

# Research Coal Technology—Article

## Fundamental and Technical Challenges for a Compatible Design Scheme of Oxyfuel Combustion Technology

Chuguang Zheng\*, Zhaohui Liu, Jun Xiang, Liqi Zhang, Shihong Zhang, Cong Luo, Yongchun Zhao

**ABSTRACT** Oxyfuel combustion with carbon capture and sequestration (CCS) is a carbon-reduction technology for use in large-scale coal-fired power plants. Significant progress has been achieved in the research and development of this technology during its scaling up from 0.4 MW<sub>th</sub> to 3 MW<sub>th</sub> and 35 MW<sub>th</sub> by the combined efforts of universities and industries in China. A prefeasibility study on a 200 MW<sub>e</sub> large-scale demonstration has progressed well, and is ready for implementation. The overall research development and demonstration (RD&D) roadmap for oxyfuel combustion in China has become a critical component of the global RD&D roadmap for oxyfuel combustion. An air combustion/oxyfuel combustion compatible design philosophy was developed during the RD&D process. In this paper, we briefly address fundamental research and technology innovation efforts regarding several technical challenges, including combustion stability, heat transfer, system operation, mineral impurities, and corrosion. To further reduce the cost of carbon capture, in addition to the large-scale deployment of oxyfuel technology, increasing interest is anticipated in the novel and next-generation oxyfuel combustion technologies that are briefly introduced here, including a new oxygen-production concept and flameless oxyfuel combustion.

**KEYWORDS** oxyfuel combustion, research development and demonstration, CO<sub>2</sub> capture

### 1 Introduction

The emission of CO<sub>2</sub>, the main greenhouse gas, from coal combustion has been the subject of intense focus in recent years in connection with international negotiations on climate change. Since 2007, CO<sub>2</sub> emissions from China have exceeded those from the United States, making China the world's largest contributor of this greenhouse gas. In 2010, China emitted over 8.2 billion tons of CO<sub>2</sub> [1]. CO<sub>2</sub> capture, utilization, and sequestration (CCUS) technology is one of the most feasible technologies to combat climate change. Pre-

liminary studies and engineering demonstrations of CCUS technology have been carried out by the government, universities, research institutes, and enterprises in China as well as through recent international cooperation [2]. Technologies and projects based on post-combustion, oxyfuel-combustion, and pre-combustion CO<sub>2</sub> capture are being developed and constructed, respectively.

As China's coal-dominated energy structure cannot be changed within a short period of time, oxyfuel technology is considered to be the best potential CCUS technology for large-scale promotion and commercialization [3, 4]. During this process, fuel is burned in a mixture of oxygen and recycled flue gas (RFG). Unlike conventional fossil-fuel-fired power stations that use air as the oxidant, an oxy-fired plant employs an air separation unit to produce an oxygen stream. The oxygen stream is combined with RFG to produce an oxygen-enriched gas for the oxidant (Figure 1). The recycling of flue gas is necessary to moderate the otherwise excessively high flame temperature that would result from burning pure oxygen. After the removal of moisture and other impurities from the flue gas exhaust stream, high-purity CO<sub>2</sub> is produced.

This technology was first proposed in 1982 for the purpose of producing a high-purity CO<sub>2</sub> stream for use in enhanced oil recovery [5]. The technology was then broadly investigated for use in producing enriched CO<sub>2</sub> in flue gas. Many reports indicate that oxyfuel is the cheapest of the three CCUS capture options; it has higher thermal efficiency and a lower energy penalty than the other options. This flexible technology can be used in new power plants and in existing power plant retrofits, with no technological barriers to its implementation. In addition, it is an environmentally friendly process that can simultaneously remove pollutants [4–7]. Pilot-scale studies have been carried out in the years following the proposal of this technology. Over the last decade, global research activity has increased to the point at which several demonstration phase projects have begun, and the commercial concept is expected before 2020.

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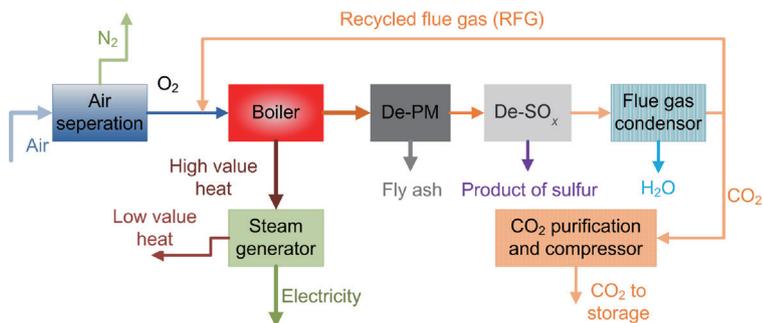


Figure 1. Schematic diagram of oxyfuel combustion.

Fundamental research on oxyfuel combustion in China began in the 1990s. Several research institutes and universities, such as the Huazhong University of Science and Technology (HUST), Southeast University (China), and the North China Electric Power University, have been involved in this field [6]. Research and platform construction of oxyfuel combustion for two main streams of coal combustion—pulverized coal combustion and fluidized bed combustion—have been active in recent years [7]. HUST has developed a roadmap for oxyfuel combustion research and development (Figure 2). A 400 kW<sub>th</sub> small pilot system was built in 2005, followed by a 3 MW<sub>th</sub> full chain system in 2011 (Wuhan, 1 t·h<sup>-1</sup> CO<sub>2</sub> uptake capacity, annual 10 000 t level). Until now, this 3 MW<sub>th</sub> full chain system was the largest oxyfuel combustion test system in China. However, a 35 MW<sub>th</sub> pilot system is expected to start operation in 2015 (Jiuda Salt Co., Yingcheng, Hubei, 12 t·h<sup>-1</sup> CO<sub>2</sub> uptake capacity, annual 0.1 Mt level). In addition, a feasibility study for a 200 MW<sub>e</sub>

demonstration project was started in 2012 (Shenmu Power plant, Guohua, annual million tons level), and project implementation is planned for 2020.

Figure 3 shows the roadmap for worldwide oxyfuel combustion research development and demonstration (RD&D) [8], provided by the International Energy Agency (IEA), and Tables 1 and 2 list worldwide semi-industrial and large-scale demonstration projects. The overall development of oxyfuel combustion technology in China keeps in step with international development. The UK government is sponsoring the front-end engineering design (FEED) study of a 426 MW<sub>e</sub> large-scale oxyfuel combustion plant. In China, the pre-FEED of a 200 MW<sub>e</sub> large-scale demonstration is approaching its target.

