



## Emissions from co-firing lignite and biomass in an oxy-fired CFBC

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### ARTICLE INFO

#### Keywords:

Oxy-firing  
Circulating fluidized bed combustion  
Emissions  
Co-firing

### ABSTRACT

The co-combustion of a high-sulfur lignite and biomass blend (up to 50% by weight) has been studied in a small oxy-fired circulating fluidized bed combustion (CFBC) pilot plant. Here the goal is to examine the effect of biomass share on NO<sub>x</sub>, SO<sub>2</sub> and CO emissions. In these tests, a series of runs has looked at the effect of increasing biomass share under air firing, followed by tests in oxy-firing mode. The results show that the emissions are remarkably insensitive to the biomass share, and are comparable to other results for coal combustion, and likely to be well below any current emission guidelines. Overall, there appear to be no direct challenges to oxy-fuel co-firing in terms of gaseous emissions, although the simple lack of studies means that significantly more data are required on CFBC oxy-firing using a much wider range of biomass and coal types. K doping was also examined and did not result in significant formation of K phases on deposit probes.

### 1. Introduction

Oxy-fuel combustion is one of the most promising routes for using fossil fuels in a CO<sub>2</sub>-neutral manner. When combined with biomass firing it may also offer net reductions of CO<sub>2</sub> emissions, the so-called Bio-Energy with Carbon Storage (BECS) option [1, 2]. Here fluidized bed combustion is arguably the best available co-firing technology [3], and it has recently been demonstrated in its oxy-combustion form at the 30 MW<sub>th</sub> scale at Ciuden, in Spain [4, 5]. However, despite rapid advances in the technology, there are remarkably few studies on the co-firing of biomass and coal in an oxy-fuel circulating fluidized bed combustion (CFBC) environment [6–9] and, for that matter, relatively few papers on the operation of oxy-fuel CFBC [7, 10–13] in general. Moreover, the necessity for recycling flue gas, in order to control the temperature in the primary reaction loop, means higher emission levels in the flue gases with the potential for fouling, corrosion and other problems [14, 15]. Given the interest in the potential for achieving negative CO<sub>2</sub> emissions [16], and the very successful oxy-fuel CFBC trials carried out by Foster Wheeler at Ciuden, in Spain, there is a need to accelerate such developments. To do this we must explore a wide range of coals and biomass products in order to investigate the behavior of oxy-fuel CFBC technology with a range of feedstocks beyond the three co-firing studies currently available, which were carried out on a 0.4 m dia. CFBC [7], and a 0.1 m dia. CFBC [8, 9], respectively. In this work, a Spanish lignite was used and K doping was adopted for the oxy-

fuel trials, to simulate the co-combustion of Turkish lignite and olive cake which typically contains high levels of K. It was anticipated that emissions would be similarly low compared with our previous studies, and that K doping would not influence those emissions. Less certain was whether the K doping would lead to measurable fouling or the production of agglomerates.

### 2. Materials and methods

#### 2.1. Characteristics of fuels and limestone

A Spanish lignite and wood pellets (obtained from a commercial supplier (Valfei, Canada) were used in this study. The proximate and ultimate analyses of these fuels are given in Table 1. In all combustion tests, the lignite particle size was < 4.76 mm and the wood pellets were approximately 20 mm in length and 5 mm in diameter.

Havelock limestone from Canada, which is a high-purity limestone (> 98% CaCO<sub>3</sub>), was also used to examine SO<sub>2</sub> emissions under co-firing conditions. Havelock has been used extensively in other FBC work at CanmetENERGY and elsewhere [17, 18].

#### 2.2. Experimental

Combustion tests were carried out in a lab-scale circulating fluidized bed combustor located at Natural Resources Canada – CanmetENERGY

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**Table 1**  
Proximate and ultimate analyses of Spanish lignite and wood pellets.

	Spanish lignite	Wood pellets
Proximate analysis, % by wt. (as fired)		
FC	29.26	15.40
VM	29.36	80.17
Ash	28.40	0.4
Moisture	12.98	4.03
Ultimate analysis, % by wt. (dry basis)		
C	44.4	49.6
H	2.92	5.92
N	0.52	0.13
S <sub>total</sub>	5.72	
O	13.83	43.93
Ash	32.64	0.42
HHV, kJ/kg (as fired)	14,600	18,900

FC: Fixed Carbon, VM: Volatile Matter, HHV: Higher Heating Value.

in Ottawa, Canada. The schematic diagram of the experimental setup is shown in Fig. 1. The experimental setup consists of a combustor, a cyclone, a return leg, a bag filter and a feeding system. The feeding system is comprised of two fuel hoppers for lignite and wood pellets. Limestone was added to the system by mixing it with lignite. The combustor is constructed of SS310 steel and has an internal diameter of 0.1 m, and height of 5.1 m. It is fitted with electric heaters capable of operating at temperatures up to 1100 °C. This facility has been described in detail elsewhere [10, 11, 19].

In all, eight tests of several hours duration were carried out in order to determine the effect of biomass share, O<sub>2</sub> percentage in the combustion gas and dense phase bed temperature on flue gas emissions. Five of these tests were air-fired and the remaining tests were oxy-fueled. The experimental matrix consists of three different experimental sets which are described in Table 2. All but one of these tests were done at 850 °C, with a final test carried out at 915 °C to determine the effect of temperature. Both the percentage of biomass and the oxygen in the recirculated flue gas were varied, and the three distinct campaigns are as shown in Table 2. Here, no attempt was made to keep the heat input constant; instead the mass fraction of biomass was changed (10, 30 and 50% weight) by changing the coal feed rate accordingly. Secondary air was not used, and all emission figures in the tables and figures have been re-calculated to 6% O<sub>2</sub>.

