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Scale-up challenges and opportunities for carbon capture by oxy-fuel circulating fluidized beds

Sadegh Seddighi^a, Peter T. Clough^b, Edward J. Anthony^{b,*}, Robin W. Hughes^c, Ping Lu^d

^a Department of Mechanical Engineering, K.N. Toosi University of Technology, Tehran, Iran

^b Department of Energy and Power, Cranfield University, Cranfield, Bedfordshire MK43 0AL, UK

^c Natural Resources Canada, CanmetENERGY, Ottawa K1A 1M1, Canada

^d School of Energy & Mechanical Engineering, Nanjing Normal University, 78 Bancang Street, Nanjing 210042, China

HIGHLIGHTS

- An extensive review of the current status of oxy-fuel CFBC combustion has been written.
- Issues related to the scale-up of the technology, and the appropriate modelling approaches are discussed.
- The potential of pressurized oxy-fuel FBC is explored.
- Emissions and pilot plant performance are exhaustively examined.

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ABSTRACT

Oxy-fuel combustion is a promising technology for carbon capture and storage (CCS) from large point sources. In particular, fluidized bed (FB) boilers represent one of the power generation technologies capable of utilizing the oxy-fuel concept. This paper reviews the published material on the key aspects of oxy-fuel circulating FB, including the boiler heat balance, heat transfer mechanisms, furnace hydrodynamics, and the mechanical and chemical mechanisms of the process. In particular, it demonstrates the challenges of utilizing high inlet O_2 concentrations in the oxy-fuel process in fluidized beds. This requires significantly more efficient gas-particle clean-up technology (especially for Cl with perhaps 19% retention and Hg with 2.15 µg/m³ in flue gases), high circulating solids flux and, hence, significant heat extraction outside the furnace (up to 60% of the boiler's total heat extraction). Scale-up of oxy-fuel CFB technology can partially compensate for the energy penalty from air separation by furnace downsizing when operating at high inlet O_2 concentrations. Critically, while there are numerous measurement campaigns and corresponding models from the pilot and, to a lesser extent, industrial scale, the paper endeavors to answer the questions about what information taken from such experimental campaigns is reliable, useful for future design, and for scale-up.

1. Introduction

The Fifth Assessment Report of the United Nations Intergovernmental Panel on Climate Change [1] notes that human impact on the climate system is certain, and recent anthropogenic emissions of greenhouse gases (GHGs) are the highest in recorded history leading to global warming, sea level rises and more frequent weather-related disasters. In particular, CO_2 emissions have risen sharply and are the main cause of and contributor to global warming [2]. Fig. 1 shows the elevation in anthropogenic CO_2 emissions since 1850, indicating a dramatic increase since ca. 1950, which was due to the rapid global economic growth after the Second World War. It is now widely accepted that action against unabated emissions of GHGs must occur, in order to minimize the damaging effects of climate change.

The main methods for reducing CO₂ emissions are:

- Carbon capture and storage (CCS) [3]
- Utilization of fuels with low C/H ratio such as natural gas [4]
- Improving energy efficiency, thus utilizing less fuel [5]
- Substituting fossil fuels with renewable or nuclear energy sources

* Corresponding author.

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E-mail addresses: sadegh.seddighi@kntu.ac.ir (S. Seddighi), p.t.clough@cranfield.ac.uk (P.T. Clough), b.j.anthony@cranfield.ac.uk (E.J. Anthony), robin.hughes@cranda.ca (R.W. Hughes).

Symbols	s and notation	$U_O \ U_{mf}$	Superficial air velocity Minimum fluidization velocity
С	Concentration	z_{gd}	Distance between the injection plane and sampling plane
d_p	Particle diameter		
D_{g}	Radial dispersion coefficient	Greek let	tters
G_i	Mass flux of species i		
g	Acceleration due to gravity	μ	Viscosity
L	path length	$\rho_{\rm f}$	Fluid density
Р	Density	$\rho_{\rm P}$	Particle density
r	Distance from combustor centreline	φ	Voidage at minimum fluidization



Fig. 1. Global anthropogenic CO₂ emissions from fossil fuel combustion, cement production and flaring in comparison to forestry and other land use [1].

[4,6,7]

Carbon capture and storage is a bridging technology permitting a smoother socio-economic shift from fossil fuels to renewable energy sources. The main capture technologies in CCS are post-combustion, pre-combustion and oxy-fuel combustion.

Post-combustion CCS relies on removal of CO_2 from the flue gases after combustion; however, since the concentration of CO_2 in the exhaust gases is relatively low (usually 7–14 vol% for coal-fired boilers and below 4 vol% for gas-fired boilers), the cost of CO_2 separation is high and imposes efficiency penalties of ~8–12 percentage points [8,9].

In pre-combustion CCS the fuel is gasified or reformed, and the CO_2 is then removed from the produced gas before it is combusted or further processed for another use. A significant benefit of pre-combustion CCS technologies is that the CO_2 concentration is typically far greater than 20 vol% (after water-gas-shift processing) and thus the separation of

 CO_2 is more economical than for post-combustion systems, with efficiency penalties of ~7–9 percentage points [8–10].

Finally, oxy-fuel CCS is the combustion of fuel in a mixture of almost pure O₂ and recirculated flue gases (RFG, predominantly composed of CO₂ and steam), resulting in a flue gas consisting mainly of CO₂ and steam, making the separation of CO₂ relatively simple. An air separation unit (ASU) produces O₂ cryogenically and the choice of O₂ purity significantly affects CO₂ purity, plant capital cost and operating power consumption [11]. Fig. 2 shows a generalized schematic of the oxy-fuel CCS process, detailing the ASU, combustion boiler and RFG lines. Oxyfuel circulating fluidized beds (CFB) for CCS typically have an efficiency penalty of ~10 percentage points [9] and, therefore, a new focus of current research in the oxy-fuel area is on pressurized oxy-fuel CCS technologies which should lower the intrinsic efficiency penalty.



Fig. 2. Generalized schematic of the oxy-fuel CCS process [12].

Table 1

Jxy-ruel CFB experimental units.					
Unit scale, Location	Years of operation	Unit thermal power	Aim of research	Type of fuel	References
Industrial-scale, CIUDEN, Spain	2011-2014	30 MW _{th}	Oxy-fuel CFB operation, sulphur capture potential	Anthracite, petcoke, sub-bit. coal and biomass	[24,36,37,47–51]
Industrial-scale, Valmet, Finland	2013-present	4 MW _{th}	Oxy-fuel CFB operation, progress of combustion, heat transfer properties, safety	Bituminous coal	[13,18]
Industrial-scale, "La Pereda" power plant	2011-present	$1.7 MW_{th}$	Unit operation and experimental validation	Coal	[52-54]
Pilot-scale, CanmetENERGY, Canada	2011-2017	0.8 MW _{th}	Oxy-fuel CFB operation, combustion characteristics, pollutant formation	Coal, petroleum coke and lignite	[39,40]
Pilot-scale, University of Utah, USA	2011- present	$0.33 \text{ MW}_{\text{th}}$	SO ₃ formation under oxy-fuel conditions	Low-sulphur bituminous coal	[55]
Pilot-scale, University of Stuttgart, Germany	2014- present	$0.15 \mathrm{MW}_{\mathrm{th}}$	Solid burnout and emission of CO and NO _x	Bituminous coal	[26]
Pilot-scale, VTT, Finland	2013- present	$0.1 \ \mathrm{MW}_{\mathrm{th}}$	Evaluation of combustion and NO _x , SO _x and other emissions	Bituminous coal	[17,57]
Pilot-scale, Czestochowa University of Technology, Poland	2010- present	0.1 MW _{th}	Evaluation of combustion and $\mathrm{NO}_{\mathrm{x}}, \mathrm{SO}_{\mathrm{x}}$ and other emissions	Bituminous coal	[19]
Pilot-scale, CanmetENERGY, Canada	2004- present	0.1 MW _{th}	Investigation of a sh properties, Effect of steam on NO_{x} and SO_{x} formation	Coal and petroleum coke	[58,59]
Pilot-scale, Chinese Academy of Sciences, China	2015- present	0.1 MW _{th}	NO_x , SO_x and evaluation of other emissions	Bituminous coal	[60,61]
Laboratory-scale, Southeast University, China	2011- present	0.05 MW _{th}	Warm file gas recycle operation, effect of O_2 concentration on combustion	Coal and petroleum coke	[62,63]

