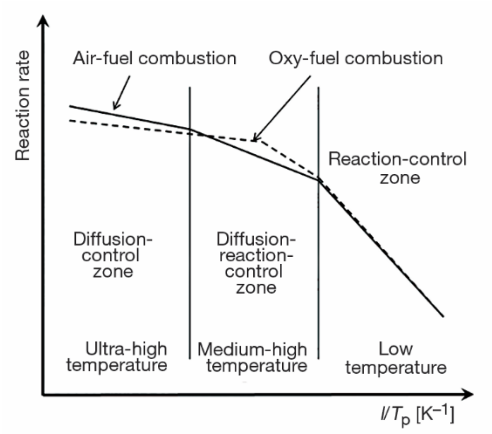

Figure 2. Effect of temperature on the reaction rate of coal char

For the burn-out characteristics of coal char, Wall *et al.* [12] studied three different Australian coal under the oxygen concentration range of 2-30% both in air-fuel and oxy-fuel atmosphere. It is found that increasing the oxygen concentration can elevate the burnout rate of all coals, and the burnout rate under oxy-fuel atmosphere is higher than that under air-fuel atmosphere at any oxygen concentration. These results are caused by CO_2-char gasification reaction. In addition, some researchers [13, 14] calculated the conversion of single-particle coal in oxy-fuel combustion and believed that CO_2 gasification reaction would reduce the particles temperature. However, their results also indicate that the presence of gasification leads to a decrease in the burnout time of particles and an increase in the conversion rate of coal char. As a conclusion, the influence of CO_2 gasification reaction on coal char burnout cannot be ignored under oxy-fuel combustion at high temperature.

Heat transfer characteristics

It mainly considers radiation heat transfer and convection heat transfer in the furnace, which depends on the flame temperature, radiative characteristics of gases and particles, emissivity of furnace wall, and the aerodynamic field of the flame. It is believed that in an oxy-fuel atmosphere, the low kinematic viscosity of CO_2 leads to a large convective heat transfer coefficient at the same flow rate [15]. Toporov [16] further verified that the convective heat transfer coefficient in oxy-fuel condition is slightly higher than that in air-fuel condition and it becomes higher as the recycle rate of flue gas increases. Therefore, oxy-fuel combustion could reduce the heating surface of the boiler, and thereby saving materials [17]. Due to the difference in heat transfer, the heat flux distribution in oxy-fuel boiler is different from that in air-fuel boiler. Wall *et al.* [12] numerically studied a 30 MW boiler and found that the wall heat flux on the burner side is greater for air-fuel combustion, while the heat flux on the boiler back wall is greater for oxy-fuel combustion. This is because the high CO_2 specific heat delays the ignition which makes flame length increase. In order to reach a same adiabatic flame temperature as air-fuel combustion, the required oxy concentrations are 35% and 28%, respectively for the dry flue gas recycle and wet flue gas recycle based on the analysis of oxy-fuel combustion of bituminous coal [12]. It is believed that there are similar heat transfer characteristics for oxy-fuel and air-fuel combustion when their temperature fields are similar in furnace. Therefore, the existing boilers can be modified by adjusting the oxygen concentration and recycled flue gas [15]. Radiation heat transfer is predominant in coal-fired boilers. Relying participating media such as triatomic gases (CO_2, H_2O) and fine particles (soot and fly ash). Nicely, oxy-fuel combustion can generate more CO_2 and H_2O gases, and meanwhile, the volume of flue gas decreases and the concentration of fine particles increases due to flue gas recycle. Thus, radiation heat transfer is higher in oxy-fuel combustion. Section *Radiant model* will introduce the oxy-fuel gas radiation model in detail.

There are also some classical experiments on radiation heat transfer. The IHI in Japan built a 1.2 MW pilot plant to study the oxy-fuel combustion of pulverized coal [18]. High-volatile bituminous coal was used in both O_2/CO_2 and O_2/N_2 atmospheres, and a heat exchange tube was set in the furnace to simulate the real radiation transfer surface in boiler. It appears that the radiative heat transfer in 27% O_2/CO_2 atmosphere with wet flue gas recycle is similar to that in air-fuel atmosphere. Andersson *et al.* [19] and Hjartstam *et al.* [20] in Chalmers University of Technology built a 100 kW oxy-fuel combustion test platform with flue gas recycle and studied radiation characteristics of lignite combustion. In the experiment, the flue gas recycle rate was adjusted to increase the oxygen concentration from 25% to 29%. It shows that both flame temperature and total radiation intensity increase, and the ratio of gas radiation to total radiation in oxy-coal combustion is slightly higher than that in air-coal combustion. These results indicate that most radiation in lignite flame is emitted by flame particles. The authors believe that the increase in gas radiation caused by higher CO_2 proportion is not obvious in coal combustion. Moreover, as long as there are similar temperature fields, the total radiation intensities are similar regardless of oxy-coal or air-coal combustions.

Pollutants emission

Oxy-coal combustion could produce environmental pollutants such as NO_x, SO_x, and solid particulate. The circulation of NO_x and SO_x in oxy-fuel flue gas causes corrosion on boiler and furnace, which seriously threatens the boiler safe running. In addition, it also has an impact on the energy consumption and safety of CO_2 compression and separation process.

