



Influence of gas-staging on pollutant emissions from fluidized bed oxy-firing



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HIGHLIGHTS

- Influence of gas-staging is experimentally surveyed in a lab-scale oxy-fired FB reactor.
- Efficiency of NO_x control is quite different depending on the volatile content of the coal.
- The increase of secondary gas ratio from 0% to 10% has been shown more effective than from 10% to 20%.
- The larger the %O₂ in the firing atmosphere, the lower the efficiency of the gas-staging.
- Side effects affecting CO and SO₂ have been also determined and related to NO_x emissions.

ARTICLE INFO

Article history:

Received 8 May 2014

Received in revised form 2 July 2014

Accepted 3 July 2014

Available online 14 July 2014

Keywords:

NO_x emissions

Gas-staging

Oxy-firing

Fluidized beds

ABSTRACT

Air-staged combustion is a known, successful way to reduce NO_x emission in solid-fired systems. Despite this measure has been more frequently applied in pulverized fuel units, it can also be found in fluidized bed boilers. In this case, air-staging affects the fluid dynamics and other closely related issues, like combustion efficiency or desulphurization. The extent of these effects is still to be completely determined in oxy-fired boilers. This paper discusses the influence of gas-staging on CO, NO_x and SO₂ emissions in fluidized bed oxy-firing. An experimental plan was carried out in a 90 kW_{th} bubbling fluidized bed with two secondary gas inlets located at different heights. The tests were conducted firing two Spanish coals (anthracite and lignite) for a variety of O₂/CO₂ atmospheres and secondary flow rates. The secondary gas ratio has been revealed as the key variable, since solid mixing and particle conversion in the dense zone are directly affected. While gas-staging is shown to be an effective measure to reduce NO_x emissions for all the coals and atmospheres tested, SO₂ is however penalized, so finally a trade-off solution is needed. These trends are deepened for the higher values of CO₂ concentration.

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1. Introduction

During the last years, oxy-firing is being developed as a technology with a view to zero CO₂ emission power plants [1–5]. The use of a feeding gas free of N₂ allows obtaining high CO₂ concentrations in the flue gases, which can be recovered and treated to be stored. Several demonstration projects support the interest on this technology [6–10].

The development of oxy-fuel was initially focused on pulverized-fuel units [11–14], but now the application to fluidized bed units is also relevant. Oxy-fired fluidized bed boilers retain the advantages of air-fired units, namely: capability of burning a wide range of fuels, quite uniform temperatures along the reactor and low emission records [15,16]. As for the latter, low NO_x and SO₂

figures are observed due to moderate combustion temperature and in situ desulphurization. Despite pollutant concentrations in oxy-fired systems are higher than those in air-fired, due to the lower gas flow rates, conversion factors and normalized emissions per energy unit are similar and even lower under O₂/CO₂ atmospheres [17,18]. Moreover, the recycling of a fraction of flue gases enables an additional reduction of pollutant emissions [19–21].

In an oxy-fired combustor, the absence of N₂ in the feeding gas drastically reduces the production of thermal-NO_x. Furthermore, this mechanism can be considered negligible in a fluidized bed reactor due to operation temperatures much below 1500 °C. The same can be said for prompt-NO_x. Then, the only NO_x source that should be considered in a fluidized bed operated under oxy-firing conditions is the so called fuel-NO_x [22].

Basu [23] reported a general review of the influence of several parameters on NO and N₂O emissions under conventional combustion. The higher the nitrogen contained in fuel, the volatile-N

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fraction, the temperature, the excess oxygen and the limestone addition, the higher the NO emissions. The tendencies observed for N₂O are the opposite, except in the case of the fuel-N and excess oxygen. On the other hand, some measures contributed to decrease the NO emitted: air-staging and addition of ammonia or urea. A direct relation is also reported between SO₂ and NO: the higher the former, the lower the latter. Duan et al. [24] showed coherent influences for limestone addition and primary/secondary air ratios. The change of fluidizing gas from air to a O₂/CO₂ mixture does not alter the tendencies observed for volatile-N fraction, temperature and excess oxygen [17,24,25].

Oxidant staging in fluidized bed reactors brings along two different combustion zones. The first region is located under the secondary injection ports, a fuel-rich zone limiting the nitrogen oxidation and reducing the formed NO by homogeneous reactions with the radical-pool or by heterogeneous reaction with the char. Over the injection ports an oxygen-rich zone is located, where oxidation of volatiles and char particles is achieved. There is also a twofold effect in the dense zone contributing to control NO_x emissions by adopting the oxidant staging: decrease of temperature and increase of CO concentration. There is a direct influence of temperature on nitrogen and carbon oxidation rates [26], while higher CO concentrations favour NO depletion either by direct interaction or acting as catalyst of the reaction between NO and char surface [27–29]. The effectiveness of oxidant staging has been demonstrated in open literature for conventional combustion [30–32] but also for oxy-combustion, both in pulverized fuel [12] and fluidized bed units [24].