1.1. The oxy-fuel process

The inlet O₂ concentration is key for scale-up of the oxy-fuel process. Utilizing a higher inlet O₂ concentration while keeping the furnace geometry constant leads to higher thermal output per furnace crosssectional area, improving the economics as it increases the process efficiency. For new facilities, however, it is more likely that a higher inlet O₂ concentration allows a smaller boiler size, with reduced capital costs [13–16]. It has been shown that higher inlet O₂ concentrations also lead to lower CO emissions [14,17-19], improved desulphurization efficiency [14], and a reduced power requirement for the flue gas recycle (FGR) blower [20]. Nonetheless, higher inlet O₂ concentration also lead to constraints in heat extraction from the boiler (influenced further by the smaller boiler size) [16,21], an increased risk of bed agglomeration within the furnace [21], and if the O_2 concentration increases beyond \sim 27 vol%, a significant increase in piping costs due to the requirement for somewhat specialized materials. An important consequence of reduced boiler size when utilizing elevated O₂ concentrations is that the solids inventory is smaller, which can result in a greater variation in the CFB bed temperature since the bed mass acts to dampen temperature changes due to its thermal capacity [22].

In this context, it is important to note that the inlet and exit O_2 concentrations are among the major control parameters in CFB oxyfired boilers [23]. Lappalainen et al. [24] concluded that FGR leads to higher perturbations in the furnace O_2 content due to the lack of control of the O_2 supply from O_2 injection, FGR and fuel O_2 . One requirement identified in controlling the O_2 content within the furnace is the online measurement of the time-dependent O_2 concentration in the RFG, and consequently the required adjusted flow of inlet O_2 in order to meet the desired air/fuel ratio [25].

Given the importance of inlet O_2 concentration, the present work focusses on this parameter as a key variable. Information on oxy-fuel pulverized combustion (PC) boilers can be found in Scheffknecht et al. [26], Toftegaard et al. [27], Chen et al. [28], Wall et al. [29] and Yin and Yan [30]. Unfortunately, the differences between oxy-fuel CFB and oxy-fuel PC make them of less relevance to CFB operation. There are far fewer reviews on oxy-fuel CFB than on oxy-fuel PC. One example is by Mathekga et al. [31], and another is by Singh and Kumar [32] focusing on the current status and experimental results from small oxy-FBC beds. However, the present review looks at the challenges and opportunities for process improvement through scale-up by reviewing the existing literature in oxy-fuel CFB modelling, heat transfer phenomena, fluidbed dynamics, and pollutant emissions.

The work provides a comprehensive (fluid dynamics, combustion, and heat transfer and emission formation) assessment of the oxy-fuel fluidized bed scale-up process, not addressed in previous reviews. Given that oxy-fuel CFB is a major carbon capture route for large coal power plants, such information is of vital importance for furnace designers and fluidized bed research and development. In particular, the authors evaluate: (1) how the available models developed for lab-, pilot- and large-scale oxy/air-fired CFBs can be adapted for modelling large-scale oxy-fuel CFB units; (2) the major challenges and opportunities in the design and scale-up of utility-scale oxy-fuel CFB boilers; and (3) the important parameters for designing large-scale oxy-fuel CFB boilers.

2. Scale-up and design

Both experimental work and modelling simulations are pivotal in the design and scale-up of oxy-fuel technology. To date, no commercial-scale (> 300 MWe) oxy-fuel CFB boiler has been built despite the technology currently having a Technology Readiness Level (TRL) of 7–8 [33]. The failure to build large-scale oxy-fuel plant arises because of a lack of effective carbon pricing, and an absence of government motivation to tackle climate change, and the scale and inherent inter-dependence of investors (private plant operator with publicly owned transport and storage network) required to get such a project up and

running [34]. Furthermore, where CCS plants have been deployed, they have typically utilized post-combustion amine scrubbing systems because of their previous demonstration at scale (such as the Niederaussem project, Dong power plant in Denmark, and the Canadian SaskPower's Boundary Dam project [35]) and the larger efficiency penalty incurred by conventional oxy-fuel technology. As a result, the scale-up of novel oxy-fuel technologies such as pressurized oxy-fuel boilers is receiving attention.

2.1. Industrial-, pilot- and lab-scale oxy-fuel CFB

Table 1 shows the major experimental oxy-fuel CFB units and indicates the extensive research carried out to date. The largest tested oxy-fuel CFB unit so far is CIUDEN (City of Energy), an industrial-scale 30 MW_{th} unit situated in northwest Spain [36,37]. Other large-scale units in operation include: a 4 MW_{th} oxy-fuel CFB demonstration plant at Valmet, Finland [13,18], an oxy-fuel CFB calciner utilized as part of a 1.7 MW_{th} post-combustion calcium looping CCS plant at La Pereda, Spain [38], and a 0.8 MW_{th} testing plant built at CanmetENERGY, Canada [39,40]. These units have all undergone major experimental campaigns focussing on the different aspects of oxy-fuel CFB operation including combustion, emissions formation and heat transfer. Examples of the scale-up process for air-fired CFBs can be seen elsewhere: Glicksman [41], Glicksman et al. [42], Zlokarnik [43], Knowlton et al. [44], and Leckner et al. [45,46].

A major difference between industrial-scale and smaller-scale units is the degree of lateral gas mixing, localized air/fuel ratios, and heat distribution within the furnace, which is influenced by the furnace width. For lab- and pilot-scale units (see Fig. 3) the furnace width is typically less than 0.2 m, while industrial-scale units can be 1 m or more and, thus, gas mixing in lab-scale units is effectively perfect [18].

2.2. Existing models for oxy-fuel CFB

The application of observed trends and correlations, and measured parameters obtained from lab- and pilot-scale units is often unsuitable for the design and operation predictions of large-scale units due to the greater complexity of larger systems over smaller systems [66]. It is necessary to be cautious when assessing the degree to which data obtained from the smaller-scale units can be utilized for larger-scale units [67].