Nitrogen oxides

Coal combustion mainly generates NO. The NO is converted to NO_2 at low temperature and N_2O may also be produced at a combustion temperature of the fluid-bed boiler. The formation mechanism of NO_x in pulverized coal combustion is shown in fig. 3. The NO_x formation in coal combustion is divided into three types: thermal NO_x formation and prompt NO_x formation from N_2, and fuel-based NO_x formation from fuel-N.

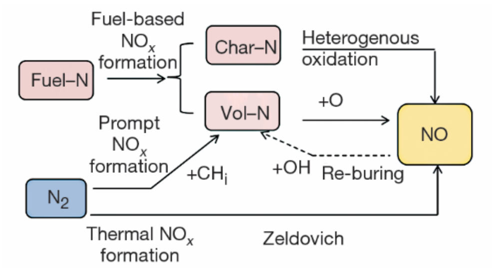

Figure 3. The NO_x formation mechanism in pulverized coal combustion

The NO_x emission in oxy-coal combustion is lower than that in air-coal combustion, which only accounts for about 1/3 of that in air-fuel atmosphere. The primary reason for the reduction of NO_x lies in the isolation of nitrogen in the atmosphere, which reduces the generation of thermal NO_x, and the re-burning of NO in the fuel-rich zone in oxy-fuel combustion. Under oxy-fuel conditions, the increase of CO_2 ratio decreases the flame temperature and NO production dramatically, causing the mechanism of NO production is transformed from the thermal to prompt route [21]. Moreover, the high CO_2 proportion also contributes to the reaction of $CO_2 + H \rightarrow CO + OH$, which promotes the re-burning of NO due to much produced OH [22].

Methods to control NO_x emission in oxy-coal combustion are also significant. The partial pressure of oxygen is an important factor affecting NO_x formation. It have been pointed out that oxygen partial pressure has a first-order increasing relationship with NO_x production during oxy-coal combustion [23], which has guiding meaning for the development of low-NO_x combustion technology. Developing burner is an important NO_x control technology. Tan *et al.* [24] conducted a series of technical tests of oxy-coal combustion and believed that the formation of NO_x could be reduced under the premise of ensuring a rational burnout by optimizing the burner design and organizing oxygen combustion. In addition, the staged combustion technology can also reduce NO_x in oxy-fuel combustion [25]. The reason lies in that staged oxy-fuel combustion can increase the de-volatilization rate, thereby increasing the reduction of NO in fuel-rich zone, and the reduction of flue gas dilution in the staged oxy-fuel combustion results in a longer residence time of NO in the reduction zone.

Sulfur oxides

Sulfur emissions in coal combustion process will cause low temperature corrosion in the economizer and tail heating surface of boiler and also affect the fly ash characteristics. Sulfur emission is directly proportional to the sulfur content of coal. Flue gas recycle makes SO_x concentration in oxy-fuel combustion furnace much higher than that in air-fuel combustion. However, most studies show that the SO_x emission rate is still lower than that in air-fuel combustion. The experimental results of oxy-fuel combustion in IHI 1.2 MW test platform with dry flue gas recycle show that the sulfur conversion rate in coal is significantly lower than that of air-fuel combustion, and SO_2 emissions decrease by 20-30% [7]. Wall *et al.* [12] conducted experiments on three coals and found that the SO_2 emission is only about 2/3 of that in air-fuel combustion. This is because the condensation deposit of sulfate in the pipe and more sulfur is absorbed by the ash in oxy-fuel combustion with flue gas recycle.

As shown in fig. 4 [26], increasing the oxygen ratio will elevate the SO_2 emission rate in oxy-fuel combustion. The fuel equivalent ratio, φ, also has a greater impact on SO_2. There is a SO_2 emission peak at φ about 1.2. This is because S remains in the unburned carbon or is converted into other reduced sulfur compounds (H_2S, COS, CS_2, *etc.*) in the fuel-rich zone where φ is greater than 1.2. In addition, S may be converted to SO_3 in the fuel-lean zone where φ is less than 1.2. Tan *et al.* [24] found that SO_3 conversion rate in oxy-coal combustion (5%) is higher than that in air-coal combustion (1-5%). Although most scholars believe that oxy-fuel combustion is beneficial to reduce the emission rate of SO_2, the mechanism of the sulfur conversion to other sulfides remains to be further studied. Flue gas recycle could increase the concentration of SO_2 and SO_3 in the furnace, and oxy-fuel combustion will also increase the acid dew point, thereby accelerating low-temperature corrosion. Therefore, oxy-fuel combustion should use low-sulfur coal and take desulfurization before flue gas recycle.

