



Optimum temperature for sulphur retention in fluidised beds working under oxy-fuel combustion conditions



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ABSTRACT

Oxy-fuel combustion is one of the leading options for power generation with CO₂ capture. The process consists of burning the fuel with a mixture of nearly pure oxygen and a CO₂-rich recycled flue gas, resulting in a product flue gas from the boiler containing mainly CO₂ and H₂O. Among the possible boiler types, fluidised bed combustors are very appropriate for the oxy-fuel process because they allow the in situ desulphurisation by feeding Ca-based sorbents into the combustor.

In this work, the effect of the temperature of the combustor on the retention of the SO₂ generated in the combustion of two coals with very different sulphur content (a lignite and an anthracite) has been studied. The experimental facility used was a bubbling fluidised bed (BFB) combustor of ~3 kW_{th}. Tests were conducted under oxy-fuel combustion mode and also under enriched-air combustion mode for comparison reasons. A Spanish limestone “Granicarb” was used as Ca-based sorbent for sulphur retention. The temperatures tested were between 800 and 970 °C using Ca/S molar ratios between 0 and 3.

It was found that in BFB combustors operating under oxy-fuel combustion conditions the optimum temperature to achieve the highest sulphur retention was 900–925 °C, whereas operating with enriched air the optimum combustion temperature was 850–870 °C. Working at the optimum temperature, the SO₂ retentions were lower in oxy-fuel combustion than in enriched air combustion conditions. It was also observed that working with lignite there was 10–15% of sulphur retention by coal ashes, however, working with anthracite the sulphur retention by coal ashes was negligible. This finding was independent of the combustion mode used, oxy-fuel or enriched air.

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1. Introduction

The emission of gases into the atmosphere from burning fossil fuels, mainly coal, in large power plants causes important environmental problems. The CO₂ produced in combustion process has been recognised as one of the major contributors to the build-up of greenhouse gases in the atmosphere and, in addition, sulphur in coal is oxidised to SO₂, which contributes to acid rain formation.

The capture and storage of CO₂, emitted in large quantities from power plants, is considered an option to be explored in the medium term for reducing CO₂ levels released to the atmosphere. Oxy-fuel combustion is one of the possibilities under investigation within the different options for CO₂ capture [1–3]. This technology uses for combustion pure O₂ (instead of air used in conventional combustion) mixed with CO₂-rich recycled flue gas, and so, the flue gas stream from the boiler finally produced is highly concentrated on CO₂. After condensing water and a cleaning step, the CO₂ can be stored.

Among the possible boiler types, fluidised bed (FB) combustors are very appropriate for the oxy-fuel process because they allow the in situ desulphurisation of the flue gas by feeding Ca-based sorbents into the combustor, such as limestone or dolomite. This could be an advantage since sulphur containing species mean a risk of corrosion and could have impacts in the furnace, during ash collection, CO₂ compression, transport and storage [4,5]. Other well known advantages are the possibility of burning a wide range of fuels and the relatively low NO_x emissions achieved [6,7]. It is also believed that oxy-fuel circulating fluidised bed (CFB) combustors could be an important candidate for new coal fired power plants, mainly because the circulation of solids in the combustor can help to an effective control of the temperature [5–11]. This will permit the use of high oxygen concentrations in the combustor, thus reducing the area of the CFB combustor, as well as the amount of recycled flue gas, thus reducing the size of some subsequent equipments and power consumption [10]. Other important advantage is the efficiency improvement, mainly due to the strong mixing in the furnace and longer residence times of solids that allows a good carbon burnout.

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The development of the oxy-fuel combustion technology in CFB combustor is currently growing. Alstom [10], VTT and Foster Wheeler [11], Metso [12], Czestochowa University of Technology [7], and Canmet Energy [5,8,9,13] have experimented in oxy-fuel combustion with CFB combustors at scales up to 4 MW_{th}. The Fundación Ciuden [14] in Spain is developing two plants able to operate from conventional air combustion to oxy-fuel combustion. One is a 20 MW_{th} PC boiler and the other is a CFB combustor of 15 MW_{th} operating in air-mode and 30 MW_{th} operating in oxy-mode. The research group of Canmet Energy [8,9] is involved in testing a CFB combustor of 100 kW_{th} with flue gas recycle. Operating at ~850 °C, they found lower sulphation in oxy-fuel combustion than in air combustion conditions, but the sulphation improved working with a petroleum coke by increasing the operation temperature, that is, changing from direct to indirect sulphation. However, the same effect was not confirmed working with coals. Recently this group has successfully converted and operated a conventional 0.8 MW_{th} pilot-plant to oxy-fuel firing [5,13]. Their results strongly support the view that this technology offers all of the advantages of air-fired CFB. However, the test experiences showed that operating the pilot-scale unit over a long period under oxy-fuel mode led to enhanced corrosion due to higher sulphur concentrations in the flue gas [5]. SO₂ concentration under oxy-fuel mode was up to four times higher compared to air firing mode. They concluded that limestone performance for sulphur removal was impacted by the combustion mode, and that this impact depended on fuel characteristics as well as on combustion temperature.