Various research groups have developed oxy-fuel models to properly understand and analyze their experimental data. Thus, the modelling scope and results are largely dependent on the type and scale of the experimental setups and their corresponding data. In general, labscale FB units are 1D and thus their corresponding extracted models are more suitable for combustion chemistry. On the other hand, pilot-scale units give valuable sets of data in relation to both axial and lateral profiles leading to models capable of comprehensive 3D analysis of oxyfuel FB processes. There are several active groups developing comprehensive mathematical models for oxy-fuel CFBs using data from experimental units, including: Myöhänen et al. [57,68,69]; Seddighi et al. [13,17,18,65]; and Krzywanski et al. [70,71]. Another modelling approach utilizes commercial computational fluid dynamics (CFD) codes modified for oxy-fuel CFB boilers, such as work by: Zhou et al. [14]; Adamczyk [72]; and Amoo [73]. Table 2 presents the major modelling tools developed specifically for oxy-fuel CFB boilers.

2.3. Design scenarios for large-scale oxy-fuel CFB boilers

The design of large-scale oxy-fuel fluidized bed boilers has been of critical importance for minimizing the experimental costs of the scaleup process. Examples of the work on the design of oxy-fuel FB boilers can be found elsewhere [78,79]. The design of next-generation largescale oxy-fuel boilers is split between two pathways of: constant-furnace-size scenario; and constant-thermal-power scenario (a detailed discussion of these two scenarios is presented elsewhere [22]).

2.3.1. Constant-furnace-size scenario

The constant-furnace-size scenario is the only option for retrofitting air-fired CFB boilers, thus providing a near-term CCS implementation. Retrofit boilers are most attractive to the power sector since they can reuse most of the plant equipment and reduce investment cost/risk, making retrofitting oxy-fuel the most competitive technology option for CCS [28,80,81]. However, to economically retrofit an oxy-fuel system into an existing air-fired boiler, the original power plant must have sufficiently high efficiency (high enough to be capable of handling around 8% efficiency drop due to the energy penalty of the air separation unit (ASU)).

With the same furnace geometry, an oxy-fuel CFB boiler with the same O_2 concentration as an air-fired boiler, i.e., 21 vol%, gives a lower furnace temperature due to the specific heat capacity of CO_2 relative to N_2 . The same furnace temperature as an air-fired furnace can be achieved with an oxy-fuel furnace, but O_2 concentration of around 27–30 vol% is required [22]. By increasing the O_2 concentration, the boiler thermal power output changes, therefore necessitating additional heat removal via external heat exchangers [22]. An increased rate of circulating solids flux to improve heat removal ability also increases the efficiency of the boiler (see Fig. 4) and reduces the unburnt carbon content of the fly ash (see Fig. 5), both of which are economically and environmentally favorable [82].

2.3.2. Constant-thermal-power scenario

The constant-thermal-power scenario involves downsizing the furnace, as it is possible to achieve the same thermal power output with a lower total volumetric flow rate and an increased O_2 concentration. Modelling by Leckner and Gomez Barea [83] found the potential to reduce the boiler size by ~ 80% by increasing the oxygen concentration from 21 vol% to 80 vol%, for a 300 MW_{th} oxy-fuel CFB boiler, as shown in Fig. 6. In addition, a more homogeneous bed temperature profile and a lower heat flux to the boiler tubes, compared to the constant-size scenario, make the constant-thermal-power scenario a better pathway for oxy-fuel CFB CCS development for new facilities [22].



Fig. 3. Schematic of pilot-scale 100 kW_{th} (left) [17] and an industrial-scale 4 MW_{th} (right) [64] oxy-fuel CFB boilers (image is taken from [65]).

Table 2

Models previously utilized for oxy-fuel CFB boilers.

Developer	Focus of model	Type of model	Dimensions
Myöhänen et al. [57,68,69]	Comprehensive: combustion, heat transfer, fluid dynamics	Mathematical model	3D
Seddighi et al. [13,17,18,65]	Comprehensive: combustion, heat transfer, fluid dynamics	Mathematical model, MATLAB	3D, 1.5D
Krzywanski et al. [70,71]	Comprehensive: combustion, heat transfer, fluid dynamics	Mathematical model	3D, 0D
Zhou et al. [14]	Comprehensive: combustion, heat transfer, fluid dynamics	CFD, Fluent	2D
Adamczyk [72]	Fluid dynamics, combustion, heat transfer	CFD, Fluent	3D
Amoo [73]	Heterogeneous combustion, emissions production	CFD, STAR-CD	3D
Bolea et al. [74,75]	Boiler heat extraction modelling	Mathematical model	1.5D
Takkinen et al. [76]	Emissions modelling		1D particle model
Pikkarainen et al. [77]	Combustion modelling	Mathematical model	1D



Fig. 4. Effects of circulating solids flux (70 and 100% of maximum flux) on the combustion and boiler efficiency [82]. Maximum continuous rating (MCR) is the maximum thermal output of the boiler under continuous and normal operational conditions over one year.



Fig. 5. Effects of circulating solids flux (70 and 100% of maximum flux) on the unburnt carbon content in fly ash [82].

2.4. Pressurized oxy-fuel fluidized bed combustion

In order to improve the competitiveness of oxy-fuel fluidized bed combustion, R&D has started on pressurized oxy-combustion. This has a number of potential benefits including a further reduction in boiler size, increased net power plant efficiency, and the provision of alternative pathways for removal of impurities such as O_2 , CO, NO_x and



Fig. 6. Furnace size reduction with increase in inlet O_2 concentration for constant thermal power of 300 MW_{th} [83].

particularly SO_x [84–86]. The challenges faced by previous air-fired pressurized fluidized bed combustion (PFBC) demonstrations [87–90] have been recognized by the research community and thus configurations that do not include hot gas filters and gas turbines are being developed.

There are at least five global research groups developing oxy-PFBC technology including those in Canada (CanmetENERGY and the University of Ottawa) [91], in the UK (Cranfield University) [92], in the USA (Gas Technology Institute, GTI) [93,94], in China (Southeast University) [95], and in Poland (Czestochowa University of Technology, AGH University of Science and Technology, and the Institute for Chemical Processing of Coal) [96–98]. The largest oxy-PFBC pilot plant built to date (1 MWth), in collaboration with GTI and Linde, is located at CanmetEnergy in Ottawa. The facility includes CO₂ purification equipment for removal of NOx, SOx, and oxygen.

3. Heat transfer challenges

Under oxy-fuel conditions, the specific heat of the furnace gas is higher than under air-fired conditions due to the higher concentrations of CO₂ and H₂O. A higher specific heat capacity of the mixed gas lowers the furnace temperature and alters the heat removal duties [13,74,75,99]. However, it has been demonstrated that raised inlet O₂ concentration can considerably increase the furnace temperature [13]. The main research questions in relation to heat transfer in oxy-fuel CFBs are: "What is the most appropriate mechanism for heat removal?" and "How is the heat/temperature distributed within the overall system?"

Within the constant-furnace-size scenario, if the inlet O_2 concentration rises above 60 vol%, the thermal power output theoretically can be tripled for an equivalent size combustor [22]; however, the furnace heat extraction area and, consequently, the maximum extractable heat from the furnace are limited. Thus, above a certain inlet

 O_2 concentration, heat must come from the CFB return leg or through external heat exchangers in order to compensate for the increased thermal power output [13,16,100]. Figs. 7 and 8 show the increase in the share of heat extraction required within the return leg and external heat exchangers due to an increase in O_2 concentration. The extent to which heat can be usefully obtained from the solids within the external heat exchanger is unclear and depends on circulating solids flux and heat transfer rates [40].