Although oxyfuel technology is widely studied, there are still some drawbacks to be overcome. Firstly, the proper control of oxygen and recycled flue gas through the burners is neces-

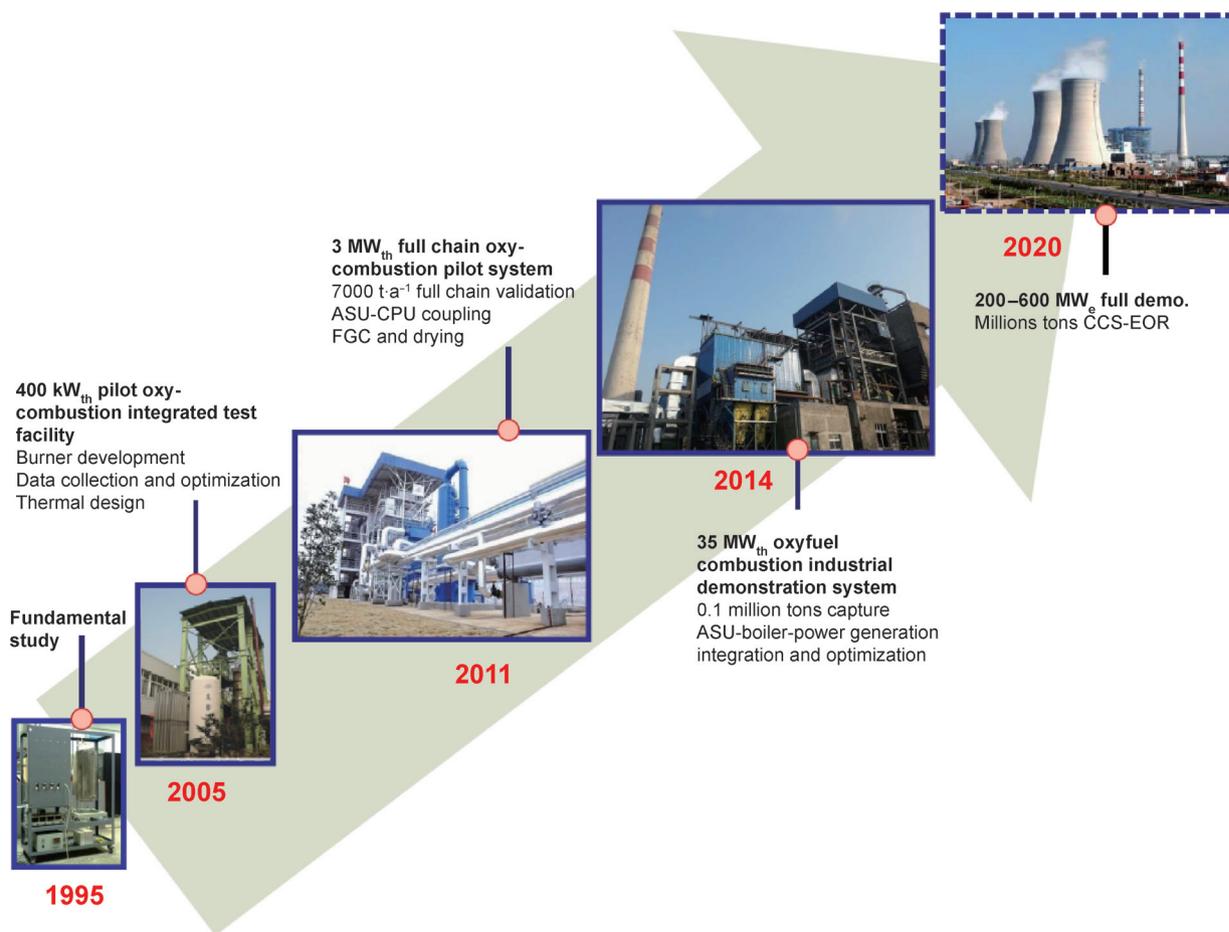


Figure 2. Roadmap of oxyfuel combustion technology at HUST.

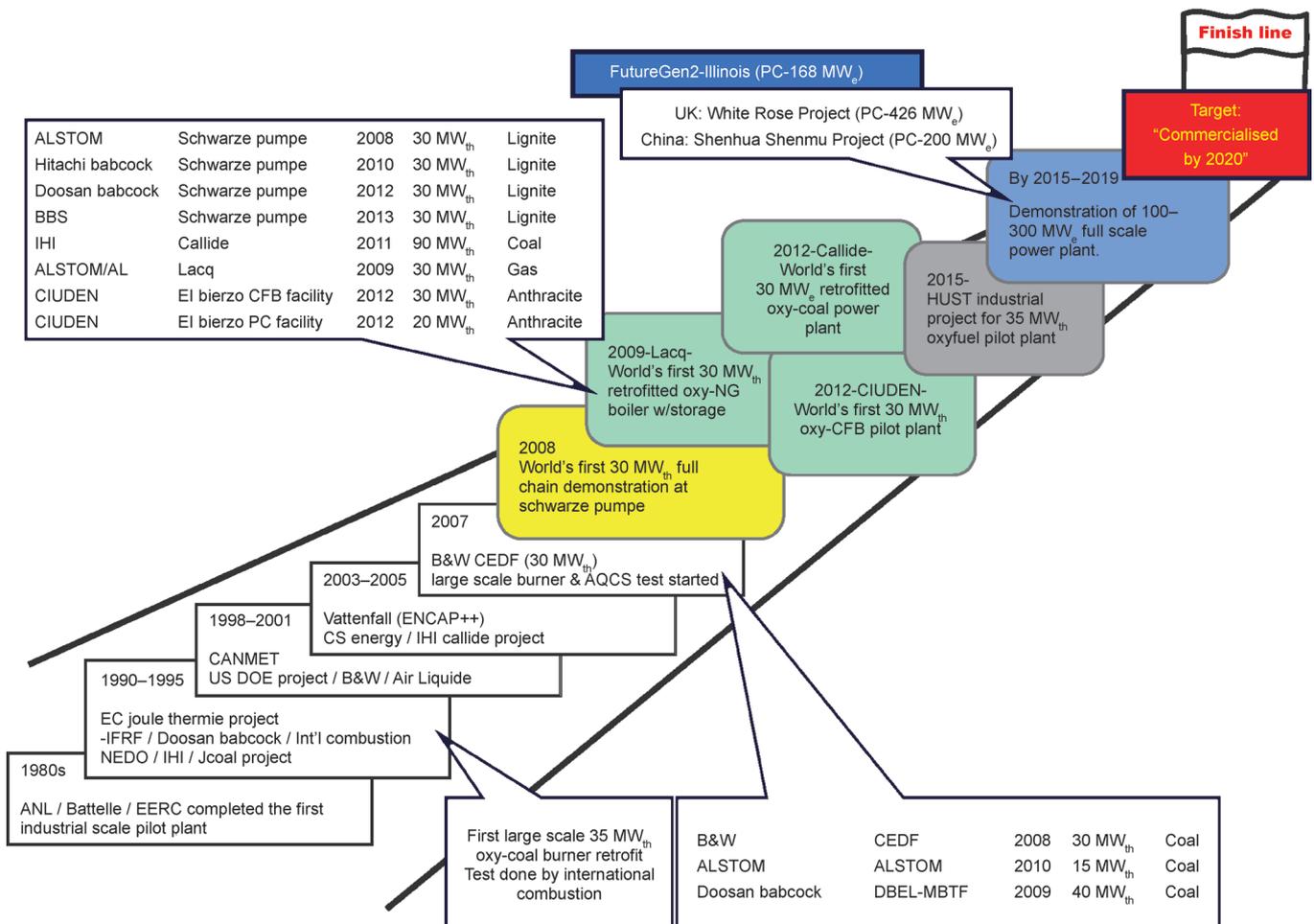


Figure 3. IEA GHG RD&D roadmap of oxyfuel combustion.