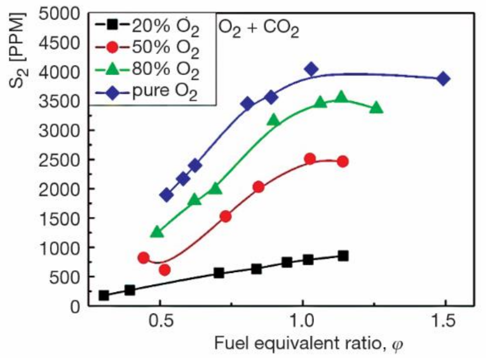

Figure 4. The influence of oxygen concentration and fuel equivalent ratio on SO_2 emission [26]

Fly ash and soot

Minerals conversion in coal is affected by combustion temperature and atmosphere [27]. The differences of atmospheres and temperatures between oxy-fuel and air-fuel combustions will affect the formation mechanism and the composition of fly ash. Compared with air-fuel combustion, oxy-fuel combustion with the same oxygen concentration reduces the adiabatic flame temperature and suppresses the combustion rate. Thus, the volatilization of volatile metals and the secondary oxides of non-volatile metals slows down and the formation rate of fly ash is also reduced [28, 29]. Increasing the oxygen concentration can reduce the difference between oxy-fuel and air-fuel atmospheres, because adjusting oxygen concentration can change combustion temperature and coal char distribution in different phases [27-29]. In Suriyawong's [30] experiment, the variation of oxygen concentration from 20% to 50% in oxy-coal combustion makes the temperature increase by 907 K and results in more fly ash with large particles. In addition, Wall *et al.* [12] pointed out that the fly ash concentration in the flue gas of oxy-coal combustion is 1.5 times greater than that of air-coal combustion due to the reduction of flue gas volume. Moreover, there will be more deposition at the bottom of oxy-fuel combustion boiler due to low flue gas flow rate. This is conducive to the subsequent ash removal work. The content of sulfur in fly ash of oxy-coal combustion is higher than that of air-coal combustion. This also verifies that oxy-fuel combustion can suppress the conversion of S in coal from solid to gas phase.

There are fewer studies on soot formation in oxy-fuel combustion. Guo *et al.* [31] pointed out that O_2/CO_2 atmosphere has an inhibitory effect on the soot formation because the high specific heat of CO_2 causes a decrease of the flame temperature. Saanum *et al.* [32] believe that the amount of soot produced by oxy-fuel combustion is also smaller than that by air-fuel combustion. The main reason is that CO_2 reacts with H free radicals to generate OH radicals, thereby promoting the oxidation of soot precursors. Morris *et al.* [33] found that bituminous coal produces less soot under oxy-fuel conditions than in air-fuel condition. Therefore, it appears that the soot production in oxy-coal combustion is generally reduced.

Trace elements

Trace elements have a content of less than 100 μg/g in coal [34], and the content is related to the coal types. Trace elements generally include mercury, lead, selenium, molybdenum, vanadium, manganese, zinc, copper, and other elements. For oxy-coal combustion, Zheng *et al.* [35] found that the emissions of mercury, chromium, arsenic, and selenium are not affected by oxy-fuel atmosphere through equilibrium calculation. Wall *et al.* [12] found that the mercury content in the ash increases in oxy-coal combustion, but the mercury could not be leachable and its form also could not be determined. Boron element exists in fly ash and flue gas, and its contents are not much different between oxy-fuel and air-fuel combustions. However, more boron can be filtered out from ash in oxy-fuel combustion, so it may exist in different forms. Suriyawong *et al.* [36] studied the release characteristics of heavy metals and particles in oxy-coal combustion and found that proportions of elemental mercury and oxidized mercury in oxy-fuel combustion are not much different from that in air-fuel combustion. Zhang *et al.* [37] found that the mass content of most trace elements in particles are basically the same in particle size range of PM_{10}, but the content of selenium in size range of PM_{10} under oxy-coal combustion with flue gas recycle is significantly higher than that in air-coal combustion. At present, there is no systematic research report on trace element emissions in oxy-fuel combustion, so there are few references and relevant research needs to be further carried out.

Numerical models

Numerical simulation can quickly, accurately and cost-effectively analyze the flow, heat transfer, and reaction process in combustion. It is widely used in fuel combustion studies. For oxy-fuel combustion, some general models of air-fuel combustion can be used. In addition, scholars have established different radiation and reaction sub-models to adapt to oxy-fuel conditions.

Turbulence model

Industrial combustion processes are mostly in turbulent condition. In theory, all turbulence can be solved by direct numerical simulation, that is, solving the Navier-Stokes equation directly, but this requires huge computer capacity, so it is mostly solved by turbulence models in practical engineering. The main models are based on RANS and large eddy simulation (LES). Turbulence models for coal combustion reactions are often based on RANS. In oxy-fuel combustion, scholars also mostly use the two-equation k-ε model based on RANS to simulate turbulent combustion as shown in [38-40], and the results are in good agreement with the experiment. Edge *et al.* [41] used both LES and RANS methods to numerically simulate oxy-coal combustion, and found that the prediction results by LES are more accurate in flue gas recycle zone. This is because LES performs direct numerical calculations on turbulent large eddies, resulting in high accuracy.

Radiation model

Radiation heat transfer is obtained by solving the radiation transfer equation (RTE) in CFD simulation. Spectral RTE with emission, absorption and scattering can be expressed [42]:

$$\frac{\mathrm{d}I_\lambda(\mathbf{s})}{\mathrm{d}s} = -(\kappa_{\mathrm{a},\lambda} + \kappa_{\mathrm{s},\lambda})I_\lambda(\mathbf{s}) + \kappa_{\mathrm{a},\lambda}I_{\mathrm{b},\lambda} + \frac{\kappa_{\mathrm{s},\lambda}}{4\pi}\int_{4\pi} I_\lambda(\mathbf{s}')\Phi(\mathbf{s}',\mathbf{s})\mathrm{d}\Omega' \tag{1}$$

where κ_a is the spectral absorption coefficient, κ_s – the scattering coefficient, and Φ – the scattering phase function. The RTE in CFD calculation is generally solved by discrete co-ordinate method [42] since it is easier to combine with various radiative models and has higher calculation speed and accuracy [43].