So far, the only way to utilize high O2 inlet concentrations is to increase the circulating solids flux, requiring dramatically higher heat extraction requirements in the external heat exchanger. For example, a 324 MW_{th} furnace with 27 vol% O₂ inlet concentration requires around 240 kg/s of circulating solids, but for a similarly sized unit operating at 70 vol% O₂ inlet concentration, the capacity becomes 1079 MW_{th}, requiring ~3000 kg/s of circulating solids [13]. Therefore, when maintaining the furnace size, the heat balance often necessitates a very high circulating solids flux. Higher O2 inlet concentrations can have dramatic impacts on the boiler's fluid dynamics, combustion efficiency, heat transfer requirements and effectiveness, and can increase erosion due to the higher solids flux. Thus, for the retrofit scenario, inlet O₂ concentrations are limited to below 40 vol% [22]. Therefore, in order to utilize greater inlet O2 concentrations, a new CFB design is required which is capable of operating with a high circulating solids flux and with large heat extraction duty within the external heat exchanger.

Effective design and scale-up of heat transfer equipment depend on heat transfer modelling which itself requires accurate empirical data and experimental data obtained under realistic conditions [101–107]. A major challenge in the design and scale-up of oxy-fired systems is the unrepresentative results obtained from the use of heat transfer models which apply data derived from lab-scale units [105]. In part, the inadequacies of lab-scale units are due to their aspect ratio being much larger than industrial- and large-scale furnaces, leading to heat transfer correlations which are unreliable at larger scales. Furthermore, the knowledge, experience and correlations gained from air-fired CFB conditions can be unreliable for oxy-fuel CFB due to significantly different solid fluxes. Thus, critical evaluation of available heat transfer models is paramount.

Numerous fluidized bed heat transfer models with different structures and assumptions exist [108]. The most important parameters in modelling heat transfer in a CFB furnace are: the solids suspension density; particle size; bed temperature; particle specific heat; and hydrodynamic conditions in the furnace, which are typically assumed to involve a core-annulus structure [107].

3.1. Convection

Convection in the CFB context refers to: (1) convection from gases to heat transfer surfaces; and (2) conduction from solid particles to the heat extraction surfaces. These two mechanisms can be modelled separately [109–113] or together in a single convective heat transfer coefficient [13,114,115].

3.2. Radiation

Radiation is the main heat transfer mode under oxy-fuel CFB conditions; however, some experimental results have shown that the radiative heat transfer coefficient decreases dramatically when suspension density exceeds 5 kg/m³ [13,73,116] due to the increased absorption of the heat by the particles. Radiative heat flux in oxy-fuel combustion can be enhanced due to higher-temperature combustion; Bordbar and Hyppänen [117] found up to a 40% increase in furnace radiative heat flux when changing from air-fired to oxy-fuel combustion (Fig. 9). The spatial radiative heat flux variations observed in Fig. 9 result from multiple cyclones, which allow the gas to bypass before injection to the furnace, in addition to the flow oscillations due to bubbles rupturing above the dense bed. With regard to heat radiation in CFB furnaces, two calculation methods that can be used are Monte Carlo analysis and the net radiation methods [118]. The Monte Carlo approach is based on statistical features relating to the physical phenomena where the radiation is simulated by modelling stochastic paths of photon bundles leaving and reaching the combustor wall [119–121]. Although it is a mature method, it requires extensive calculation resources for complex systems and geometries [122]. This is especially so for the simulation of oxy-fired systems with high solids flux and hence greater complexity.

The required heat extraction duty from a furnace is usually overestimated due to the high level of uncertainty linked to the expected heat losses of the overall system [13,123]. In addition, the uneven solid flow rate within the furnace leads to errors in the measurements and modelling of the heat flux distribution [124].

3.3. Furnace temperature

Control of temperature levels in oxy-fired CFB furnaces is a critical issue for their scale-up [83,125]. The temperature profile is largely homogeneous in air-fired CFB furnaces, but under oxy-fuel conditions with increased inlet O_2 concentration, can lead to large, localized gradients in the furnace temperature profile due to hotspots [13,126].

The furnace temperature of oxy-fuel CFBs can be lower than those of air-fired CFBs for equivalent inlet O_2 concentrations due to the higher heat capacity of the gas in the oxy-fuel atmosphere, caused by the recirculation of CO_2 [13,24,73,127]. The steam content of oxy-fired CFB furnaces will vary due to the use of wet or dry FGR and, in turn, higher furnace steam concentrations can reduce the furnace temperature [127].

Fig. 10 shows the effect of the recirculation rate on the temperature distribution in an oxy-fired CFBC furnace [82]. Increasing the fly ash recirculation rate from 0 to 8 t/h leads to a reduction of the bed temperature from 1233 K to 1153 K, but also causes the furnace exit temperature to rise from 1033 K to 1083 K. The primary reason for the temperature change is the transfer of more heat from the bed's dense phase to its dilute phase.

High-temperature oxidation under oxy-fuel conditions is particularly problematic for furnaces since it leads to fireside corrosion from the reactions between the reactor internals and the surrounding hot gaseous environment. This can eventually lead to the failure of boiler tubes, superheaters, reheaters, and water walls by typical metal loss mechanisms or by the generation of cracks [128]. In addition, the corrosion in oxy-fuel furnaces can be intensified due to the FGR producing elevated concentrations of corrosive gases (HCl and SO₂) [128–130]. Syed et al. [128] and Hussain et al. [131] both found



Fig. 7. Change in boiler heat extraction duty from air-fired to oxy-fuel (70 vol% O₂) CFB according to data from Alstom and U.S. Department of Energy [100].



Fig. 8. Increased share in loop seal heat extraction duty when increasing the O₂ concentration in oxy-fuel CFB. H_{WW} refers to heat extraction from water walls, H_{seal} is heat extraction from loop seals, H_{flue} is heat extraction from flue gases and H_{panel} refers to heat extraction from wing and division walls. Information on right of the graph refers to the heat extraction from a typical air-fired CFB boiler which is taken as reference case [16].



Fig. 9. Radiative heat flux for air-fired (diamond symbols) and oxy-fired (solid line) combustion [117].



Fig. 10. Effect of recirculation rate on temperature distribution in an oxy-fired CFB furnace [82].

thicker ash-oxide layers on the combustor internals under oxy-fuel conditions compared to air-fired conditions, up to an $\approx 40\%$ increase. They also found that sulphur penetrated deeper into the oxide layer under oxy-fuel conditions, leading to higher levels of S-stabilized corrosion compounds such as alkali-iron tri-sulphates.

3.4. Maximum heat flux

The maximum heat flux to the boiler water tubes is of great significance for the furnace design and is used for selecting the materials utilized for the construction of the water tubes [13,132,133]. Heat flux prediction and modelling become more important in large-scale supercritical units, where the values of heat flux should be designed to avoid steam generation in water tubes [134]. The furnace maximum heat flux must be designed to ensure a reasonable safety margin before reaching the critical heat flux, to avoid vapor films, overheating and potentially the rupture of the water tube walls [13,123,135]. The location of the maximum heat flux depends on the fuel type, thermal power, secondary gas injection arrangements and the heat extraction panel arrangement [136].