The first campaign was carried out in an air-fired mode to determine the effect of biomass share on flue gas emissions. In these tests both temperature in the dense bed ( $T_{\text{dense phase}}$ ) and superficial velocity in the combustor were kept effectively constant, at about 850 °C and between 2.5 and 3 m/s, respectively, as far as possible. The experiments involved combustion of Spanish lignite, and co-combustion of Spanish lignite and wood pellet fuel mixtures containing 0, 10, 30, and 50% by weight of wood pellets (Tests no. 1–1 to 1–4).

In the second campaign the tests were designed to determine the effect of O<sub>2</sub> percentage in the combustion gas on emission levels in the flue gas. The experiments included co-combustion of Spanish lignite and wood pellets (1:1 weight ratio) for two different oxygen levels fed to the combustor (25% and 30% O<sub>2</sub>) (Tests no. 2–2 and 2–3) and these results are compared along with those for air firing (Test no. 2–1).

In the last campaign, three experiments were conducted to investigate the effect of biomass and limestone interaction and bed temperature on ash composition and deposit formation under oxy-firing conditions (30% O<sub>2</sub>). Three tests (3–1, 3–2 and 3–3) were carried out with K doping bringing the K<sub>2</sub>O content in the “biomass ash” to an overall 34–39%. This is roughly equivalent to levels associated with Turkish olive cake, which is a potentially important fuel in Turkey since it produces about 8% of the world's olives [20]. All three tests were carried out for a nominal 24 h period. To simulate the K effect, K<sub>2</sub>CO<sub>3</sub> solution was sprayed onto the wood pellets, which were then air dried prior to feeding. The amount of K<sub>2</sub>CO<sub>3</sub> added was based on the ash

content of the wood pellets and the K content typical of olive cake ash.

Spanish lignite/wood pellets (1:1 weight ratio) mixture was oxy-fired (30% O<sub>2</sub>) at 850 °C in Test no. 3–1. Test no. 3–2 was the same test with limestone addition (0–2 mm particle size). Tests no. 3–1 and 3–2 were used to see the effect of limestone addition to the fuel mixture on ash composition. The third test (Test no. 3–3) in this campaign was a repeat of test 3–2 with the operating temperature raised to 915 °C to see the effect of dense phase temperature on ash composition.

In order to carry out this work, a special “deposit sampling probe” was designed and manufactured to simulate a heat exchanger tube in the combustor (Fig. 2). The surface of the deposit sampling probe is cooled internally down to 550 °C with air in order to represent the surface temperature of the superheaters. The deposit sampling probe was used in order to collect fly ash samples during the combustion process. The particles accumulated on the probe's surface were collected at the end of the test. After the probe was removed from the combustor, the particles collected on the probe were very slowly scraped from the surface into a sampling pot. The deposit sampling probe itself was located at a level of 4.5 m above the distributor plate. The probe was made from a stainless steel tube with a 26.7 mm outer diameter (OD) and its length was 230 mm. The deposit samples themselves accumulated on the surface of the probe on the detachable ring which has a length of 60 mm, and a 26.7 mm OD. The probe temperature at this level was around 790–830 °C for the tests where dense phase temperature was kept at a nominal 850 °C (C-1, CB-1 to CB-8), and a temperature of 845 °C for the test where dense phase temperature was kept at 915 °C (CB-9).

The deposit characteristic X-ray diffraction (XRD) data were collected on a Rigaku Ultima IV XRD spectrometer over the angular range 7 to 80° (2-theta) in 0.02° steps at 1°/min. The XRD system operates in the theta:theta geometry, and uses Cu(K $\alpha$ ) radiation, 0.154 nm. The generator voltage and current settings were 40 kV and 44 mA, respectively. At the end of the co-combustion tests (Test nos 3–1 to 3–3), bed ash from the combustor and fly ash from the bag filter were collected. X-ray fluorescence (XRF) and XRD analyses of ash samples were carried out.

The diffraction spectra were processed using the JADE version 9.0 XRD processing software. Identification of the chemical or mineral compounds was performed using the search/match option in JADE. Accurate integrated intensities of diffraction peaks were derived through “peak deconvolution” or more commonly called “line profile fitting”, another software tool in JADE. These data were used for quantitative analysis. Quantitative analysis of the samples was done using a variety of different internal spiking standards. This technique is known as the reference intensity ratio (RIR) method. For quantitative XRD, each sample was mixed with a spiking agent. The resulting mixture was thoroughly ground using a mortar and pestle to ensure homogeneity of the phases. The resulting powdered mixture was loaded into a holder. The holder was transferred to the XRD instrument and data were collected. Integrated intensities for strong diffracting peaks of each phase were obtained using JADE. Concentrations of the chemicals identified in each sample were calculated using the method mentioned above. The amorphous content was estimated by difference.

For this work EPA Method 0031 was used for volatile organic compounds (VOC) sampling. The VOC sampling train consists of a heated glass-lined probe, two sorbent tubes containing Tenax®-GC (1.6 ± 0.1 g each), a third sorbent tube containing Anasorb®-747 (5.0 g ± 0.1 g), flow measurement and other related devices. Once steady state operation of the oxy-fuel mini-CFB was achieved, flue gas samples were pumped through the VOC sampling train. As the flue gas passed through the sorbent tubes, VOCs were absorbed onto the Tenax and the Anasorb sorbents. At the end of the VOC sampling period, the sorbent cartridges were analyzed to determine the amount of each volatile organic compound absorbed. Together with the volume of flue gas sample passing through the VOC sampling train, concentrations of VOCs were determined.

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