Table 1. Worldwide oxyfuel combustion industrial-scale demonstration projects.

Country	Project name	Scale (MW <sub>e</sub> )	Type	Start date	Fuel	Power gen.
Germany	Vattenfall	10	New	2008	Coal	N
Australia	Callide	30	Retro	2010	Coal	Y
France	TOTAL	10	Retro	2009	Gas	Y
Spain	CIUDEN	7	New	2010	Coal	N
China	Yingcheng	12	Retro	2011	Coal	Y

Table 2. Worldwide large-scale oxyfuel combustion demonstration projects.

Country	Project	Scale and parameters	Technology source	Progress and launch time
UK	Capture Power White Rose	426 MW <sub>e</sub> supercritical	ALSTOM AP	FEED study, commission before 2020
China	Shenmu	200 MW <sub>e</sub> subcritical	Shenhua, HUST, DEC	FEED study, commission before 2020

Notes: ALSTOM—ALSTOM Power Inc.; Ap—Air product; DEC—Dongfang Electric Corporation.

sary to ensure fast ignition and flame stability. Secondly, because of air leakage and accumulation in the process of flue gas recirculation, pollutant concentration in the exhaust gas is relatively high in oxyfuel combustion (although the gross emission is substantially reduced). Thirdly, the recirculation process decreases the overall systematic efficiency. To solve these problems and to further understand the details of oxyfuel combustion, systematic experiments and simulations were conducted. Over the past 15 years, researchers have developed a compatible design philosophy involving both air combustion and oxyfuel combustion. In this concept, the system and the boiler can operate at full load in both air combustion mode and oxyfuel combustion mode. In this paper, we discuss and clarify the fundamental study and key

technology innovation of this compatible design, regarding ignition and combustion characteristics, heat transfer and heat surface arrangement, and the transition of oxyfuel combustion. In doing so, we hope to provide useful information for the demonstration and promotion of oxyfuel combustion technology.

## 2 Critical challenges for oxyfuel combustion

### 2.1 Ignition and flame stability

Maintaining flame stability is a major challenge for oxyfuel combustion, as reported in early pilot-scale experimental studies [9, 10]. This challenge stems from the lower adiabatic

flame temperature, lower flame propagation speed, and delayed ignition of coal particles in a CO<sub>2</sub>-diluted environment.

The lower flame transportation velocities in an O<sub>2</sub>/CO<sub>2</sub> environment are evident. Chen et al. [11] investigated the laminar flame speed using the CHEMKIN flame speed calculator model. The adiabatic flame temperature and flame propagation speed of CH<sub>4</sub> in an O<sub>2</sub>/CO<sub>2</sub> atmosphere were both lower than those in an O<sub>2</sub>/N<sub>2</sub> atmosphere. In addition, this difference increased with increasing CO<sub>2</sub> concentration. The lower flame propagation speed of pulverized-coal combustion in CO<sub>2</sub> was reported by Kiga et al. [12] using a microgravity experimental facility.

Two types of ignition mechanism have been observed for pulverized-coal combustion: homogeneous ignition and heterogeneous ignition [13]. Chen et al. [11] used a closed homogeneous CHEMKIN reactor model with the GRI-Mech reaction mechanism to investigate homogeneous ignition, where methane is used as a substitute for the volatiles, ignition delay is significantly longer in the CO<sub>2</sub>-diluent gas than in the N<sub>2</sub>, and the final combustion temperature is about 400 K lower in the CO<sub>2</sub>-diluent gas. Shaddix and Molina [14] observed similar delays in the ignition of high-volatile bituminous coal particles when burning in an O<sub>2</sub>/CO<sub>2</sub> atmosphere compared with an O<sub>2</sub>/N<sub>2</sub> atmosphere. Huang et al. [15] and Huang [16] investigated the ignition characteristics of differently ranked Chinese coals during oxyfuel combustion using a flat-flame supported entrained flow reactor. The results, as shown in Figure 4, indicate that the ignition delay time is inversely proportional to  $Y_{O_2}^n$ , where  $n$  is 0.15–0.2. The presence of high-concentration CO<sub>2</sub> leads to increased ignition delay time and decreased ignition stability for high-volatile coal. In contrast, the presence of high-concentration CO<sub>2</sub> leads to decreased ignition delay time and increased ignition stability for low-volatile coal. The effects of CO<sub>2</sub> on the heterogeneous ignition temperature of two coal chars were studied using a wire-mesh reactor [17].

Lower laminar burning velocity and narrower flammability limits may cause combustion destabilization in oxyfuel combustion [11]. This opens up opportunities for advanced oxy-coal burner design including the following variables: ① flue gas recycle ratio; ② oxygen concentrations in the primary, secondary, and tertiary streams; ③ the distribution of gas volumes in the different streams and their momentum; and ④ the preheat temperature of different streams.

A series of swirl oxyfuel burners was designed and applied to 0.4 MW<sub>th</sub>, 3 MW<sub>th</sub>, and 35 MW<sub>th</sub> oxyfuel combustion boilers [18, 19]. The effects of the blockage ratio of bluff body, swirl number, oxygen injection tube number and size, oxygen concentrations in the primary stream, and recycle ratio were investigated. To achieve a stable pulverized-coal flame under both air-fuel and oxyfuel combustion, the following design rules were discovered and adopted: The oxygen is premixed with flue gas before its injection to the burner as primary stream or secondary stream; the oxygen partial pressure of the primary stream is maintained below 18% to ensure safety while oxygen is supplied from the secondary stream; the tertiary stream is switched off to ensure a similar momentum ratio of the primary stream and secondary stream; and the mo-

