



Research article

Mercury emissions from coal combustion in fluidized beds under oxy-fuel and air conditions: Influence of coal characteristics and O₂ concentration



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ABSTRACT

In this work, the fate of mercury in a bubbling fluidized bed combustor working under oxy-combustion conditions has been studied and compared with air combustion. The influence of burning three different rank coals, with sulphur content ranging from 0.65% to 5.17%, on Hg partitioning has been studied. The presence of limestone as sorbent for SO₂ capture as well as the concentration of O₂ at the entrance of the combustor were also evaluated.

Coal rank does not have a direct influence on coal partitioning. However, sulphur content of the coal is an important parameter to describe not only the high percentage of particle-bound mercury, up to 87%, in presence of limestone but also the prevalence of Hg(0) as the main species in the gas phase when burning the lignite. The O₂/CO₂ ratio has little effect on Hg partitioning in experiments carried out in presence of limestone at 925 °C (the optimum temperature for sorbent sulphating under oxy-combustion conditions) and little differences are found with air combustion at 850 °C (the optimum temperature for sorbent sulphating under air combustion conditions).

Percentage of particle-bound mercury shows a maximum at 925 °C independently of the coal studied, which it is related with the maximum sulphur retention for each coal at this temperature.

1. Introduction

The basic principle of oxy-combustion is to burn a fuel in a mixture of oxygen and recycled flue gas (RFG) instead of air [1–3]. So, flue gas is mainly composed by CO₂ and H₂O, and the CO₂ rich gas is purified and then cooled, compressed and ready to transport and storage.

In addition to the advantage to avoid the CO₂ separation from flue gas, NO_x emission can be reduced to less than one-third of that of combustion in air because not only of the reduction of thermal NO_x (due to the air-nitrogen elimination) but also because of the reburning of recycled NO_x with volatiles as well as reaction of recycled NO_x with char [1,3,4]. On the other hand, the SO₂ concentration increases during oxycoal combustion with RFG [1,5] and gas purification by flue gas desulphurisation systems is mandatory because of the high risk of in-furnace corrosion and CO₂ transportation systems [6].

Fluidised bed (FB) combustors working under oxyfuel conditions allow the in situ flue gas desulphurisation by the injection of the appropriate sorbents in the boiler [7]. With respect to the accepted advantages of the circulating fluidised bed (CFB) [8,9], the abatement of SO₂ is also an important issue in oxycoal combustion.

Among trace elements coming from coal combustion, mercury is a leading concern among the air toxic metals because of its volatility, persistence, bioaccumulation in the environment and its neurological health impacts [10].

Mercury present in the flue gas can be found as elemental mercury, Hg(0), oxidized mercury, Hg(II), and particle-bound mercury, Hg(p). In general, mercury speciation depends on: coal properties, combustion conditions, the flue gas composition, fly ash properties, the time/temperature profile between the boiler and air pollution control devices, and the flue gas cleaning methods, if any, in use. Mercury can be oxidized in the SCR units, which can favour its adsorption on particulate matter and removed from the flue gas in the particulate matter control devices or, because of its solubility, Hg(II) can be retained in the WFGD systems. Thus, mainly Hg(0) would be released [11].

There are some studies concerning the fate of mercury during pulverized coal (PC) oxy-combustion indicating that the average oxidized mercury emission to stack are higher regarding conventional combustion [12,13]. Recent studies during air and oxy-fuel firing transitions at the Callide Oxy-fuel Project [14] have shown that oxy-fuel fly ash contains lower amounts of mercury than air fired fly ash; the authors

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found this trend consistent with the lower LOI values.

For FB boilers, two new aspects have to be added regarding PC oxy-combustion: the presence of a sorbent, which it is expected to have some mercury capture ability, and the type of reactor, which allows extended contact time between the sorbent and the Hg.

The effect of combustion temperature and recycled gases (NO, SO₂ and H₂O) on the fate of mercury emissions from coal combustion in fluidized beds (FB) has been previously studied [15]. In that study, it was concluded that the particle-bound mercury exhibited a maximum at a temperature about 925 °C, with different Ca-sorbents and Ca/S ratios which coincided with the highest degree of the limestone sulphation and so the lowest SO₂ concentration. However, the fate of mercury comparing air and oxy-combustion mode in FB, under different O₂ concentration at the entrance of the combustor and with different rank coals exhibiting different sulphur content is almost unexplored.

The quality of CO₂ to be transported and sequestered has been subject of research, concerning impurities participating in mineralization and precipitation reactions in sequestration conditions [16]. However, the role of mercury during CO₂ transport has not been deeply studied, in particular corrosion issues on steel pipelines when some humidity is present [17].

Moreover, for coal oxy-combustion, mercury is not an environmental issue alone but also an operational issue, particularly about where mercury could accumulate within the CO₂ processing unit. The removal of Hg is necessary to prevent Hg attack on the aluminium heat exchangers [18,19].

In this work, mercury emissions from a bubbling fluidized bed (BFB) combustor burning three different rank coals with a wide range of sulphur content operating under oxy-fuel conditions have been evaluated, testing the influence of oxygen concentration at the entrance of the combustor, comparing with air combustion baseline and the presence or absence of limestone in the bed.