For air-fired CFB boilers with an electrical power generation capacity in the range of 1–200 MW_e, heat fluxes are typically found to be in the range of $120-150 \text{ kW/m}^2$, which can increase by a third for oxy-fuel conditions [13,114,117,137,138]. Fig. 11 provides a comparison of the reported maximum heat fluxes for CFB and PC units under both air and oxy-fired conditions.

The heat flux to the furnace water walls is much lower in a constantthermal-power scenario than in a constant-furnace-size scenario because the furnace net heat extraction duty is higher in the latter scenario at high O_2 concentration [22].

3.5. Slagging, fouling and sintering

Deposition of ash on heat transfer surfaces which occurs through slagging and fouling is a major cause of boiler tube and combustor wall damage [144]. Ash deposition has been widely explored for air-fired CFB, but little such data exist for oxy-fuel CFB conditions.

Ash deposition through slagging usually occurs in furnace locations with high temperatures while fouling typically occurs at lower temperatures. The co-utilization of coal and biomass has been studied by various groups such as [145–147] and can correspond to increased slagging and fouling on heat transfer surfaces [148]. Oxy-fuel conditions can increase the rate of ash deposition compared to air-fired conditions, owing to the gas physical properties, which lead to changes in deposition behavior [149,150]. In addition to an increased rate of ash deposition, the sulphur content of the ash deposits also increases under oxy-fuel conditions (Fig. 12), producing a higher risk of corrosion of fireside surfaces when approaching the acid dew point [151].

The deposition process varies largely among different ash particles depending on their composition and size. Once ash particles are deposited on the surface of a combustor component they can begin to



Fig. 11. Maximum heat fluxes to the furnace water walls reported for CFB and PC furnaces under air-fired and oxy-fuel-fired conditions [65]. Data presented from Wang et al. [139], Xu et al. [140], Seltzer et al. [141], Butler et al. [142], Huilin et al. [114], Sun et al. [137], II'chenko et al. [138], Seddighi et al. [13,143].



Fig. 12. Sulphur retention of the ash particles under oxy-fuel and air-fired conditions [149]. AAEM refers to alkali and alkaline earth metal species.

coalesce and fuse via sintering. Particle sintering typically occurs at temperatures lower than the melting temperature of the ash material, but higher than the Tamman temperature.

Like slagging, increased fouling can lead to the following issues [139,148,152]:

- Reduction of boiler efficiency and availability;
- Increase in boiler temperature due to lower heat extraction capability;
- \bullet Higher levels of NO_x emissions due to the promotion of thermal $NO_x;$
- Lower thermal power output of the boiler; and
- An increase in ash deposition due to the formation of alkali sulphates on the surfaces encouraging the collection and agglomeration of ash particles.

The increased furnace temperature of an oxy-fired furnace can also lead to the enhanced oxidation rate of boiler tubes which will increase their required replacement rate [153]. The frequency of boiler tube replacement is influenced by erosion and abrasion caused by solid particles impacting the surfaces and by oxidation and corrosion caused by higher oxygen partial pressures and ash-influenced corrosion [154].

A critical problem in CFB furnaces is the melting and sintering of solid bed particles, which increases at elevated temperatures and in the presence of alkaline earth compounds [23]. Ash melting temperature varies for different fuels: for biomass it typically occurs above 1250 K, for lignite above 1350 K and for hard coal above 1450 K [155–157].

Some key elements in ash contributing to corrosion in CFB boilers include Cl, Br, Zn and Pb [158]. In the upper part of the furnace, ash deposition is mainly of K-, Na- and $CaSO_4$ -derived materials. However, in the lower parts of the furnace alkali chlorides and bromides are found in the form of K and Na compounds and $CaSO_4$, while the heavy metals in ash found on water walls are typically in the form of Zn, Pb, and Cu sulphates and chlorides [158]. With this information, it is possible to design and implement suitable materials for the type of deposition expected at each location in the furnace. For a more complete discussion of corrosion in oxy-combustion systems and for suggestions regarding materials of construction, see elsewhere [159].

Iron oxide is an important contributor to slagging on heat transfer surfaces during coal combustion and its effects can be enhanced further by the formation of pyrrhotite (FeS_x) and FeO–FeS, which derive from pyrite and have low melting points and, therefore, enhance the slagging process [144,160–164]. In oxy-fuel conditions, pyrite undergoes faster oxidation due to the increased steam concentration, leading to the production of magnetite, which can impact ash deposition rates [144].

4. Fluid dynamics challenges

The operation of a large-scale CFB furnace consists of multiphase flow with interactions significantly influenced by the contact between the gas and solid phases. Fig. 13 shows a schematic of the main transport regions in a CFB loop.

4.1. Furnace flow characteristics

In a typical CFB furnace, the formation and break-up of particle clusters is the main characteristic of the flow at the meso scale; here we



Fig. 13. Regions of a CFB loop according to the distribution of the solids [65]. Arrows show the direction of solids motion.

refer to the meso scale as the length scale between the largest particle size and the diameter of the furnace. The dense-bottom/dilute-top and dense-wall/dilute-core structures are the characteristic of the flow at the macro scale [65,165]. Here we refer to the micro scale as that which is relevant to a length equal to or smaller than the particle diameter [166].

The furnace dense bed contains the major proportion of the solids and it is the region where most of the char particles burn [167,168], while the volatiles and lighter char particles predominantly combust in the O₂-rich bubble phase [17]. The rise of the bubbles generates the general upward motion of the solids; the bubbles rupture at the top of the dense phase – the so-called 'splash zone' – producing a further driving force for the upward motion of the solid particles [169]. The 'transport zone' is located above the splash zone and is characterized by a core-annulus structure and a dispersed outer phase which generally falls back into the bed resulting in back-mixing [168,170,171].

4.2. Circulating solids

Circulating solids play a key role in shaping the hydrodynamics of the furnace and temperature profile in the CFB loop. Circulating solids also transfer furnace heat to external heat exchangers, which are located in the loop seals of the return leg (Fig. 13).

Increasing the amount of circulating solids avoids excessive temperatures but can significantly increase operating costs. A major challenge for modelling, design and scale-up is that the quantity of circulating solids varies considerably among different boiler manufacturers and units [13]. While circulating solids have been reported as high as $25 \text{ kg/m}^2 \text{ s}$ [171,172], the typical value is less than $10 \text{ kg/m}^2 \text{ s}$ [16]. A higher circulating solids flux increases the heat extraction capability of the CFB and minimizes the amount of unburnt carbon in the fly ash [82]. Nevertheless, practical methods for increasing the circulating solids flux when utilizing lower gas flow rates are challenging; one potential solution would be increasing the efficiency of the cyclones and decreasing their cut-off particle diameter, thereby circulating more solids back into the furnace.

In CFB furnaces, particle segregation due to differences in particle size and density can have a major impact on flow behavior and reaction kinetics [173]. Within the fluidized bed itself there exists a certain amount of axial particle segregation that is caused by the smaller particles fluidizing and entraining within the gas stream more easily than the larger particles [173]. The concentration of solids and, hence, fuel in the dense zone of the fluidized bed can be around 1000 times that at the top of the bed [17]. Jang et al. [174] found that the average ash particle size under oxy-fuel conditions was smaller than in air-fired conditions due to a reduced ash particle growth mechanism caused by the different properties of the oxy-fuel gas.

