



Full Length Article

Effect of co-firing on emissions and deposition during fluidized bed oxy-combustion



Carlos Lupiáñez^a, M. Carmen Mayoral^b, Isabel Guedea^a, Sergio Espatolero^a, Luis I. Díez^{a,*}, Sergio Laguarda^b, J. Manuel Andrés^b

^a Center of Research of Energy Resources and Consumptions, University of Zaragoza, Mariano Esquillor 15, 50018 Zaragoza, Spain

^b Instituto de Carboquímica-CSIC, Miguel Luesma 4, 50018 Zaragoza, Spain

HIGHLIGHTS

- Oxy-combustion of anthracite and corn stover blends is experimentally characterized in a lab-scale reactor.
- SO₂ emissions are affected by the chlorine content in the biomass while NO_x are more dependent on operating conditions.
- In comparison to air combustion, oxy-firing increases the chlorine detected in fly ashes.
- Deposition rates are affected by the KCl content in the corn rather than the firing atmosphere.
- K₂Ca(SO₄)₂ and K₃Na(SO₄)₂ are found in deposits for the more severe conditions.

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ABSTRACT

The relevance of coal oxy-firing in fluidized bed reactors has increased during the last years as an alternative for the development of CO₂ capture technologies. The addition of biomass in the fuel blend is well-known in conventional combustion, but few experiences are found under oxy-combustion conditions. This paper discusses the results obtained when oxy-firing anthracite and corn stover in a lab-scale fluidized bed, paying attention to pollutant emissions, deposition rates and composition of the ashes. While SO₂ emissions are affected by the chlorine content supplied with the biomass, NO_x are much more dependent on operating conditions in a similar way to conventional combustion. As concerns the ash composition, chlorine is detected in fly ashes while the bed solids are mostly composed by aluminosilicates. Oxy-firing increases the chlorine detected in fly ashes in comparison to the air-fired tests. Deposition rates are barely modified by the O₂/CO₂ atmosphere; severe deposition is only detected for the blend with the highest chlorine content. Mixed Ca-K sulfates are found in deposits, minimizing the risk of chlorine-induced corrosion.

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1. Introduction

During the last years, an important effort has been done to demonstrate oxy-firing as a feasible technology for capturing CO₂ in power stations [1,2]. Firstly on pulverized burners [3,4] and later on fluidized bed reactors [2,5], O₂/CO₂ combustion shows promising solutions to reduce the CO₂ emissions.

The application of oxy-firing on fluidized bed reactors offers the inherent advantages of this kind of technology, i.e. fuel flexibility, moderate combustion temperature and low pollutant emissions. Moreover, fluidized bed reactors do not require the development

of new burners suitable for the oxidizer composition, since they are able to operate with high O₂ concentration [6–9] which favours smaller boilers and ancillaries [1].

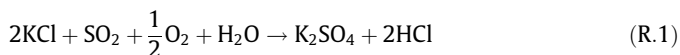
Biomass combustion widens the energy use of residues, with a neutral CO₂ contribution. The possibility of burning 100% biomass in fluidized beds has been demonstrated under conventional combustion conditions [10,11]. Nevertheless, the heterogeneous chemical composition of the biomasses requires a detailed analysis to avoid several inconveniences during the operation [12–14]. Some elements as alkali or chlorine promote the formation of deposits on heat exchangers tubes, which enhance corrosion and diminish heat transfer.

Co-firing is conceived as a way to introduce biomass in large-scale coal-fired plants, but the synergies between the mineral matter of the fuels have to be well determined. Aho and

* Corresponding author.

E-mail address: luisig@unizar.es (L.I. Díez).

Ferrer [15] studied the effect of coal ashes composition on the chlorine deposition during co-firing experiments in fluidized bed, reporting significant influences for the coals with higher contents in Al, Si and S. In this way, KCl from biomass mineral matter can follow reactions (R.1) and (R.2), in presence of sulfur oxides from the coal:



Then, HCl is released in the gas-phase as result of the KCl transformation and K_2SO_4 is found in the deposits. While fouling propensity by K_2SO_4 is increased in comparison to potassium aluminosilicates, corrosion promoted by KCl can be to the contrary attenuated.

Co-firing can also affect the NO_x emissions in comparison to coal combustion. Even though NO_x formation is more sensitive to combustion conditions (temperatures and O_2 concentrations), the volatile content and composition can also play a role. Therefore, the trends reported by previous works show a wide variety of behaviours [16,17]. For fluidized beds, it can be stated that the lower the volatile matter of the fuels and the lower the N-bound in the volatiles, the lower the NO_x emissions. This is due to several reasons: (a) in the dense phase, char particles participate in NO_x -reduction mechanisms [18,19], (b) in the diluted phase, volatiles oxidation increases the temperature and then the NO_x formation rates.