Since primary fluidizing gas is reduced when gas-staging is implemented, fluid-dynamics and particles mixing can be significantly affected. Ersoy et al. [33] analysed the influence of the location of the injection ports and the ports arrangement (radial/tangential). They observed a decrease of fluidization velocity and an increase of solids holdup below the injection section for all the geometries. Over the secondary air ports, solids holdup only was increased for the tangential cases. In addition, Koksai and Hamdullahpur [34] reported that tangential injections enhanced mixing of gas and solids in a larger extent than radial ones.

Desulphurization is also modified due to the lower amount of primary gas. Lyngfelt [35,36] found a negative effect of air-staging on sulphur retention caused by the CaSO₄ decomposition due to larger CO concentrations:



The same trend was later reported by Kahn [37] who explained it by the reductive conditions below the secondary injections and the lack of enough O₂ to complete retention by the limestone.

Okasha [31] reported a different effect of air-staging on SO₂ emissions depending on the value of the secondary air ratio. For ratios below 10%, sulphur retention was increased due to the smaller bubbles sizes and higher residence times, which favours the SO₂ retention. This result was also reported by Christensen [38,39]. Nevertheless, if secondary air ratio was increased over 10%, particle-gas mixing was not adequate enough to have a proper desulphurization and SO₂ emissions increased [31].

Under oxy-firing conditions, there are additional issues that should be surveyed. The high CO₂ concentration in the feeding gas and the increased concentration of H₂O, if wet recycling is adopted, enhance the release of CO according to the reactions (R.2) and (R.3), and then indirectly affect the emissions of NO_x and SO₂ [40–43]:



This paper aims at increasing the knowledge of emissions control in oxy-fired fluidized bed reactors, by experimentally characterising CO, NO_x and SO₂ emissions when two different coals are fired under a variety of O₂/CO₂ atmospheres and secondary gas flow rates. Firstly, experimental setup and test campaigns are described, and secondly, results are analysed and discussed.

2. Experimental setup

The experiments reported in this paper were carried out at CIRCE oxy-firing facility (see Fig. 1). It consists on a 90 kW_{th} bubbling fluidized bed reactor, which is 2.5 m height. The lower part of the reactor, up to 1 m over the distribution plate, is water-cooled in order to control bed temperature. The freeboard is refractory isolated. A detailed description of the entire facility, as well as instruments and auxiliary equipment, can be found elsewhere [17,44–47].

The feeding stream is injected in the reactor as primary and secondary gas. Primary gas is fed through a distributor plate located at the reactor bottom, while secondary gas can be supplied at two different heights: 40 and 80 cm over the plate. The first inlet is usually around the splash zone and the second one is at the bottom of the freeboard, in a region with lower solid concentrations. Both jets are tangentially introduced to produce a progressive mixing of the fresh oxidizer with the gases generated in the lower part of the reactor (Fig. 2). Flow rates are controlled by valves and measured by calibrated rotameters. Composition of primary and secondary gas is the same, and then oxygen concentration cannot be modified in height.

Gas streams are on-line monitored by thermocouples, pressure sensors and a gas analyzer. Samples from the feeding gas and the flue gases can be alternatively analysed. Non-dispersive IR technique is used to measure CO (0–5000 ppm), CO₂ (0–100%), SO₂ (0–30,000 ppm) and NO (0–1000 ppm), while a paramagnetic sensor is used to measure O₂ concentration (0–100%). Experimental uncertainties respectively are 1% for the measured temperatures, 2% for the measured pressures and 1% of the full-scale for the gas composition.

NO measurements reported and discussed in the following section are the combination of NO + NO₂, as delivered by the gas analyser. All the results have been normalized according to Eq. (1), in order to enable the comparison between the different tests,

$$[\text{gas}]_{6\%} = [\text{gas}]_{\text{fg}} \frac{[\text{O}_2]_{\text{inlet}} - 6}{[\text{O}_2]_{\text{inlet}} - [\text{O}_2]_{\text{fg}}} \quad (1)$$

where oxygen concentrations are all expressed in %.

Two Spanish coals were selected to proceed with the tests, low volatile anthracite and high sulphur lignite. Both coals were sieved to a particle size ranging 0.7–1.2 mm. Granicarb limestone was used as SO₂ sorbent, with a mean particle size of 0.5 mm. Calcium-to-sulphur ratio was kept 4 for all the tests conducted. The composition of coals and limestone is summarised in Table 1. The inert material of the bed was silica sand, with a mean particle size of 0.55 mm.

Experiments were carried under air-firing conditions and oxy-firing conditions, for two different O₂/CO₂ atmospheres. Gases are directly taken from commercial bottles, and mixed to get the prescribed composition. No flue gas recycling (FGR) was used during these tests, in order to avoid instabilities in recycled flow rates and species concentrations due to uncontrolled air in-leakages. If this FGR operation is coupled to secondary gas supply, it is not possible to distinguish their independent effects on emissions. This is a somewhat limiting condition of the facility, since concentration levels are below the expected values in a real plant with FGR, but at least tendencies can be deemed as reliable.

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