Due to the large CO_2 proportion in oxy-fuel combustion, the gas radiation characteristics are significantly different from that in air-fuel combustion. In order to obtain accurate gas radiation characteristics under oxy-fuel conditions, the absorption spectra of CO_2 and H_2O as well as the overlap between them must be considered. Because it is quite complicated to directly calculate the absorption coefficient, the weighted-sum-of-gray-gases (WSGG) model is developed for gas radiation characteristics. It has simple principle and high calculation efficiency and also can be combined with various RTE solving methods, leading to widely application [44]. The core idea of WSGG model is to replace the actual non-gray gas with N gray gases, and the total absorption coefficient or radiative heat flux is equal to the weighted sum of each gray gases [45]. The emissivity on the path-length L is expressed by WSGG model:

$$\varepsilon = \sum_{i=1}^{n} a_i[1 - \exp(-\kappa_i XPL)] \tag{2}$$

where X is the mole fraction of the mixed gas, expressed as the sum of the mole fractions of H_2O and CO_2:

$$X = X_{\mathrm{H_2O}} + X_{\mathrm{CO_2}} \tag{3}$$

The absorption coefficient κ_i of each gray gas is constant in a certain spectral region, and a_i represents the weight. The sum of the weights must be one, which also includes the white gas weight a_0, whose absorption coefficient κ_0 is zero; thus, there is:

$$\sum_{i=0}^{n} a_i = 1 \tag{4}$$

The focus of WSGG model development is to determine the absorption coefficient, κ_i, and weight coefficient, a_i, of each gray gas. For the oxy-fuel atmosphere, Yin *et al.* [46] proposed a new WSGG model of four gray gases based on the exponential wide band model. Johansson *et al.* [47] developed a new WSGG model based on the statistical-narrow-band (SNB) model of EW2C database. Dorigon *et al.* [48] further developed an oxy-fuel WSGG model based on line-by-line model of HITEMP2010 database. Recently, Shan *et al.* [49, 50] have further developed a WSGG model of oxy-fuel combustion under pressurized conditions based on SNB model. Centeno *et al.* [51] found that WSGG models have greater impact on the results of temperature field and wall heat flux in oxy-fuel combustion furnace through the numerical simulation. The WSGG model developed by Dorigon has better results, while the WSGG model for traditional air-fuel combustion is not ideal. Recently, their group still used

Dorigon's WSGG model to calculate gas radiation characteristics when analyzing radiation-turbulence interactions in an oxy-fuel atmosphere and idea results were obtained [52]. Besides, Jovanović *et al.* [53] used Yin's WSGG model to simulate a 0.5 MW furnace burner for pulverized coal combustion and found that the numerical temperature fields in both air and oxy-fuel conditions are in good agreement with the experimental results. Zhang *et al.* [54] used different WSGG models developed by Yin and Johansson to perform numerical simulation of a 100 MW oxy-fuel combustion boiler for pulverized coal. In addition to gas radiation, the particles and soot radiation in the furnace also cannot be ignored. Johansson *et al.* [55] found that particle radiation is dominant when studying particle and gas radiation in a cylindrical reactor. Backstrom *et al.* [56] and Yin [57] also gave similar conclusions. Therefore, although gas radiation is enhanced in oxy-fuel combustion, the radiation heat transfer in the furnace is dominated by particle radiation.

Reaction model

Oxy-coal combustion is a heterogeneous reaction. Due to the high CO_2 concentration, it will cause the CO_2 gasification reaction and the O_2 diffusion rate decrease. Considering the diffusivity, Khare *et al.* [58] numerically simulated the oxy-fuel and air-fuel combustions on a vertical furnace of pilot plant. Different mass diffusion-limited rate constants and kinetic-limited rate pre-exponential factors are selected for oxy-fuel combustion and air-fuel combustion based on drop-tube furnace experimental results. It is found that the simulation results are not very consistent with the measured results since the CO_2 gasification reaction is not considered.

It can improve the accuracy of the char consumption rate simulation if CO_2 gasification reaction is considered. Kuhr *et al.* [59] used two models to simulate the pulverized coal combustion in an O_2/CO_2 atmosphere in a vertical reactor without flue gas recycle. One is an inherent model that only considers oxy-char combustion, and the other also considers a Boudouard reaction model (CO_2-char gasification). The results obtained are shown in fig. 5. It appears that the predicted oxygen concentration by the model without gasification is obviously higher, while the model considering the gasification reaction predicts the oxygen concentration very well. Thus, the influence of char gasification on the reaction cannot be ignored under oxy-coal conditions. In addition, the concentration of H_2O in flue gas cannot be ignored under wet recycle conditions, some numerical studies [60, 61] utilized both char-CO_2 and char-H_2O gasification reactions in the char combustion model.