A major problem for fluid dynamics scale-up is that the majority of experimental work in this field has mimicked air-firing conditions such as in [175]. However, simulation of the fluid dynamics and solid flows is different in oxy-fuel conditions compared to typical air-fired conditions due to the varying inlet O_2 concentrations and, consequently, differences in volumetric flow rates and FGR rates [24,50,51]. Another difference in the oxy-fuel CFB furnaces lies in their considerably higher solids fluxes when high inlet O_2 concentrations are specified, which leads to a shift toward the fast fluidization regime.

4.3. Mixing

Mass transfer phenomena can be divided into two categories: lateral (perpendicular to the flow); and axial (vertical movement of gas and solids within the combustor) mixing. In the lower section of the transport zone, the effects of solid-solid mixing are dominant, while in the upper section gas-gas mixing is more significant [176].

A major reason for studying mixing is that the reaction mechanisms and the combustion kinetics by themselves fail to predict the measured furnace gas profiles in both lab- and large-scale units [17,68,177]. Simulation studies often add adjustment coefficients to the reaction rates in order to take into account the mixing effects of the gas and solids [68,177,178]. Others reconcile the lack of exact kinetic data in largescale systems (partly due to the hefty calculation costs) by assuming that CFB combustion can be modelled as mixing-controlled combustion, thus lumping kinetics and mixing together [179].

4.3.1. Lateral mixing

In general, the extent of lateral mixing is better in lab-scale units than in large-scale units; therefore, information obtained from lab-scale experiments is less robust for direct application to large-scale units [17].

Lateral gas mixing in fluidized bed furnaces can be described by dispersion, which may be assumed to be the sum of the dispersion coefficients from large-scale structures (eddies - generated by bubble eruptions and localized pressure differences within the bed), and localized small-scale particle motions [180–183].

It has been demonstrated previously, that lateral gas mixing within the core region of an FBC can be modelled as turbulent flow within a pipe (Eqs. below) [181,183–185].

$$0 = -U\frac{\partial C}{\partial z_{\rm gd}} + D_{\rm g}\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial C}{\partial r}\right) \tag{1}$$

$$\frac{\partial G_{\rm i}}{\partial t} = D_{\rm g} \nabla^2 G_{\rm i} \tag{2}$$

where *r* is the distance from the combustor centreline, z_{gd} is the distance between the injection plane and sampling plane, G_i is the mass flux of species *i*, *C* is the tracer-gas concentration, D_g is radial dispersion coefficient and *U* is velocity. The two main approaches for increasing the gas dispersion coefficient and, hence, the degree of lateral gas mixing within the bed are: (1) increasing the concentration of solids; and (2) increasing the fluidization velocity. The method of increasing the furnace solids concentration works by increasing the fluidization velocity, but is limited as it requires considerable design changes to the existing large-scale boiler which is unlikely as the fluidization velocity utilized lies in a limited range [13].

Overall, the poor lateral gas mixing commonly associated with CFBs can often lead to maldistribution of the reactant gases and incomplete combustion [186]. The maldistribution of gas is greater under oxy-fuel conditions given the higher fuel feeding rates (in the cases where the geometry is kept constant), but the higher solids flux may positively influence lateral gas mixing. Improving the lateral mixing is important to ensure homogeneous heat distribution throughout the combustor, which in turn will minimize any localized hot spots that could lead to the formation of ash melts [18].

Table	3
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Modelling approaches for axial gas mixing in CFB boilers.

Unit power [MW _{th}]	Unit width [m]	Air-/oxy-fired	Description	Reference
0.1	0.08	Air and oxy-fuel	Semi-empirical relation used for axial gas mixing	[17]
4	1	Air and oxy-fuel	Semi-empirical relation used for axial gas mixing	[18]
12-300	1.5-8.5	Air	Adjustment coefficient applied to kinetic rates to fit measured gas concentration	[178]
15	3.6	Air	Adjustment coefficient applied to kinetic rates to fit measured gas concentration	[68]
-	0.8	Air	Adjustment coefficient applied to kinetic rates to fit measured gas concentration	[177]

4.3.2. Axial mixing

Prediction of the interaction between the solid and gas phases is critical in establishing effective axial mixing in fluidized bed boilers. The distribution of the phases within CFB furnaces will affect how the reactants mix. Table 3 presents some of the past approaches used for modelling axial gas mixing.

Axial gas mixing in a CFB furnace is often divided into three distinct zones. From the bottom to the top of the combustor, they are: (1) restricted mixing in the dense bed; (2) improved mixing close to where the bubble eruptions occur; and finally (3) restricted mixing in the transport zone [17]. Axial gas mixing is of course dependent on the furnace geometry, the properties of the solids and gases, and the operating conditions. Mixing in the dense phase of the CFB bed can be limited because of poor mixing between the combustible gas containing emulsion and the oxygen containing bubbles, but provided that there is sufficient fluidization mixing generally proceeds rapidly [187].

Secondary air injection has an important effect as this additional gas, injected at an angle into the furnace, will cause the eruption of bubbles into a relatively dense bed of material, improving axial gas mixing [17]. Secondary injection can dominate fluid dynamics across the entire cross-section of a lab-scale CFB furnace, while affecting only a portion of the injection plane in larger units [17,18]. Varol et al. [188], using a 30 kW_{th} CFB unit, suggested that the secondary air should be injected into the furnace at an angle of 10–15 degrees to the horizontal in the upward direction, to produce optimal mixing between the injected air and the furnace flow. While such studies are valuable in optimizing the properties of secondary gas injection, concerns about the generality of their conclusions is such that further measurements in industrial-scale oxy-fuel CFBs with high inlet O_2 concentrations are required.

4.4. Fluid dynamics scaling

Dimensional analysis is one of the most useful tools in providing scaling laws between lab-scale and large-scale units. Geometric and dynamic similarity are both important in scaling studies, while geometric similarity is a prerequisite for dynamic similarity [42]. Geometric similarity is valid when the dimensions of one unit relate to the second unit with just one constant factor. Thus, dynamic similarity has two conditions: first, geometric similarity is valid; and second, all independent dimensionless numbers are the same. Dynamic scaling can be divided into three major parts: fluid dynamics scaling; combustion scaling; and boiler design scaling [45]. Fluid dynamics scaling has been studied in detail by Glicksman et al. [42].

The set of dimensionless numbers which contribute to fluid dynamics similarity are as follows:

$$\frac{U_0^2}{gL}, \frac{\rho_p}{\rho_f}, \frac{\rho_p U_0 d_p^2}{\mu L}, \frac{\rho_f U_0 L}{\mu}, \frac{G}{\rho_p U_0}, \emptyset, PSD$$
(3)

However, solving this set of dimensionless numbers would lead to a unique set of parameters like particle size, bed size and unit size. While finding a unique set of parameters is desirable, often it is practically impossible to find the correct solution in such a constrained set of parameters. Simplifying the dimensionless numbers gives more choices and fewer constraints on the dimensions of the unit. For example, typically in CFB units, friction between the gas and the furnace walls is small, compared to other forces and if eliminated from the gas momentum equation, allows the set of dimensionless numbers in Eq. (3) to be simplified to [42]:

$$\frac{U_0^2}{gL}, \frac{U_0}{U_{\rm mf}}, \frac{G}{\rho_{\rm p} U_0}, \varnothing, PSD$$
(4)

In order to achieve fluid dynamics similarity between two units of varying size, the dimensionless numbers indicated in (4) and the geometry should be similar. The comparison of lab-scale units with industrial or even other lab-scale units is often difficult due to the lack of geometric similarity. Industrial CFB units used for electricity production typically have an aspect ratio lower than 10 [189], whereas lab-scale units have aspect ratios > 30 [45]. In such narrow units, lateral mixing tends to be better than in large-scale units [17]. This lack of geometric similarity raises the question of the extent to which CFB transport and mixing phenomena are transferable from a narrow small-scale unit to a wider large-scale unit.