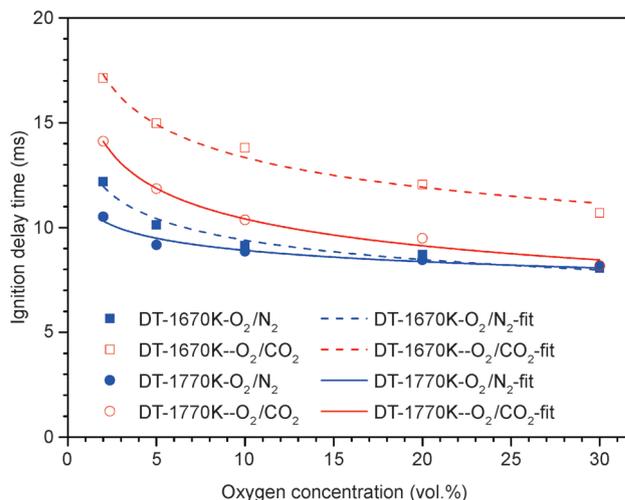


Figure 4. Ignition delay of DT bituminous coal under O<sub>2</sub>/N<sub>2</sub> and O<sub>2</sub>/CO<sub>2</sub> conditions.

mentum of the primary stream is kept the same to ensure its pneumatic conveyance capabilities. The performance of the 3 MW<sub>th</sub> swirl burner was extensively validated at the 3 MW<sub>th</sub> pilot facility, and showed that stable combustion was achieved, and a burnout rate of 98% was obtained under oxyfuel combustion, in comparison to 95% under air combustion. Figure 5 shows the predicted temperature profiles in the 3 MW furnace. Generally, the profile is very similar under air combustion and oxyfuel combustion, although the temperature peak of the latter decreases by about 200 K. These rules for combustion system design were also adopted for the design of a combustion system for a 200 MW<sub>e</sub> oxyfuel utility boiler, both tangential fired and wall-fired.

## 2.2 In-furnace heat transfer

In oxyfuel combustion, high concentrations of gases participating in radiative heat transfer (i.e., CO<sub>2</sub>, H<sub>2</sub>O) can result in significantly different emissivity within the boiler, influencing the radiative transfer and the absorption of heat within the system.

A few modified weighted-sum-of-grey-gases (WSGG) models were proposed in recent years to consider the wide range of H<sub>2</sub>O to CO<sub>2</sub> ratios for oxyfuel combustion. Based on the up-to-date HITEMP 2010 database, we obtained new parameters of a WSGG model for oxyfuel combustion by combining the features of a full-spectrum  $k$ -distribution (FSK) model with a WSGG model, in which the weighting factor and absorption coefficient were directly obtained from the  $k$ -distribution, and the fitting formula of the absorption coefficient was improved [20]. The new parameters of this WSGG model were extensively validated by comparing the radiative source terms and radiative heat fluxes predicted with the line-by-line (LBL) model integration of the HITEMP 2010 database for a one-dimensional slab system. Figure 6 shows the predicted emissivity of different parameters in WSGG models. The results show that the new parameters can significantly improve prediction accuracy in oxyfuel combustion.

Different researchers have observed conflicting heat-transfer results, and have considered that these should be optimized to ensure efficient operation. However, for a retro-

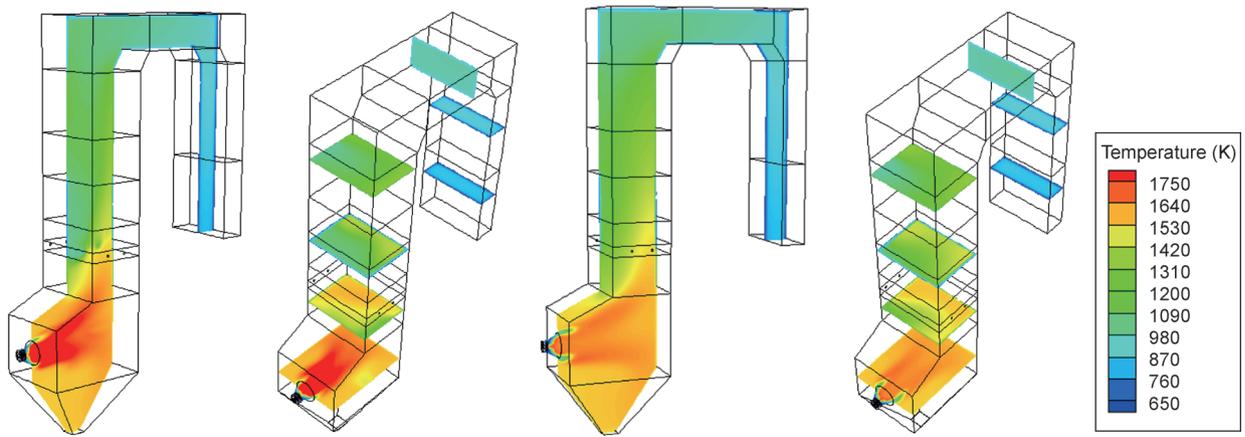


Figure 5. Predicted temperature profile of 3 MW furnace with a single swirl burner. Left: air combustion, and right: oxyfuel combustion.

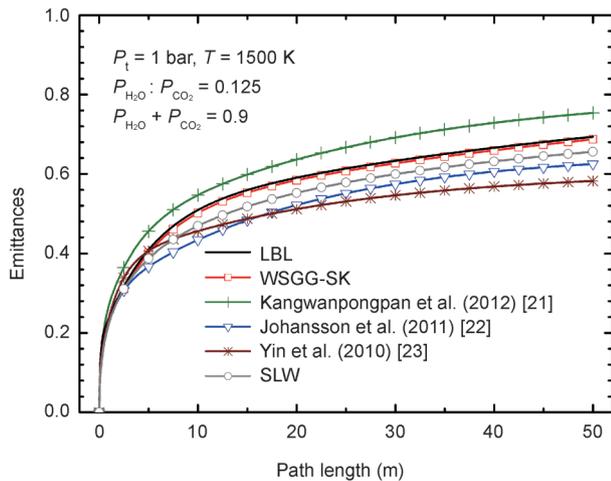


Figure 6. The total emittances calculated by different models for dry recycle oxyfuel combustion.

fit case where in-furnace heat transfer is matched, and for a given flue gas oxygen concentration, the oxyfuel case results in a decreased furnace exit-gas temperature, for which the match of convective heat transfer in the duct of the flue gas would be questionable. Andersson et al. [24] found that the recycle ratio is an appropriate tuning parameter to achieve similar boiler performance for air and oxyfuel combustion, which is a goal if existing boilers are to be changed from air to oxyfuel operation. They found that the temperature, and thereby the total radiation intensity of the oxyfuel flames, increases with decreasing flue gas recycle rate. In addition, the total radiation intensities are similar during oxyfuel and air-fuel operation as long as the temperature distributions are similar. Smart et al. [25] studied convective heat-transfer coefficients in oxy and air coal combustion. They found that an acceptable operational range of the flue gas recycle ratio exists in which the convective heat transfer in oxy-coal combustion can be approximately matched to that of air combustion. Thus, an existing coal-fired power plant can be retrofitted to function as an oxy-coal plant without replacing the boiler or heat exchangers.