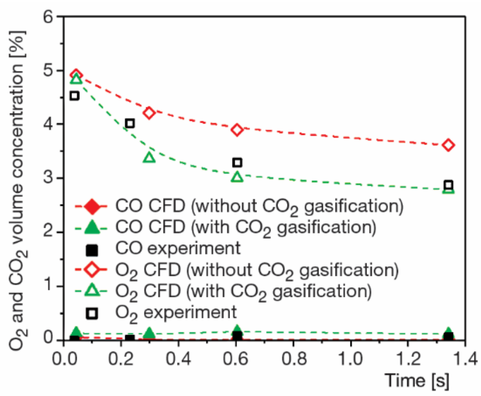

Figure 5. Influence of gasification model on simulation results [59]

Gas phase reactions in coal combustion are also very important, including the pyrolysis and combustion of gaseous components. It is necessary to model their reaction mechanisms and turbulent-chemical interactions. For reaction mechanism model, the volatile matter combustion was simplified as a one-step reaction in early oxy-fuel combustion numerical simulation [62], but this could not accurately predict the CO concentration. Later, Toporov *et al.* [60] proposed a two-step simplified model for reaction mechanism of volatile matter combustion:

$$C_xH_yO_lS_nN_m + \left(\frac{x}{2} + n - \frac{l}{2}\right)O_2 \rightarrow xCO + \frac{y}{2}H_2 + nSO_2 + \frac{m}{2}N_2 \tag{5a}$$

$$CO + \frac{1}{2}O_2 \rightarrow CO_2 \tag{5b}$$

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O \tag{5c}$$

Simulation results show that the temperature distribution and O_2 concentration are in good agreement with the experimental results, but the CO concentration distribution is not given in the paper. Andersen *et al.* [63] modified the two-step and four-step reaction mechanisms widely used in air-fuel combustion to adapt to oxy-fuel combustion. The reaction products were predicted through detailed chemical reaction kinetic simulation, and the results shows that both predicted temperature and CO concentrations are closer to the experimental results than the unmodified model results. In general, oxy-coal combustion is more complicated, and its mechanism model needs further study. Especially for heterogeneous reactions based on different coal types, thermo-gravimetric experiment of oxy-fuel combustion for coal char should be carried out to obtain the reaction kinetic parameters.

In turbulent flame, the coupling of turbulence and chemical reactions is also the focus in oxy-fuel combustion simulation. Most simulations use the eddy-dissipation model (EDM). In addition, a finite rate/eddy dissipation model is proposed, that is, the reaction rate takes a smaller one between the Arrhenius rate and the eddy dissipation rate. Furthermore, the eddy dissipation conceptual (EDC) model is proposed as an extension of EDM to adapt to the multi-step chemical reaction in turbulent flow. It assumes that the reaction takes place in a small turbulent structure controlled by Arrhenius' rate. Cui *et al.* [64] used EDC model for the coupling of turbulence and chemical reaction and performed numerical calculation on oxy-coal combustion for a swirl burner, and the results shows a better prediction of flame temperature and ignition position. Jovanović *et al.* [53] simulated the oxy-fuel burner for pulverized coal with a finite rate/eddy dissipation model and the results are in a good agreement with the experiment. Liu *et al.* [65] presents a numerical investigation on oxy-coal swirling flame and compares the influence of EDM and EDC models, it founds that superior results for EDC model in the prediction of exhausted flue gas mixture and CO concentration.

Summary and outlook

Oxy-fuel combustion of pulverized coal is different from air-fuel combustion in heat transfer and combustion characteristics. At present, the researches of oxy-fuel combustion mainly focus on the combustion characteristics, heat transfer, pollutant emission, and numerical simulation methods. This review is not meant to be exhaustive and only representative works are selected. Nevertheless, it is hoped to be helpful for researchers in quickly knowing the underlying knowledge and research topic of oxy-coal combustion. The summaries of key results and directions that can be further developed in the future include as follows.

- Due to high specific heat and density of CO_2, the ignition, flame and heat transfer characteristics of oxy-coal combustion are different from that in air-fuel combustion. Therefore, it is necessary to develop boiler design guidelines suitable for oxy-coal combustion. In addition, it also needs to develop suitable burner for oxy-coal combustion.

- It is found that oxy-coal combustion can reduce NO_x emissions, and flue gas recycle can reduce SO_2 emissions. However, the principle is still unclear. The migration law of N and S in oxy-coal combustion, their mutual coupling mechanism and the relationship with other elements need to be further studied. There is no systematic report on the emission of fly ash and trace elements, so further investigation is needed.
- High triatomic gas concentration makes the radiative heat transfer of flue gas an important subject of oxy-fuel combustion. Using WSGG model can simulate the gas radiative characteristics in oxy-fuel atmosphere more accurately and efficiently. The radiation of solid particles in oxy-coal combustion is dominant. Therefore, it is an interesting subject to develop a WSGG model or other radiation model that can describe the mixed radiation characteristics of particles and gases. In addition, there are few reports on the combination of turbulence and radiation effects in oxy-coal combustion, which should be paid more attention.
- Considering heterogeneous models of CO_2-char gasification and H_2O-char gasification can better predict the concentration reaction products. However, the oxy-coal combustion is more complicated; thus, the thermo-gravimetric experiment of oxy-fuel combustion for char should be carried out to obtain reaction kinetic parameters for heterogeneous reactions based on different coals. It is also an important subject to establish a reaction kinetics database for oxy-coal combustion through neural network technology based on experimental results.