In all, if the results of the experimental campaigns presented in Section 2 are divided into three categories of combustion chemistry, axial mixing and lateral mixing, the fluid dynamics scaling for the design of large-scale oxy-fuel CFB boilers can take the lateral mixing information only from pilot- and industrial-scale experimental campaigns.

5. Combustion challenges

The combustion reactions are critical for an oxy-fired CFB, starting with char combustion.

5.1. Char combustion

Char combustion is the most important energy-releasing reaction for the combustion of solid fuels and is the main source of energy, CO and CO_2 in solid fuel boilers. Murphey and Shaddix found a dramatic increase in the particle temperature in pulverized fuel flames when increasing the O_2 concentration [190].

Heterogeneous reactions consist of three stages: (1) adsorption of oxygen onto the char surface; (2) surface reaction between char and O_2 ; and (3) desorption of CO_2 or CO formed. Modelling the formation of the combustion reaction products requires the simulation of several parallel adsorption-desorption processes, each with individual activation energies [190,191]. The most widely used approach for detailed reaction mechanism evaluation is the Langmuir–Hinshelwood derivation [190,192]. The Arrhenius equation is also widely used for quantifying kinetic rate constants across a range of temperatures for various reactions [17,18,65,191,193–195]. The most important heterogeneous reactions relevant to combustion efficiency and temperature profile in oxy-fuel CFBs are the char combustion and gasification reactions, as described below.

CO is an effective marker to observe the progression of combustion in CFB furnaces. CO measurements can also show to what extent the combustion is complete or incomplete at different heights along the furnace [17,18,196,197]. Riaza et al., using an entrained flow reactor, reported that char burnout slows down at low inlet O_2 concentrations during oxy-fuel combustion owing to higher heat capacity and lower diffusivity of CO_2 in comparison with N_2 [198]. Riaza et al. also noted that switching to oxy-fuel conditions led to an increased char particle ignition temperature [198]. Yuzbasi and Selçuk reported that the char combustion process is retarded in oxy-fuel conditions as compared to air-fired conditions at the same oxygen levels, and is characterized by a lower rate of reaction and higher burnout temperature [199,200].

Solid fuel conversion in oxy-fuel conditions produces a char with a higher carbon content due to the loss/conversion of more hydrogen and oxygen. Furthermore, these chars typically have a higher specific surface (almost six times). Furthermore, the high concentration of CO_2 prevents secondary char formation (by shifting the equilibrium position of the Boudouard reaction) and tar polymerization reactions, but may lead to additional CO production [201].

A major challenge with modelling char combustion via Langmuir–Hinshelwood type mechanisms or Arrhenius methods is that most of them are semi-empirical models valid only in the range of temperatures and oxygen partial pressures for which the correlations parameters are derived [17,190,202]. Fitting factors, such as the tortuosity factors and the effectiveness factors are used to adapt intrinsic kinetic parameters derived at the lab scale for a larger-scale system,

although this is still limited to the range over which the data are gathered.

5.2. Gasification reactions

Although gasification reactions are negligible in air-fired CFB combustion due to their slow reaction rates compared to combustion reactions, they become more relevant in the oxy-fuel atmosphere [14,170]. The predominant equations, the Boudouard and water–gas reactions, are given by Eqs. (5) and (6), respectively:

$$C + CO_2 \leftrightarrow 2CO \tag{5}$$

$$C + H_2 O \leftrightarrow CO + H_2 \tag{6}$$

Many studies are available on gasification reactions in atmospheres with high concentrations of CO_2 and steam [203–205].

However, the importance of the gasification reactions is highly temperature- and fuel-dependent. For example, Jia and Tan [206] reported that the gasification reaction between CO_2 and carbon becomes relevant at temperatures above 1033 K for low-rank, high-reactivity coals (such as lignite) and at temperatures above 1173 K for anthracite and petroleum coke.

5.3. NO_x and N₂O emissions

NO arises from three mechanisms: prompt-, fuel- and thermal-NO_x, with fuel-NO_x the primary source of NO_x from FBC systems. Typically, prompt-NO occurs in gas flames, while thermal-NO_x will only become significant at temperatures above 1273 K.

A schematic of the fate of fuel nitrogen in combustion is shown in Fig. 14. Air-fired FBCs typically have lower total NO_x emissions compared to conventional PC combustion, due to the lower combustion bed temperature. It should be noted that N_2O emissions are not a problem for woody biomass, where the bulk of the fuel-N is released in the form of NH_3 rather than HCN, or during char-N oxidation [207]. The dependence of the NO_x emission on O_2 concentration is also discussed elsewhere [208].

 NO_x pollutants can be generated from either char-bound or volatilebound nitrogen compounds but the type and quantity of emissions depend significantly on the operating conditions and fuel characteristics [209].

Hofbauer et al. [56] concluded that the NO_x emissions increased with increasing inlet O_2 concentration (26–36 vol%) when conducting

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Fig. 15. NO_x concentration and the percentage of nitrogen conversion for airfired and oxy-fired combustion showing that increasing the oxygen concentration from 26% to 36% increased the overall NO_x production [56].

the combustion reactions in a 150 kW_{th} pilot-scale CFB unit, the results of which are shown in Fig. 15.

de Diego et al. [211] reported that, regardless of initial concentration of NO and temperature, oxy-CFB FGR leads to more than 60% of the recycled NO being converted to N₂, which is mainly due to the reburning process, while less than 5% is converted to N₂O [212,213]. In pilot plant runs with a 0.8 MWth CFBC, NO_x emission were shown to increase modestly, around 18%, when changing bed temperature from 1123 K to 1193 K utilizing bituminous coal [40]. These pilot plant results also demonstrated that the final emitted NO_x was considerably lower in oxy-fuel conditions. Duan et al. [62] concluded that the emission of NO_x under oxy-fuel was much lower due to enhanced gas phase reduction from FGR.

It is widely known that the presence of char and CO in the bed leads to reduction of the NO_x into N_2 and N_2O ; this was demonstrated recently by Duan et al. [214], using a 50 kW_{th} oxy-fuel CFB with a 21% inlet O_2 concentration. Further, they found that increasing the inlet O_2 concentration from 21 vol% to 40 vol% led to a higher conversion of the fuel-N to NO. In a modelling study, Peng et al. [215] suggested that the levels of NO and N_2O emissions increase with the increase in excess oxygen. This has also been supported experimentally for air-fired systems but is less clear for oxy-fired systems [209].

Air staging is a proven method for reducing NO_x emission from CFB furnaces [216]. Duan et al. [214] concluded that O_2 staging in oxy-fuel CFB was more effective in reducing NO emissions compared to air-fired conditions, while de las Obras-Loscertales et al. [217] reported that wet



Fig. 14. Possible fates of nitrogen contained in coal [210].