Figure 7 shows a summary of the relative heat transfer of oxyfuel to air-firing, based on data from our simulation work of a 200 MW<sub>e</sub> tangential-fired furnace, and data from experiments and simulations on a large pilot- or full-scale furnace.

It is evident that relative heat transfer is slightly dependent on oxygen concentration, as revealed by the predicted results in the study by Black et al. [26], and the experiment in the Callide 30 MW<sub>e</sub> [27] and the Schwarze Pumpe 30 MW<sub>th</sub> furnaces [28]. In particular, experimental data from the pilot-scale facility shows that the relative heat-transfer ratio increases by several percent in a wide scope of oxygen concentration, that is, from 25% to 30% by volume. In this study, similar heat transfer can be achieved at 28.5% and 27.1% oxygen concentration for dry and wet recycle, respectively. However, the required oxygen concentration is approximately 26% in design calculations of boiler performance, which are based on thermochemistry equilibrium assumptions, regardless of the influence of the relatively slow CO reaction. It is critical to keep in mind the difference between numerical simulation and design calculations of boiler performance, which should be validated with experimental data from future large-scale demonstrations. The optimum oxygen concentration should depend on a comprehensive consideration of the combustion properties and heat-transfer properties.

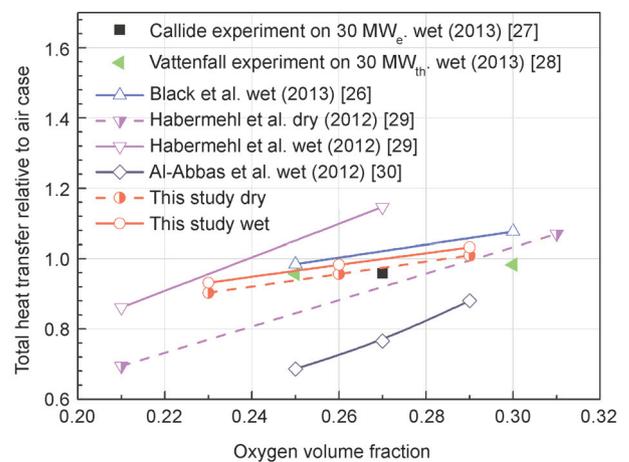


Figure 7. Normalized total heat transfer of large-scale furnaces.

### 2.3 System operation and optimization

Under oxyfuel conditions, a typical operation would involve starting up under air firing, and then gradually switching to oxyfuel mode by recycling the flue gas into the boiler [31]. However, the process of smoothly switching modes from air

firing to oxyfuel is still considered to be a challenge [11]. Very little research has been conducted on appropriate switching strategies or on valuable operational highlights such as controlling recycle flue gas, oxygen, and air streams. Related research that is reported in the literature can be divided into two types: pilot experiments and process simulations. On the experimental front, researchers have operated several oxy-combustion pilot plants as a means to obtain practical operational experience for commercial demonstration [32, 33]. These operational results provide a benchmark for understanding the mode-switching process in an oxy-combustion power plant. However, deeper insight into the dynamic characteristics of this novel technology is still constrained, due to a lack of sufficient experimental measurements. Applying dynamic process simulation is a powerful way to identify the comprehensive characteristics of an oxy-combustion power plant.

A dynamic model of an oxyfuel combustion system was developed, based on a 3 MW<sub>th</sub> oxyfuel pilot system and using Aspen Dynamics. The performance of this model was evaluated in detail by comparing its numeric results with experimental data, with a focus on both steady-state and dynamic behavior [34]. In addition, the transition between air combustion and oxyfuel combustion was optimized through dynamic simulation [35]. This transition was implemented by adjusting the air intake valves, recycled flue gas valves, exhaust valves, and oxygen intake valves; by monitoring oxygen concentration at the outlet of the furnace chamber and in the primary/secondary gas flow; and by monitoring the primary/secondary gas flow rates. Figure 8 illustrates the variation in primary gas flow rate and oxygen concentration. In order to maintain the stability of radiative heat transfer in the boiler, the inlet oxygen concentration was controlled so that it increased slowly during the transition, eventually remaining at 26%. Fluctuation of the oxygen concentration at the outlet of the chamber was regulated at 2%–5% (eventually stabilizing around 3%) to ensure safe and economical combustion. In addition, the fluctuation of the primary gas-flow rate was small enough, around 18% with an oxygen partial pressure, to permit safe and reliable pulverized-coal transportation. The transition process took 30 min to complete, and the mode-switching strategy studied in the model was successfully applied at the 3 MW<sub>th</sub> test facility.

In addition to the issue of the mode-switching process, the issue of air-leakage monitoring and control at oxyfuel combustion plants must be resolved. CO<sub>2</sub> concentration decreases by around 6% when air leakage is at 8% [36]. This decrease is significant when assessing the influence of recirculation in the flue gas species content, since CO<sub>2</sub> purity is one of the challenges for CO<sub>2</sub> capture in oxyfuel combustion. At the 3 MW<sub>th</sub> test facility, a positive furnace-pressure operating condition was investigated to determine its effect on reducing air leakage in the overall system. As shown in Figure 9, the CO<sub>2</sub> concentration gradually increased when the furnace pressure was set at 60 Pa. This obvious increase in CO<sub>2</sub> concentration indicates that a positive furnace-pressure operating condition is preferable in oxyfuel combustion. Figure 9 shows that the CO<sub>2</sub> concentration of the flue gas can be raised to 80% (vol,

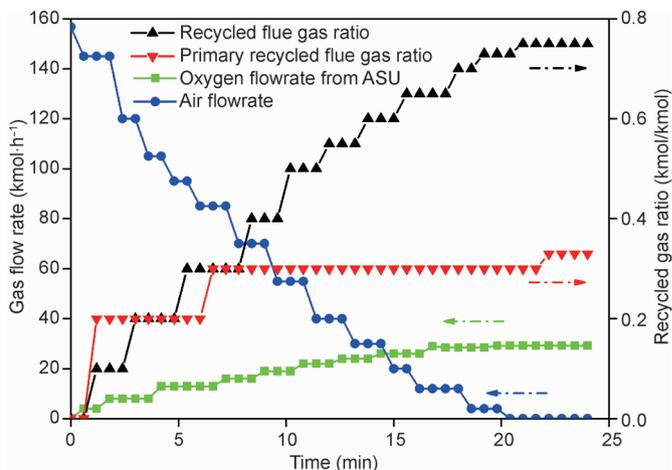


Figure 8. The change of gas flow rates during the transition process.