Acknowledgment

This work was supported by National Postdoctoral Program for Innovative Talents of China (BX2021254) and Open Project of Key Laboratory of Solar Energy Utilization & Energy Saving Technology of Zhejiang Province (ZJS-OP-2020-14).

References

[1] Boot-Handford, M. E., *et al.*, Carbon Capture and Storage Update, *Energy & Environmental Science*, *7* (2014), 1, pp. 130-189
[2] Abraham, B. M., *et al.*, Coal-Oxygen Process Provides CO_2 for Enhanced Recovery, *Oil Gas Journal*, *80* (1982), 11, pp. 68-75
[3] Zhou, Z., *et al.*, Process Design and Optimization of State-of-the-Art Carbon Capture Technologies, *Environmental Progress & Sustainable Energy*, *33* (2014), 3, pp. 993-999
[4] Molina, A., Shaddix, C. R., Ignition and Devolatilization of Pulverized Bituminous Coal Particles During Oxygen/Carbon Dioxide Coal Combustion, *Proceedings of the Combustion Institute*, *31* (2007), 2, pp. 1905-1912
[5] Glarborg, P., Bentzen, L. L. B., Chemical Effects of a high CO_2 Concentration in Oxy-Fuel Combustion of Methane, *Energy & Fuels*, *22* (2008), 1, pp. 291-296
[6] Liu, F., *et al.*, The Chemical Effect of CO_2 Replacement of N_2 in Air on the Burning Velocity of CH_4 and H_2 Premixed Flames, *Combustion and flame*, *133* (2003), 4, pp. 495-497
[7] Kiga, T., *et al.*, Characteristics of Pulverized-Coal Combustion in the System of Oxygen/Recycled Flue Gas Combustion, *Energy Conversion and Management*, *38* (1997), Suppl., pp. S129-S134
[8] Moron´, W., Rybak, W., Ignition Behaviour and Flame Stability of Different Ranks Coals in Oxy Fuel Atmosphere, *Fuel*, *161* (2015), Dec., pp. 174-181
[9] Shaddix, C. R., Molina, A., Particle Imaging of Ignition and Devolatilization of Pulverized Coal During Oxy-Fuel Combustion, *Proceedings of the Combustion Institute*, *32* (2009), 2, pp. 2091-2098
[10] Rathnam, R. K., *et al.*, Differences in Reactivity of Pulverized Coal in Air (O_2/N_2) and Oxy-Fuel (O_2/CO_2) Conditions, *Fuel Processing Technology*, *90* (2009), 6, pp. 797-802
[11] Saastamoinen, J. J., *et al.*, Pressurized Pulverized Fuel Combustion in Different Concentrations of Oxygen and Carbon Dioxide, *Energy & Fuels*, *10* (1996), 1, pp. 121-133

