

Pollutant emissions in a bubbling fluidized bed combustor working in oxy-fuel operating conditions: Effect of flue gas recirculation

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HIGHLIGHTS

- ▶ Tests in a BFB combustor at oxy-firing combustion conditions were carried out.
- ▶ The effect of flue gas recirculation on pollutant emissions was analyzed.
- ▶ SO₂ recirculation increased the Ca-based sorbent utilization.
- ▶ About 60–70% of the NO recirculated was reduced to N₂ and N₂O.
- ▶ Steam recirculation decreased NO emission.

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ABSTRACT

In this work, pollutant emissions in a continuous bubbling fluidized bed combustor (~3 kW_{th}) at oxy-firing conditions were measured. An anthracite coal was used as fuel and a limestone was added for sulfur retention. Flue gas recirculation was simulated by mixing different gases (CO₂, SO₂, steam, and NO) and the effect of varying the gas composition of the recycled flow on the pollutant emissions was analyzed. It was observed that the most important effect of CO₂ recirculation was the increase of the optimum temperature for SO₂ retention from ~850 °C (conventional air combustion) to 900–925 °C. SO₂ recirculation increased the Ca-based sorbent utilization and did not affect the N₂O emissions at any temperature. In addition, at 850 °C the CO emission increased and the NO emission decreased, however, at 925 °C the CO variation was negligible and the NO reduction was low. About 60–70% of the recycled NO was reduced to N₂ and N₂O, being the NO converted to N₂O lower than 5%. Steam recirculation mainly led to a sharp decrease in NO emission. A synergetic effect among the different recycled gases was not found in any case.

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1. Introduction

Nowadays, there is an important public awareness in relation to the hazard of pollutant gas emissions into the atmosphere from fossil fuel combustion to obtain energy. On the one hand, CO₂ gas has been recognized as one of the major contributors to the build-up of greenhouse gases and, on the other hand, sulfur and nitrogen content in coal are oxidized to SO₂ and NO_x respectively, which contribute to acid rain formation.

According to the IPCC 2005 [1], carbon dioxide capture and storage technologies would be needed to mitigate CO₂ emissions from large power plants into the atmosphere for stabilization of atmospheric greenhouse gas concentrations. Oxy-fuel combustion is a CO₂ capture technology which is characterized by the use of a mix-

ture of pure O₂ and CO₂-rich recycled flue gas (instead of air which is used in conventional combustion) to perform the combustion process. As a consequence, with this technology, the CO₂ concentration in the flue gas may be enriched up to 95 vol.% (dry), and therefore it is possible to achieve an easy CO₂ recovery.

There are mainly two types of boilers for coal combustion, pulverized coal (PC) boilers and fluidized bed boilers. Although the majority of the researches in oxy-fuel combustion have been developed for PC boilers [2,3], fluidized bed combustors (FBC), and specially circulating fluidized bed (CFB) combustors, are also very appropriate for this combustion system. This technology has the advantage that external solid heat exchangers can be used to extract heat from the combustion process [4]. This will allow the use of high oxygen concentrations in the combustor reducing the amount of recycled flue gas and the area of the CFB combustor. Moreover, in situ desulfurization of combustion gases, by feeding a low cost sorbent such as limestone [4,5], and relatively low NO_x emissions can be achieved [6,7].

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Currently, the oxy-fuel combustion technology in CFB combustors is growing. Alstom [8], VTT and Foster Wheeler [6], Metso [9], Czestochowa University of Technology [7], and Canmet Energy [4,5,10,11] have performed oxy-fuel combustion experiments with CFB combustors at scales up to 4 MW_{th}. The Fundación Ciuden [12] in Spain has built two plants able to operate at both conventional air and oxy-fuel combustion conditions. The first plant is a 20 MW_{th} PC boiler and the second plant is a CFB combustor of 15 MW_{th} operating in air-mode and 30 MW_{th} operating in oxy-mode. However, there are few works published which deal with the effect of the flue gas recycle on the pollutant gas emission and thus there is still some lack of knowledge. The research group of Canmet Energy has successfully operated two CFB combustors of 100 kW_{th} [4,5,13] and 0.8 MW_{th} [10,11] with flue gas recycle. At an operating temperature of ~850 °C, they found lower SO₂ retention by calcium sorbents in oxy-fuel combustion than in air combustion conditions, but the SO₂ retention improved in oxy-fuel combustion by increasing the operation temperature, that is, when the conditions changed from direct to indirect sulfation. In addition, mainly due to flue gas recirculation, lower NO_x emissions in oxy-firing operation were observed compared to air operation.

Our research group has recently carried out several tests in a continuous bubbling fluidized bed (BFB) combustor (~3 kW_{th}) where the effect of the temperature on the sulfur retention by limestone addition was analyzed [14]. It was concluded that the optimum operating temperature from the point of view of SO₂ retention shifted from 850–870 °C in combustion with enriched air to 900–925 °C in oxy-fuel combustion mode.