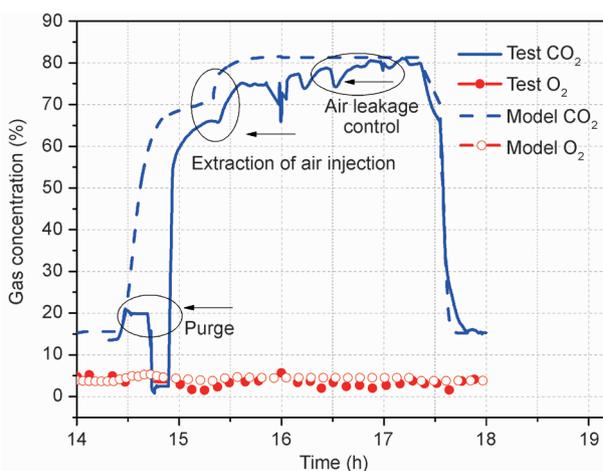


Figure 9. CO<sub>2</sub> and O<sub>2</sub> concentrations during the transition process.

dry) in both model and test results, by optimum regulation.

#### 2.4 Mineral impurities and corrosion issues

Mineral impurities in coal may cause ash deposition, erosion, slagging, and fouling on the surface of heat exchangers, which may lead to tube explosion, heat transfer, and boiler-efficiency loss. Therefore, mineral behavior during oxyfuel combustion has attracted extensive research in recent years.

High CO<sub>2</sub> concentration under oxyfuel conditions will aggravate the deposition of ash [37]; the migration and transformation of various minerals are also different under these conditions [38]. Early studies based on the X-ray diffraction analysis of total ash show only a weak indication of mineral carbonation under oxyfuel combustion conditions. Recent studies focus more on the behavior of iron- and calcium-related minerals, and all these studies indicate enhanced deposition and slagging potential under oxyfuel conditions. Compared to a traditional coal-combustion atmosphere, weight loss due to pyrite decomposition increased in an oxyfuel atmosphere, and a high CO<sub>2</sub> concentration resulted in a shortened decomposition process and a slight extension of the oxidation process. The melting endothermic of pyrite was higher in an oxyfuel atmosphere (30.4 J·g<sup>-1</sup>) than in air combustion (5.0 J·g<sup>-1</sup>). The decomposition of calcite was clearly delayed in the oxyfuel atmosphere, and TG-DSC curves indi-

cated a decomposition temperature of up to 867 °C in the oxy-fuel atmosphere, compared to 697 °C in air combustion. The melting endothermic of calcite was higher, up to 32.4 J·g<sup>-1</sup> in the oxyfuel atmosphere, compared to 15.3 J·g<sup>-1</sup> in air combustion. The melting temperatures of minerals were lower in the oxyfuel atmosphere compared to those in air, which resulted in easier mineral melting, and caused serious ash deposits in the oxyfuel combustion.

Figure 10 shows a recent study [39] of the mineral behavior of a sub-bituminous ZD coal. In this study, all ash is generated in a high-temperature (1700 K) drop-tube furnace. The ash is characterized by a computer-controlled scanning electron microscope (CCSEM). Figure 10 clearly shows that low-melting-point carbonates (Ca, Fe, Mg) are generated under oxyfuel conditions, which indicates enhanced slagging. At the same time, more silicates and aluminosilicates are generated, which have medium melting points and indicate enhanced coalescence of minerals due to high carbon monoxide yield during char oxidation.

The generation and elimination of sulfur trioxide (SO<sub>3</sub>) is another critical issue for oxyfuel combustion, as SO<sub>3</sub> can cause corrosion in the combustion chamber, flue gas duct, coal mill, and so on. Due to the enrichment effect of flue gas recycle, the SO<sub>3</sub> concentration in oxyfuel combustion is usually 3–5 times greater than in air firing, which increases the acid dew point of flue gas from around 400 K to 430 K.

The conversion of sulfur dioxide to sulfur trioxide has been well studied for air firing. Sulfur dioxide can be converted through high-temperature gaseous oxidation or mineral

catalyst conversion. If the selected catalyst reduction (SCR) method is used to control nitrogen oxides, most of the SO<sub>3</sub> is generated via the SCR catalyst. Normally, 1%–5% of SO<sub>2</sub> is converted to SO<sub>3</sub> in both air-firing and oxyfuel combustion. That is, the higher SO<sub>3</sub> concentration in oxyfuel combustion is due to the higher SO<sub>2</sub> concentration. No evidence exists of a higher conversion rate under oxyfuel conditions. Our recent study even shows that the conversion ratio of SO<sub>2</sub> to SO<sub>3</sub> by an SCR catalyst is significantly decreased in a high-CO<sub>2</sub> environment (Figure 11), due to competition between CO<sub>2</sub> and SO<sub>2</sub> at the active absorption site on the SCR catalyst. Therefore, for a compatible air combustion/oxyfuel combustion power plant design that includes SCR, which is necessary to meet strict environmental legislation, SO<sub>3</sub> corrosion may be more severe than for a power plant design that does not include SCR.

Modern wet flue gas desulfurization (WFGD) can effectively remove SO<sub>2</sub> at efficiencies of up to 95%–99%. Therefore, for coal with a sulfur yield higher than 1%, the flue gas should recycle after WFGD in order to decrease SO<sub>2</sub>/SO<sub>3</sub> concentration, and thus the potential for corrosion. Another practice that is usually adopted in order to inhibit corrosion is to reduce the moisture content of the flue gas, using flue gas condenser (FGC), which cools and dries the flue gas to temperatures around 300 K. However, the use of WFGD and FGC results in relatively high investment and operation costs; therefore, alternatives are required. In-furnace injection of a calcium-based SO<sub>2</sub> sorbent and the injection of sodium-based or calcium-based SO<sub>3</sub> sorbents before the flue gas pre-heater can be a cheaper choice.