[12] Wall, T., *et al.*, An Overview on Oxyfuel Coal Combustion-State of the Art Research and Technology Development, *Chemical Engineering Research & Design*, *87* (2009), 8, pp. 1003-1016
[13] Hecht, E. S., *et al.*, Effect of CO_2 and Steam Gasification Reactions on the Oxy-Combustion of Pulverized Coal Char, *Combustion & Flame*, *159* (2012), 11, pp. 3437-3447
[14] Kim, D., *et al.*, Effect of CO_2 Gasification Reaction on Char Particle Combustion in Oxy-Fuel Conditions, *Fuel*, *120* (2014), 1, pp. 130-140
[15] Chen, L., *et al.*, Oxy-Fuel Combustion of Pulverized Coal: Characterization, Fundamentals, Stabilization and CFD Modeling, *Progress in Energy & Combustion Science*, *38* (2012), 2, pp. 156-214
[16] Toporov, D. D., *Combustion of Pulverized Coal in a Mixture of Oxygen and Recycled Flue Gas*, Elsevier., Waltham, Mass., USA, 2015
[17] Buhre, B. J. P., *et al.*, Oxy-Fuel Combustion Technology for Coal-fired Power Generation, *Progress in Energy and Combustion Science*, *31* (2005), 4, pp. 283-307
[18] Nozaki, T., *et al.*, Analysis of the Flame Formed During Oxidation of Pulverized Coal by an O_2-CO_2 Mixture, *Energy*, *22* (1997), 2-3, pp. 199-205
[19] Andersson, K., *et al.*, Radiation Intensity of Lignite-Fired Oxy-Fuel Flames, *Experimental Thermal and Fluid Science*, *33* (2008), 1, pp. 67-76
[20] Hjartstam, S., *et al.*, Combustion Characteristics of Lignite-Fired Oxy-Fuel Flames, *Fuel*, *88* (2009), 11, pp. 2216-2224
[21] Ren, F., *et al.*, Effects of Strain Rate and CO_2 on NO Formation in CH_4/N_2/O_2 Counter-Flow Diffusion Flames, *Thermal Science*, *22* (2018), Suppl. 2, pp. S769-S776
[22] Glarborg, P., Bentzen, L. L. B., Chemical Effects of High CO_2 Concentration in Oxy-Fuel Combustion of Methane, *Energy & Fuels*, *22* (2008), 1, pp. 291-287
[23] Zhang, J., *et al.*, Ignition in 40 kW Co-Axial Turbulent Diffusion Oxy-Coal Jet Flames, *Proceedings of the Combustion Institute*, *33* (2011), 2, pp. 3375-3382
[24] Tan, Y., *et al.*, Combustion Characteristics of Coal in a Mixture of Oxygen and Recycled Flue Gas, *Fuel*, *85* (2006), 4, pp. 507-512
[25] Kaß, H., *et al.*, The Combustion of Dry Lignite Under Oxy-Fuel Process Conditions in a 0.5 MWth Test Plant, *Energy Procedia*, *1* (2009), 1, pp. 423-430
[26] Hu, Y., *et al.*, CO_2, NOx and SO_2 Emissions from the Combustion of Coal with High Oxygen Concentration Gases, *Fuel*, *79* (2000), 15, pp. 1925-1932
[27] Sheng, C., Li, Y., Experimental Study of Ash Formation During Pulverized Coal Combustion in O_2/CO_2 Mixtures, *Fuel*, *87* (2008), 7, pp. 1297-1305
[28] Sheng, C., *et al.*, Ash Particle Formation During O_2/CO_2 Combustion of Pulverized Coals, *Fuel Processing Technology*, *88* (2007), 11-12, pp. 1021-1028
[29] Sheng, C., *et al.*, Fine Ash Formation During Pulverized Coal Combustion-a Comparison of O_2/CO_2 Combustion *vs.* Air Combustion, *Energy & Fuels*, *21* (2007), 2, pp. 435-440
[30] Suriyawong, A., *et al.*, Sub-Micro Meter Particle Formation and Mercury Speciation Under O_2+CO_2 Coal Combustion, *Energy & Fuels*, *20* (2006), 6, pp. 2357-2363
[31] Guo, Z., *et al.*, The Impact of Combustion Characteristics and Flame Structure on Soot Formation in Oxy-Enhanced and Oxy-Fuel Diffusion Flames, *Science China Technological Sciences*, *56* (2013), 7, pp. 1618-1628
[32] Saanum, I., Ditaranto, M., Soot Formation in Diffusion Flames in Oxy-Fuel Atmospheres, Report No. 115, SINTEF Energy Research AS, Trondheim, Norway, 2015
[33] Morris, W. J., *et al.*, Soot, Unburned Carbon and Ultrafine Particle Emissions from Air- And Oxy-Coal Flames, *Proceedings of the Combustion Institute*, *33* (2011), 2, pp. 3415-3421
[34] Xu, M., *et al.*, Overview of Trace Elements Research in Coal Combustion Process, (In Chinese), *Proceedings of CSEE*, *10* (2001), 21, pp. 33-38
[35] Zheng, L., Furimsky, E., Assessment of Coal Combustion in O_2+CO_2 by Equilibrium Calculations, *Fuel Processing Technology*, *81* (2003), 1, pp. 23-34
[36] Suriyawong, A., *et al.*, Submicrometer Particle Formation and Mercury Speciation Under O_2-CO_2 Coal Combustion, *Energy & Fuels*, *20* (2006), 6, pp. 2357-2363
[37] Zhang, L., *et al.*, Study on Pollutant Emission Characteristics From Oxy-fuel Combustion of Coal with Recycled Flue Gas, (In Chinese), *Proceedings of CSEE*, *29* (2009), 29, pp. 35-40
[38] Nikolopoulos, N., *et al.*, Numerical Investigation of the Oxy-Fuel Combustion in Large Scale Boilers Adopting the ECO-Scrub Technology, *Fuel*, *90* (2011), 1, pp. 198-214