Our research group have recently carried out several tests in a thermogravimetric analyser (TGA) and in a batch fluidised bed reactor to analyse the behaviour of limestones for SO₂ retention under oxy-fuel operating conditions [15,16]. We found that the major effect of increasing the CO₂ concentration in the reacting gas was to shift the CaCO₃ decomposition to CaO until a higher temperature. The higher temperature needed to work in conditions of indirect sulphation with increasing CO₂ contents had a negative influence on the sulphation conversion reached by the limestones. It was concluded that the optimum temperature for sulphur retention in oxy-fuel combustion mode in FB reactors was incremented up to ~925 °C from the 850 °C normally used at conventional air combustion conditions. This conclusion seems in good agreement with the results found by Canmet Energy working in its CFB combustors [5,9]. However, more work should be carried out in continuous fluidised bed reactors to confirm this conclusion.

The objective of this work was to determine the optimum operating temperature to achieve the maximum sulphur retention in fluidised bed reactors working under oxy-fuel operating conditions. Tests were carried out in a continuous bubbling fluidised bed (BFB) combustor (~3 kW_{th}) operating with limestone under calcining or non-calcining conditions. Two coals of different rank and sulphur content were used. The effect of Ca/S molar ratio with respect to the sorbent sulphation process was also analysed.

2. Experimental section

2.1. Materials

Two Spanish coals, an anthracite and a lignite with a high sulphur content, were selected for this study. Both coals were 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 coals. Sulphur in coal can exist in both inorganic and organic forms. The inorganic sulphur is mostly pyrite and/or marcasite, together with small amounts of sulphates depending on the level of oxidation of the coal. The organic sulphur exists either in aromatic rings

Table 1
Analysis of coals and its ashes.

	Coals	
	Lignite	Anthracite
Proximate analysis (wt.%)		
Moisture	12.6	1.0
Ash	25.2	31.6
Volatiles	28.7	7.6
Fixed C	33.6	59.9
Ultimate analysis (wt.%, dry)		
C	45.43	59.64
H	2.50	1.67
N	0.65	0.93
S	5.17	1.52
Sulphate	1.05	0.02
Pyritic	1.76	1.50
Organic (by diff.)	2.36	0.00
LHV ^a (kJ/kg)	16,252	21,807
Ash composition (wt.%)		
Al ₂ O ₃	24.81	24.93
CaO	6.54	2.73
Fe ₂ O ₃	20.05	10.22
K ₂ O	1.38	4.18
MgO	1.44	1.84
Na ₂ O	0.15	0.94
SiO ₂	41.00	51.47
TiO ₂	0.80	0.94

^a Low heating value.

or in aliphatic functional groups. The distribution between organic and pyritic sulphur depends on the rank and total sulphur content of the coal [17]. The release of the sulphur to form SO₂ in the oxy-fuel combustion process can depend on the sulphur distribution. So, it is also included in Table 1 the different forms of the sulphur and the composition of the coal ashes. In this case, all the sulphur present in the anthracite was as pyritic form meanwhile sulphur present in lignite was distributed in three forms. The analysis of the ashes shows that both coals contained some CaO, which could contribute to the reduction of the SO₂ emissions.

For sulphur capture tests, a high purity Spanish limestone “Granicarb” (97.1 wt.% CaCO₃) was used as Ca-based sorbent. Table 2 gives the analysis of the Granicarb limestone. Particle size of the limestone was in the range of 0.3–0.5 mm. The porosities of the raw and after calcination sorbent were 3.7% and 49%, respectively.

In addition, inert silica sand of size 0.2–0.6 mm was fed together with the coals and the limestone during all the tests to decrease the residence time of the sorbent in the fluidised bed reactor, as it will be commented later.

2.2. Experimental installation

The experimental installation of ~3 kW_{th} consisted of a fluidised bed combustor and different auxiliary systems for gas supply,

Table 2
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 during 10 min.

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