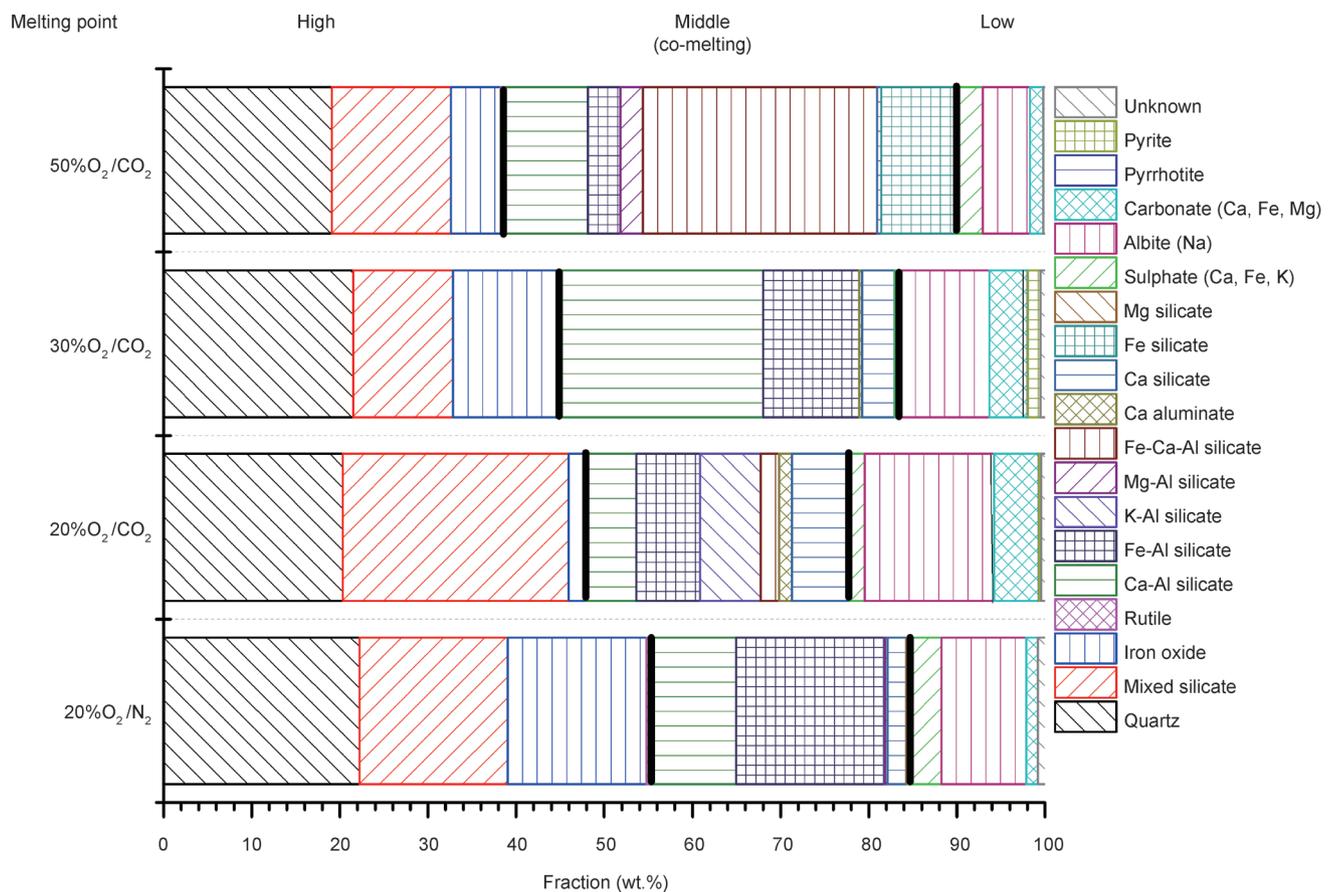


Figure 10. Weight fraction of various mineral associations for ZD coal under air-firing and oxyfuel combustion.

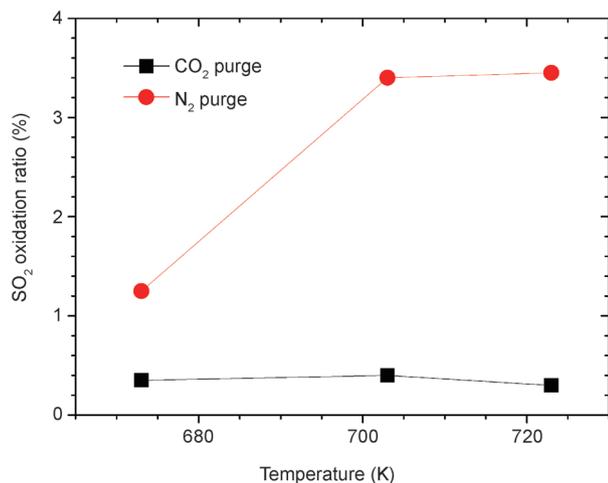


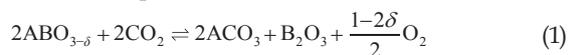
Figure 11. SO<sub>2</sub> conversion ratio under nitrogen and carbon dioxide purge [40].

### 3 Next-generation oxyfuel combustion

#### 3.1 A novel air separation concept

An air separation unit (ASU) provides the oxygen for combustion in an oxyfuel power plant. The current state of technological development necessitates the application of a cryogenic distillation unit. Preliminary calculations indicate that this operation alone requires about 60% of the power consumption for carbon capture, and reduces the overall efficiency of the power plant by 7%–9% [41]. Further studies have been conducted to reduce ASU energy consumption by investigating the use of high-temperature looping technology for oxygen production as an alternative to conventional cryogenic distillation methods. A new application of perovskite-type oxides as oxygen carriers provides pure O<sub>2</sub> or O<sub>2</sub>/CO<sub>2</sub> gas streams for oxyfuel combustion [42]. This oxygen production process consists of two main steps, as shown in Figure 12: ① oxygen adsorption; and ② oxygen desorption.

In the first step, air is used as a feed gas to saturate the perovskite oxygen carrier with O<sub>2</sub>; in the second step, CO<sub>2</sub> is used as a sweep gas to desorb O<sub>2</sub> from the perovskite, producing an O<sub>2</sub>-enriched CO<sub>2</sub> flue gas stream. The adsorption/desorption process of perovskite-type oxygen carriers is reversible and can be expressed as follows:



Several types of perovskite oxygen carriers have been investigated for oxygen production application [43–45]. The major problem encountered with these perovskite materials is their relatively low oxygen desorption ability. Recently, Shen et al. [46] developed a novel perovskite-type oxygen carrier, Ba<sub>1-x</sub>Sr<sub>x</sub>Co<sub>0.8</sub>Fe<sub>0.2</sub>O<sub>3-δ</sub>, which had an excellent cyclic stable oxygen-production performance. However, current investigations into perovskite looping for oxygen-production technologies are still at the conceptual or laboratory scale.

#### 3.2 A new concept: Moderate or intense low-oxygen dilution (MILD)-oxy combustion

As mentioned earlier, although oxyfuel technology is widely studied and applied, there are still drawbacks to be overcome.

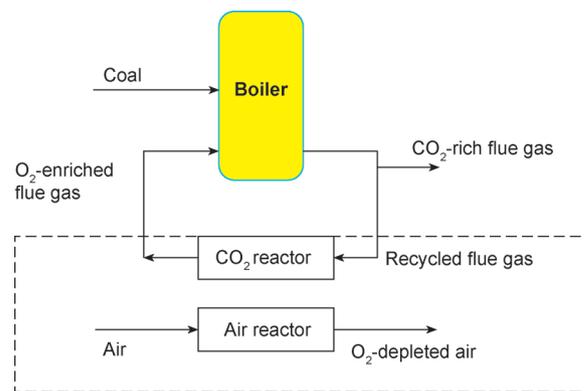


Figure 12. Simplified schematic diagram of perovskite-type oxides for O<sub>2</sub>/CO<sub>2</sub> production for oxyfuel combustion.

Firstly, an oxyfuel flame is less stable than a conventional air flame. Secondly, even though gross emission is substantially reduced in oxyfuel combustion, NO<sub>x</sub> concentration in the exhaust gas is relatively high due to nitrogen leakage and accumulation in the process of flue gas recirculation. Thirdly, recirculation decreases the overall systematic efficiency.