2. Experimental

The experimental facility used was a BFB combustor of 3 kW_{th}. It consists of a fluidized bed combustor and different auxiliary systems for gas and solid supply, solid recovering and gas analysis. Temperature, pressure and gas flow were followed on-line. More details of the experimental installation can be found elsewhere [20].

CO₂, CO and SO₂ concentrations at the exit of the combustor were continuously analysed continuously by a Non-Dispersive Infrared analyser (NDIR, Siemens/Ultramat 23) and O₂ concentration was continuously analysed in a paramagnetic analyser (Siemens/Oxymat 6).

Sampling of gaseous mercury, Hg(0) and Hg(II) in the gaseous outlet stream was performed at steady state operation [15,21]. Oxidized mercury was trapped in impingers containing 100 ml of KCl 1 M. Saturated KMnO₄ (about 5%) solution was added to the KCl during the breakdown to remove SO₂ by the reaction between KMnO₄ and SO₂ preserving the mercury in sample solutions. Mercury content in impingers solutions was analysed using an automatized mercury analyser LECO AMA254 gold amalgam atomic absorption spectrometer. Elemental Hg was monitored on-line by a cold vapor atomic absorption spectroscopy (CVAAS-VM300) apparatus. As SO₂ is a spectral interferent, CVAAS signal was previously tested at different SO₂ concentrations in order to subtract its contribution to Hg real measurement [22].

Mercury content in solids elutriated from the combustor (recovered in a cyclone, CS) as well as bed materials (recovered in a drainage deposit, BS) was determined for each operating condition as a total Hg (p). Mercury contents were directly analysed on solids using the LECO AMA254 apparatus.

In order to evaluate the quality of Hg measurements, one of the experiments was repeated twice (lignite combustion at 925 °C under ratio O₂/CO₂ of 35/65) in two different days and the same experimental

Table 1
Analysis of coals and limestone.

wt%	Anthracite	Bituminous	Lignite	wt%	Limestone
Proximate				CaCO ₃	97.1
Moisture	2.3	5.2	12.5	MgCO ₃	0.2
Ash	31.7	12.9	25.2	Na ₂ O	1.1
Volatiles	5.6	32.7	28.7	SiO ₂	< 0.1
Fixed carbon	60.4	49.2	33.6	Al ₂ O ₃	< 0.1
				Fe ₂ O ₃	< 0.1
Elemental					
C	59.64	65.63	44.43		
H	1.67	4.64	2.50		
N	0.93	1.50	0.65		
S	1.52	0.65	5.17		
Sulphur forms					
Pyritic	1.50	n.d.	1.05		
Sulphate	0.02	n.d.	1.76		
Organic ^a	0.00	n.d.	2.36		
Hg, mg/kg	0.274	0.056	0.11	Hg, mg/kg	0.0002
LHV (kJ/kg)	21,807	25,398	16,252		

n.d.: not determined

^a By difference.

procedure was followed. In addition, two different Hg(0) measurement procedures were used during the experiment carried out with the anthracite under ratio O₂/CO₂ of 35/65 in presence of limestone: Hg(0) gas was determined on line with the VM3000 device and, simultaneously, the complete train of impingers according to OHM was used in order to confirm the continuous Hg(0) measurement. In these cases, solid samples from bed and cyclone were also obtained and Hg content determined in AMA254 device.

Tests were conducted under air or oxy-fuel conditions with an anthracite (A) coming from León (Spain), a high volatile bituminous coal (B) coming from Cerrejón (Colombia) and a lignite (L) coming from Teruel (Spain). A Spanish limestone (Granicarb) was used for sulphur capture. Main characteristics of the materials are given in Table 1.

To ensure constant Ca/S molar ratio the coal and the limestone were fed together. Sand was fed separately. It was introduced inside the FB in order to short the residence time of the sorbent in the fluidized bed reactor and, accordingly, to ensure the sorbent reaching the steady state in each experiment. In all the experiments the total inlet gas flow was constant (2.23 Nm³/h) and the O₂ in the flue gas was around 4% (d.b.). Detailed description of the experimental procedure can be found elsewhere [7,15,20].

3. Results and discussion

A summary of the experiments carried out in this study is depicted in Table 2.

Table 2
Summary of the experimental conditions and Hg mass balance (O₂ in flue gas around 4%).

Coal	T °C	O ₂ /CO ₂ %	Ca/S	Coal g/h	Limestone g/h	SO ₂ vppm	SR ^a %	Hg _{out} /Hg _{in} %
A	850	AIR	3	340	50	217	87.2	74.0
	925	25/75	3	397	50	361	69.5	74.2
	925	35/65	0	593	–	3047	0.0	73.6
	925	35/65	3	584	85	699	71.1	82.2
	925	45/55	3	780	100	925	74.7	84.3
B	850	35/65	3	530	30	891	24.8	76.4
	925	35/65	0	530	–	1298	0.2	81.9
	925	35/65	3	530	30	761	36.5	87.4
	950	35/65	3	530	30	1068	18.0	79.3
L	850	35/65	3	689	350	6560	68.2	96.5
	925	35/65	3	699	350	2651	77.1	118.2
	950	35/65	3	687	350	3593	48.5	95.7

^a SR: sulphur retention.

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