In this work, pollutant gas emissions at different operating temperatures, mainly working at oxy-fuel combustion mode, were measured in a continuous BFB combustor (~3 kW_{th}). However, the main objective of this work was to analyze the effect of the flue gas recycled into the reactor on the pollutant emissions working in oxy-firing mode. The gas inflow recycled was simulated by mixing different gases (CO₂, SO₂, H₂O, and NO), which allowed us to observe the influence of the different gases, both separately and combined with each other.

2. Experimental section

2.1. Materials

A Spanish anthracite coal was selected as fuel for this study. The coal was crushed and sieved, and the particle size in the range of 0.2–1.2 mm was used. Table 1 gives the proximate and ultimate analyses of the coal. A high purity Spanish limestone “Granicarb” (97.1 wt.% CaCO₃) was used as calcium-based sorbent for sulfur retention. Table 2 gives the analysis of the Granicarb limestone. The particle size of the limestone was in the range of 0.3–0.5 mm. The porosities of the raw and after calcination sorbent

Table 1
Analysis of anthracite coal.

| Proximate analysis (wt.%) | |
|-------------------------------|-------|
| Moisture | 2.3 |
| Ash | 31.7 |
| Volatiles | 5.6 |
| Fixed C | 60.4 |
| Ultimate analysis (wt.%, daf) | |
| C | 90.36 |
| H | 2.53 |
| N | 1.41 |
| S | 2.30 |
| LHV ^a (kJ/kg) | 21807 |

^a Lower heating value.

Table 2
Granicarb limestone characteristics.

| Composition (wt.%) | |
|--------------------------------|------|
| CaCO ₃ | 97.1 |
| MgCO ₃ | 0.2 |
| Na ₂ O | 1.1 |
| SiO ₂ | <0.1 |
| Al ₂ O ₃ | <0.1 |
| Fe ₂ O ₃ | <0.1 |
| Porosity (%) | |
| Raw | 3.7 |
| Calcined ^a | 49 |

^a Calcined in N₂ atmosphere at 900 °C for 10 min.

were 3.7 and 49 vol.%, respectively. Inert silica sand of size 0.2–0.6 mm was fed together with the coal and the limestone during all the tests to control the residence time of the sorbent in the fluidized bed reactor.

2.2. Experimental installation

The experimental installation consisted of a fluidized bed combustor (~3 kW_{th}) and different auxiliary systems for gas supply, solid feeding, solid recovering, and gas analysis. Fig. 1 shows a schematic diagram of the installation.

The combustor consisted of a stainless steel reactor of 9.5 cm i.d. and 60 cm height and a freeboard of 15 cm i.d. and 50 cm height. The height of solid in the BFB was maintained constant at 40 cm. A heat exchanger located inside the bed allowed the perfect control of temperature. This heat exchanger could be moved vertically through the reactor to modify the contact surface inside the bed, which allowed the extraction of the needed heat from the combustor to reach the desired temperature.

The reactant gases, air, CO₂, and O₂, were supplied from cylinders by means of electronic mass-flow controllers to simulate typical gas compositions entering into the reactor in oxy-firing mode. The gases were fed into the reactor through a gas distributor plate. An air pre-heater allowed the introduction of hot air inside the

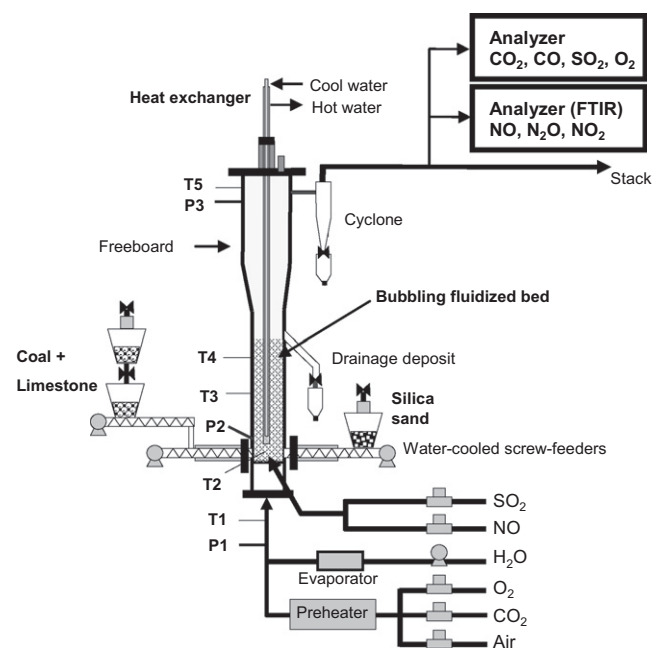


Fig. 1. Oxy-fuel bubbling fluidized bed combustor at ICB-CSIC. Measurements of temperature (T) and pressure (P).

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