MILD combustion, also known as flameless combustion, is a high-efficiency combustion technology characterized by near-isothermal distribution across the furnace, and by high reactant temperature but low temperature increment in the reaction zone [47, 48]. Typically, in order to establish MILD combustion, the reactant temperature must exceed the fuel auto-ignition temperature, and the reactants must be diluted past the flammability limit [49–51]. Given these limitations, the combination of oxyfuel combustion and MILD combustion—or MILD-oxy combustion—is a novel concept in this field, and has these advantages:

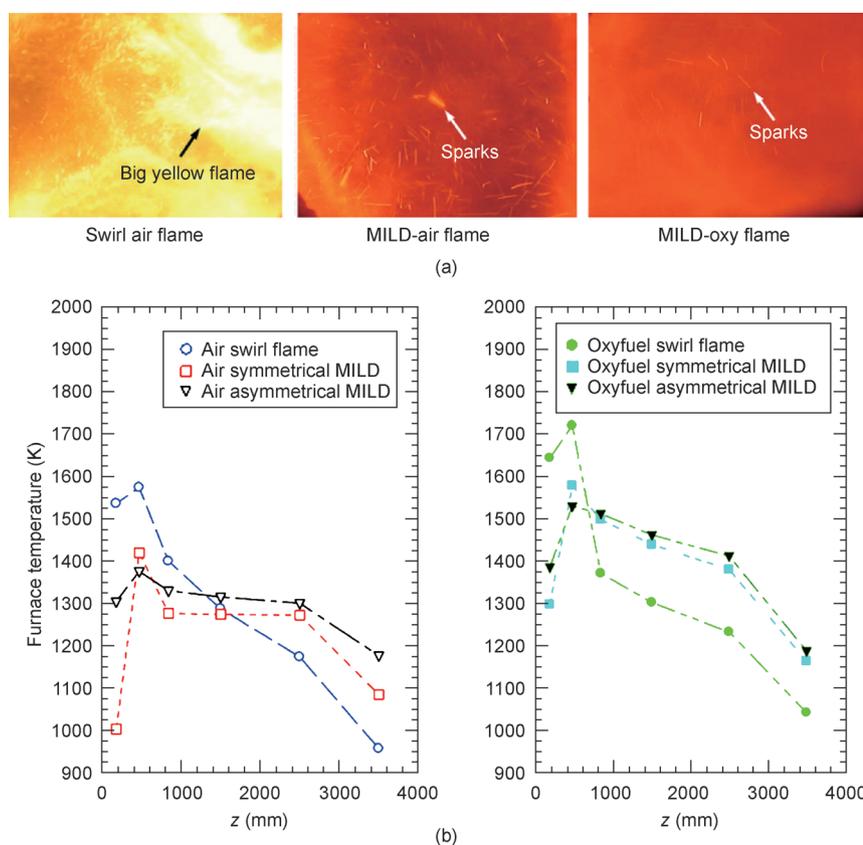
- (1) The high-temperature reactant and uniform temperature distribution of MILD combustion can improve oxyfuel flame stabilization.
- (2) NO<sub>x</sub> formation is highly suppressed, due to the intense oxygen dilution and low temperature increment in MILD combustion.
- (3) Overall thermal efficiency is greatly promoted by utilizing hot recycled flue gas for MILD combustion.
- (4) CO<sub>2</sub> dilution tends to eliminate the visible flame, and CO<sub>2</sub> participates in the reactions, rather than remaining inert.

Taking both MILD combustion and carbon capture into account, Li et al. [52] investigated the characteristics of the MILD-oxy combustion of different gaseous fuels in a recuperative furnace. They concluded that the replacement of N<sub>2</sub> by CO<sub>2</sub> can promote the establishment of MILD combustion, and that the region of the MILD combustion becomes wider as the reactant is diluted by high-concentration CO<sub>2</sub>. MILD combustion occurs without preheating as long as a sufficiently high reactant dilution is achieved under fixed CO<sub>2</sub> and O<sub>2</sub> concentrations.

Coal has less reactivity and mobility than gaseous fuels, due to its solid nature, so it is usually more difficult to achieve MILD-oxy combustion of pulverized coal than of gaseous fuels. Stadler et al. [53] tested a novel approach for MILD-oxy combustion of pulverized coal by increasing

the oxidizer ( $O_2/CO_2$ ) injection velocity up to  $100\text{--}280\text{ m}\cdot\text{s}^{-1}$ . They found that coal MILD-oxy combustion can be established without oxidizer preheating, that is, at ordinary temperatures, when the entrainment of burned gas caused by high-speed injection is sufficient strong. They also discussed the effect of gasification reactions on char reactivity, burnout, and species in the MILD-oxy combustion of pulverized coal. Saha et al. [54] investigated the MILD-oxy combustion of pulverized coal in a recuperative furnace. Their results showed that by means of the internal recirculation of hot burned gas in the recuperative furnace, the MILD-oxy combustion of both high and low rank coals can be successfully established without preheating, given an appropriate jet velocity, which is reported as  $86.2\text{ m}\cdot\text{s}^{-1}$  in the study.

It is worth noting that an experimental investigation [55] on the MILD-oxy combustion of pulverized coal at ordinary temperatures was conducted at a 0.4 MW pilot-scale facility at HUST. MILD-oxy combustion of pulverized coal was successfully achieved (Figure 13) without highly preheating the oxidant. The integral system of flue gas recirculation was applied in this experiment, including ash separation and water vapor condensation. This result indicates the feasibility of an industrial application for this technology.



**Figure 13. Experimental results of MILD-oxy combustion of pulverized coal in a 0.4 MW facility at HUST [55].** (a) Flame images for different combustion modes; (b) temperature distribution along the furnace centerline.

## 4 Summary and future challenges

After 30 years of development, oxyfuel technology has matured; it now possesses the fundamental characteristics necessary for commercial application. Most importantly, oxyfuel combustion is suitable for use in the large number of existing coal-fired power plants in China. The development of oxyfuel combustion technology in China keeps in step with international developments, and the fundamental studies described here provide a useful and invaluable reference for the design of key equipment, operation modes, industrial system flow, and more.

In order for China's coal-power-dominated energy mix to achieve greenhouse

gas emission reduction targets, large-scale demonstrations must be launched as soon as possible to increase the likelihood of commercializing oxyfuel technologies. At the same time, in order to reduce the high cost of this  $CO_2$ -capture technology, this novel concept and its methods must be strongly promoted and thoroughly developed.

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## Compliance with ethics guidelines

Chuguang Zheng, Zhaohui Liu, Jun Xiang, Liqi Zhang, Shihong Zhang, Cong Luo, and Yongchun Zhao declare that they have no conflict of interest or financial conflicts to disclose.

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