FGR caused a sharp decrease in NO emissions and a slight increase in $N_{2}O$ emissions.

5.4. Chlorine

During the combustion process, molecular chlorine, Cl_2 , and hydrogen chloride gas, HCl, may be produced; modelling has shown that Cl_2 formation is favored at temperatures above 600 °C and in oxygenrich environments [218]. Chlorine-containing product formation is highly temperature-dependent and thus may vary when changing to oxy-fired CFB. Font et al. reported that due to the high organic affinity of chlorine in coal, the retention of chlorine at any stage of the process or gas clean-up was difficult under oxy-fuel conditions [219].

Lupiáñez et al. [146] found that oxy-firing increased the chlorine detected in fly ashes in comparison to the air-fired tests, whereas, no chlorine was detected in the solids taken from the bed bottom under both air- and oxy-fired conditions.

Chlorine is a key contributor to the formation of aerosols and submicron particles [220] and high-temperature corrosion in boilers firing biomass and waste, due to the formation of KCl and NaCl compounds.

5.5. Bromine

In boilers firing solid recovered fuels or sludge, bromine plays a key role in aerosol formation and high-temperature corrosion. The bromine in sludge comes from water treatment chemicals containing bromine [158]. The formation and reduction behavior of Br mimics the Cl behavior and will form similar halogenated compounds with alkaline earth metals and heavy tars, if present [221]. While fuels typically have only tens of ppm levels of Br, the slags and wall deposits are reported to contain up to 3 wt%, mainly in the form of KBr and NaBr [158]. While Br is very important in emission evaluation and ash deposition, there is no information available on it in oxy-fuel CFB conditions.

5.6. Mercury

Suriyawong et al. [222,223] investigated mercury speciation under O₂/CO₂ coal combustion in a tubular furnace with a coal feeding system, and experimental results indicated that the distribution of Hg was similar between air and O₂/CO₂ combustion. Font et al. [219] quantitatively analyzed the fate of mercury in a 90 kW_{th} bubbling fluidized bed in O2/CO2 combustion conditions, and found that elemental Hg was the major species in the exhaust gas, while the major mercury species retained in bag filters was Hg²⁺. Wang et al. evaluated mercury speciation in 50 kW_{th} [224] and 6 kW_{th} [225,226] fluidized beds, and observed a distinct difference in mercury speciation between air and O₂/CO₂ coal combustion. Gharebaghi et al. [227] simulated the results of mercury oxidation and speciation under oxy-coal combustion using the combined homogeneous-heterogeneous model. Contreras et al. [228] conducted a series of thermodynamic equilibrium calculations on the fate of mercury, in which they found that chlorine speciation was the key factor affecting the fate of mercury in O2/CO2 combustion.

 Cl_2 , SO_x , and NO_x also play important roles in Hg oxidation in both air- and oxy-fuel combustion [229]. Chatel-Pelage et al. [230] studied mercury emission in a pilot-scale (1.5 MW_{th}) pulverized coal-fired boiler in O_2/CO_2 . SO_x and NO_x were both important factors in determining the mercury emission. Fernandez-Miranda et al. [231] pointed out that SO_2 , NO_x , and HCl can accelerate Hg⁰ oxidation. Wu et al. [232] provided evidence that NO concentration can influence mercury oxidation.

The level of mercury emissions also depends on the furnace temperature, with higher temperatures leading to the reaction of elemental mercury with chlorine [233,234], which may lead to capture of both elements in the form of fly ash. Font et al. [219] used a 90 kW_{th} oxy-fuel bubbling fluidized bed (BFB) with coal and limestone and reported high

levels of mercury emissions in the exhaust gases $(2.15 \,\mu\text{g/m}^3)$; of these total mercury emissions, 82% was elemental and the rest was Hg²⁺, which is in gaseous form and is of great environmental concern. The retention of chlorine and mercury in the form of bottom ash is desirable since it minimizes the risk of atmospheric emission in the case of less effective flue gas filters. Further research on the interaction of mercury, chlorine and Na-based sorbents is necessary for efficient in-furnace emission removal [235].

6. Conclusions - Scale-up challenges and opportunities

Oxy-fuel CFB combustion is based on mature fluidized bed boiler technology, and offers a good opportunity for reducing CO_2 emissions from heat-generating facilities. The major conclusions from this study are:

Scale-up: Lab- and pilot-scale units give valuable information on the chemical kinetics since they offer more or less the same residence time and environment as larger-scale oxy-fuel boilers. While there are numerous measurement campaigns in pilot- and industrial-scale oxyfuel CFB units, only limited results taken from such experimental campaigns are reliable for design and scale-up of large-scale oxy-fuel CFB units. For instance, data and models on reaction mechanisms are transferable from lab- and pilot-scale furnaces to large-scale units. However, utilization of heat transfer correlations, ash deposition data and furnace hydrodynamics requires further investigation. In particular, mixing is very much dependent on the unit size and, therefore, scale-up requires experimentation into the mixing issue in industrial- or large-scale boilers.

Boiler design: The two roadmaps of Constant-Furnace-Size Scenario and Constant-Thermal-Power Scenario are of technical and economic importance. Both scenarios rely on the economic potential of oxy-fuel CCS at increased O2 concentrations. The Constant-Furnace-Size Scenario is the more suitable option for retrofitting air-fired CFB boilers where, with the same furnace geometry, elevated O₂ concentration causes enhanced boiler thermal power. In the Constant-Thermal-Power Scenario, the furnace size becomes smaller when increasing O₂ concentration due to the reduced volumetric flow rate of RFG at increased O₂ concentration. Achieving high inlet O₂ concentration is important for reducing the cost of generated power by increasing the boiler thermal power or by furnace size reduction. In addition to the considerable furnace size reduction, the more homogeneous temperature profile and the lower heat flux from furnace water walls make the Constant-Thermal-Power Scenario a better roadmap to commercialization.

Heat transfer: Radiative heat extraction is normally the dominant heat extraction mechanism. Control of the furnace maximum heat flux and local temperatures under oxy-fuel conditions is necessary to avoid damage to water walls at high inlet O_2 concentrations. Ash deposition mechanisms on the heat extraction surfaces can be different from those seen in typical air-fired conditions due to temperature variations and different chemistry.

Fluid dynamics: The considerable increase in circulating solids at high inlet O_2 concentration leads to changes in furnace hydrodynamics including high solids concentration, high share of fine particles and aerosols (down to nanoparticles) in the furnace and the need for much more efficient cyclones. From a hydrodynamics point of view, the retrofit scenario is applicable only at low to medium inlet O_2 concentrations. For high inlet O_2 concentrations, a new boiler design is necessary and the semi-empirical data taken from typical air-fired CFB or from pilot-scale oxy-fuel CFB should be re-evaluated if used for design of large-scale units with an inlet O_2 concentration above 40%.

Combustion: Major reactions can vary from air-fired to oxy-fuel. Char gasification reactions become important while in-furnace profiles of gases like CO, SO_x and NO_x vary considerably compared to typical air-fired CFB. However, indirect sulphation is likely to be the dominant sulphur capture route. NO_x emission should be lower under oxy-fired conditions due to the NO_x reburn process and higher concentrations of steam coming from the wet RFG.

Data Statement

No new data was generated in the course of this research.

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Seddighi, Sadegh

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