[39] Andersen, J., *et al.*, Global Combustion Mechanisms for use in CFD Modeling Under Oxy-Fuel Conditions, *Energy &Fuels*, *23* (2009), 3, pp. 1379-1389
[40] Qi, F., *et al.*, Numerical Study on Ladle Baking Process of Oxy-Fuel Combustion, *Thermal Science*, *24* (2020), 6A, pp. 3511-3520
[41] Edge, P., *et al.*, LES Modelling of Air and Oxy-Fuel Pulverised Coal Combustion – Impact on Flame Properties, *Proceedings of the Combustion Institute*, *33* (2011), 2, pp. 2709-2716
[42] Porter, R., *et al.*, Evaluation of Solution Methods for Radiative Heat Transfer in Gaseous Oxy-Fuel Combustion Environments, *Journal of Quantitative Spectroscopy and Radiative Transfer 111* (2010), 14, pp. 2084-2094
[43] Juric, F., *et al.*, Assessment of Radiative Heat Transfer Impact on a Temperature Distribution Inside a Real Industrial Swirled Furnace, *Thermal Science*, *24* (2020), 6A, pp. 3663-3672
[44] Coelho, P. J., *et al.*, Numerical Simulation of Radiative Heat Transfer from Non-Gray Gases in Three-Dimensional Enclosures, *Journal of Quantitative Spectroscopy and Radiative Transfer*, *74* (2002), 3, pp. 307-328
[45] Modest, M. F., The Weighted-Sum-of-Gray-Gases Model for Arbitrary Solution Methods in Radiative Transfer, *Journal of heat transfer-ASME*, *113* (1991), 3, pp. 650-656
[46] Yin, C., *et al.*, New Weighted Sum of Gray Gases Model Applicable to Computational Fluid Dynamics (CFD) Modeling of Oxy-Fuel Combustion: Derivation, Validation, and Implementation, *Energy & Fuels*, *24* (2010), 12, pp. 6275-6282
[47] Johansson, R., *et al.*, Account for Variations in the H_2O to CO_2 Molar Ratio when Modelling Gaseous Radiative Heat Transfer with the Weighted-Sum-of-Grey-Gases Model, *Combustion & Flame*, *158* (2011), 5, pp. 893-901
[48] Dorigon, L. J., *et al.*, WSGG Correlations Based on HITEMP2010 for Computation of Thermal Radiation In Non-Isothermal, Non-Homogeneous H_2O/CO_2 Mixtures, *International Journal of Heat and Mass Transfer*, *64* (2013), Sept., pp. 863-873
[49] Shan, S., *et al.*, New Weighted-Sum-of-Gray-Gases Model For Typical Pressurized Oxy-Fuel Conditions, *International Journal of Energy Research*, *41* (2017), 15, pp. 2576-2595
[50] Shan, S., *et al.*, New Pressurized WSGG Model and the Effect of Pressure on the Radiation Heat Transfer of H_2O/CO_2 Gas Mixtures, *International Journal of Heat and Mass Transfer*, *121* (2018), June, pp. 999-1010
[51] Centeno, F. R., *et al.*, Comparison of Different WSGG Correlations in the Computation of Thermal Radiation in a 2D Axisymmetric Turbulent Non-Premixed Methane-Air Flame, *Journal of the Brazilian Society of Mechanical Sciences & Engineering*, *35* (2013), 4, pp. 419-430
[52] De Rocha Barcelos, B., Centeno, F. R., Numerical Assessment of the Effect Of Inflow Turbulators on the Thermal Behavior of a Combustion Chamber, *Thermal Science*, *25* (2021), 1A, pp. 209-220
[53] Jovanović, R., *et al.*, Experimental and Numerical Investigation of Flame Characteristics During Swirl Burner Operation Under Conventional and Oxy-Fuel Conditions, *Thermal Science*, *21* (2017), 3, pp. 1463-1477
[54] Zhang, J., *et al.*, Numerical Investigation of Oxy-Coal Combustion in a Large-Scale Furnace: Non-Gray Effect of Gas and Role of Particle Radiation, *Fuel*, *139* (2015), 1, pp. 87-93
[55] Johansson, R., *et al.*, Influence of Particle and Gas Radiation in Oxy-Fuel Combustion, *International Journal of Heat & Mass Transfer*, *65* (2013), 5, pp. 143-152
[56] Backstrom, D., *et al.*, Measurement and Modeling of Particle Radiation in Coal Flames, *Energy & Fuels*, *28* (2014), 3, pp. 2199-2210
[57] Yin, C., On Gas and Particle Radiation in Pulverized Fuel Combustion Furnaces, *Applied Energy*, *157* (2015), 15, pp. 554-561
[58] Khare, S. P., *et al.*, Factors inFluencing the Ignition of Flames from Air-Fired Swirl Pf Burners Retrofitted to Oxy-Fuel, *Fuel*, *87* (2008), 7, pp. 1042-1049
[59] Kuhr, C., *et al.*, Modeling of Char Combustion in CO_2/O_2 and N_2/O_2 Atmospheres, *Proceedings* 35^{th}, International Technical Conference on Clean Coal and Fuel Systems, Clearwater, Fla., USA, 2010
[60] Toporov, D., *et al.*, Detailed Investigation of a Pulverized Fuel Swirl Flame in CO_2/O_2 Atmosphere, *Combustion and Flame*, *155* (2008), 4, pp. 605-618
[61] Muller, M., *et al.*, Advanced Modeling of Pulverized Coal Combustion Under Oxy-Fuel Conditions, *Proceedings* 35^{th}, International Technical Conference on Clean Coal and Fuel Systems, Clearwater, Fla., USA, 2010

[62] Wang, C. S., *et al.*, Combustion of Pulverized Coal Using Waste Carbon Dioxide and Oxygen, *Combustion and Flame*, *72* (1988), 3, pp. 301-310
[63] Andersen, J., *et al.*, Global Combustion Mechanisms for Use in CFD Modeling Under Oxy-Fuel Conditions, *Energy & Fuels*, *23* (2009), 3, pp. 1379-1389
[64] Cui, K., *et al.*, Numerical Simulation of Oxy-coal Combustion for a Swirl Burner with EDC Model, *Chinese Journal of Chemical Engineering*, *22* (2014). 2, pp. 193-201
[65] Liu, J., *et al.*, Mathematical Modeling of Air- and Oxy-Coal Confined Swirling Flames on Two Extended Eddy-Dissipation Models. *Industrial & Engineering Chemistry Research*, *51* (2012), 2, pp. 696-708

Paper submitted: March 29, 2021
Paper revised: June 11, 2021
Paper accepted: June 18, 2021