Few experiences are reported so far as concerns oxy-combustion of coal and biomass blends. Jurado et al. [20] carried out some experiments in a pulverized burner highlighting the relevance of the increase of corrosive species in the reactor due to dry-FGR (Flue Gas Recycling) and a similar amount on sulfur in the deposits formed. Ekvall et al. [21] simulated oxy-co-firing in a pulverized burner reporting a high influence of H_2O and SO_2 in FGR to control sulfation of alkali compounds, while the increase of HCl inhibits the alkali sulfation ratio. Regarding fluidized bed experiments, Tan et al. [22] demonstrated the suitability of fluidized bed reactors to carry out oxy-co-firing with low pollutant emission, including VOCs. Duan et al. [23] studied the influence of operational parameters (temperature, O_2 concentration and primary oxidant fraction) on NO_x emissions: the higher the three parameters, the higher the NO emitted.

Aiming at increasing the available results related to oxy-co-firing in fluidized bed units, this paper reports an experimental investigation carried out firing anthracite and corn stover in a lab-scale oxy-fired bubbling fluidized bed. Discussion is focused in the effect of adding corn stover with different chlorine contents on the gaseous emissions (SO_2 , NO_x , HCl) and the ashes composition, as well as the deposits characterization.

2. Experimental work

2.1. Facility

The experiments were run in CIRCE fluidized bed laboratory (Fig. 1). The reactor is 2.5 m height and its inner diameter is 0.203 m. The facility can be operated under conventional combustion or under oxy-firing conditions, then feeding O_2 and CO_2 from commercial cylinders. Fuel is fed by regulated endless screws and bed temperature is controlled by four water-cooled probes. The installation is fully instrumented with temperature and pressure meters. The uncertainties of the measurements are: 1% full-scale for the thermocouples and 2% full-scale for pressure sensors. More details about the facility can be found elsewhere [24,25].

An air-cooled deposition probe (AISI304) was introduced at 80 cm over distributor to gather the deposits formed during the

experiments. The probe consists of two concentric tubes, 16 mm and 36 mm inner diameter respectively. Compressed air is introduced through the inner tube and flows back through the outer tube. The probe is equipped with three thermocouples (air inlet temperature, air outlet temperature and probe tip temperature). The air-cooling flowrate is regulated by a PID controller to keep the probe temperature within the range 450–500 °C, resembling the performance of a superheater tube. A removable coupon is placed near the tip probe that can be replaced for each experimental condition. Once every test is finished, the coupon is removed for analysis and a new one is placed in the probe.

To detect the chlorine in the gas-phase, a suction system can be inserted at 90 cm over the distributor. A gas sample of 0.4 m³/h is conveyed through three consecutive impingers containing Na_2CO_3 dissolution (0.1 M), respectively named Trap1, Trap2 and Trap3. This is done to ensure the complete capture of sulfates and chlorine ($\text{HCl} + \text{Cl}_2$) of the gas sample. The SO_4^{2-} and Cl^- concentrations in every trap are determined by ion chromatography in a Metrohm Chromatograph equipped with a Metrosep A Supp 5 column.

Gas analyser takes on-line samples from the flue gas circuit to measure the content of CO_2 , CO, SO_2 and NO (NDIR sensors) and O_2 (paramagnetic sensor). The uncertainty of these measurements is 1% full-scale.

2.2. Fuels

The selected fuels for the co-firing experiments were anthracite and corn stover. The former because of the large amount of reserves available at Northwest Spain, the latter due to the growing interest in valorising domestic agricultural residues for energy use. Table 1 shows their ultimate and proximate analysis and heating value, as well as ash composition obtained by ICP. It is worth mentioning the very different content in volatiles, as well as the differences in the ash content and composition: the major components found in the coal ashes are Al and Si, while the biomass shows a larger content in K.

Semi-quantitative analysis of the mineral matter was performed by XRD, confirming the presence of aluminosilicates (48% wt.), quartz (47% wt.) and pyrite (5% wt.) in the anthracite, and sylvite (KCl, 56% wt.), Ca-Mg carbonates (30% wt.), quartz (10% wt.) and halite (NaCl, 4% wt.) in the corn stover. No sulfates were detected in the original ashes.

Corn stover possesses a significant content in chlorine, which is an outstanding specificity in relation to fouling and corrosion phenomena. Since the chlorine content in the original samples was low (0.35%) in comparison to the ranges available in other works [26–28], the received stuff was doped with KCl in order to increase its chlorine concentration. Doping was carried by spraying the particles of corn stover with the KCl solution, followed by a natural drying to stabilize moisture. Then, three different contents were finally tested during the experiments: 0.35% (as received), 1% and 2%.

2.3. Experimental matrix

Table 2 shows the experimental conditions during the co-firing tests. They can be divided in two groups according to the fluidizing gas: air-firing (A1–A2) and 30/70% O_2/CO_2 oxy-firing (O1–O5). Most of the tests were carried out for 80/20 coal-to-biomass energy ratio, except test O3 when the amount of biomass was reduced. The corn stover supplied covered three different chlorine contents: 0.35%, 1% and 2%. The addition of limestone was also included in tests O4 and O5, with molar ratio Ca/S = 2.5. Limestone is typically used in fluidized bed reactors to get in-situ capture of SO_2 by sulfation. Ca/S molar ratios usually range 2–6, depending on the sulfur content of the coal. For the tested anthracite in this paper, previous experiences have shown Ca/S = 2.5 as a reasonable

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