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## **Oxyfuel Combustion Using CFBC Recent Canadian Work**

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*Oxyfuel FBC has been examined in a 100 kW pilot plant operating with flue gas cooling. This technology offers almost all of the advantages of air-fired FBC-emissions such as CO or NO<sub>x</sub> are lower or comparable to air firing, and it is possible to switch from air firing to oxy firing easily, with oxygen concentrations as high as 60-70% and flue gas recycle levels of 50-60%. Only sulphation is poorer, which is not in good agreement with other studies, and the reasons for this discrepancy need further exploration. However, longer tests have confirmed these findings with two coals and a petroleum coke. It also appears that changing from direct to indirect sulphation with the petroleum coke improves the sulphation, although a similar effect could not be confirmed with coal.*

Key words: *oxifuel combustion, circulating fluidized bed CFB, emission reduction, sulphur capture*

### **Introduction**

Anthropogenic CO<sub>2</sub> production is primarily driven by fossil fuel combustion and the current energy demand situation gives no indication that this will change in the near future. Thus, it is necessary to find ways to reduce these emissions when fossil fuel is used, and of the various potential options, CO<sub>2</sub> capture and storage (CCS) appears to be among the most promising. The CCS technologies involve producing a pure stream of CO<sub>2</sub> either by concentrating it in some manner from the flue gases, or by using effectively pure oxygen as the combustion gas [1]. The latter option, oxyfuel combustion, has now been well studied for pulverized coal combustion, but to date has received relatively little attention in the case of oxyfuel circulating fluidized bed combustion (CFBC). Recently, Alstom and Foster Wheeler have explored the concept using pilot-scale equipment. Alstom's work included testing in a unit of up to 3 MW<sub>t</sub> in size, but did not involve recycle

of flue gas [2]. Foster Wheeler's work [3], also involved pilot-scale testing in a small (30-100 kW) CFBC, and this work along with CETC-O's work with its own 100 kW CFBC appear to be the first such units operated with oxyfuel combustion using full flue gas recycle.

The advantages of FBC are already well known in terms of its ability to burn a wide range of fuels, both singly and co-fired, to achieve relatively low  $\text{NO}_x$  emissions, and to remove  $\text{SO}_2$  with limestone [4]. An additional advantage of CFBC technology in the context of oxyfuel firing is the fact that hot solids are kept in the primary reaction loop by means of a hot cyclone. This solid circulation potentially provides an effective means, in conjunction with the recycle of flue gas, to control combustion, and effectively extracts heat during the combustion process, thus allowing either a significant reduction of the amount of recycled flue gas or alternatively, permitting the use of a much higher oxygen concentration in the combustor. This allows the economics of oxy-fired CFBC to be significantly improved over PC or stoker firing, or perhaps a reduction in area of the CFBC boiler island by as much as 50% [2].

Advantages of the technology that are more difficult to define relate to the possibility of co-firing biomass, so that with CCS, the overall combustion process may potentially result in a net reduction of anthropogenic  $\text{CO}_2$ ; and the potential for this technology to be used with more marginal fuels, as premium fossil fuels come into short supply.

This paper will discuss results generated in this unit, along with a brief description of CETC-O's much larger 0.8  $\text{MW}_t$  CFBC pilot plant, which will also be run this year in oxyfuel CFBC mode, with flue gas recycle.

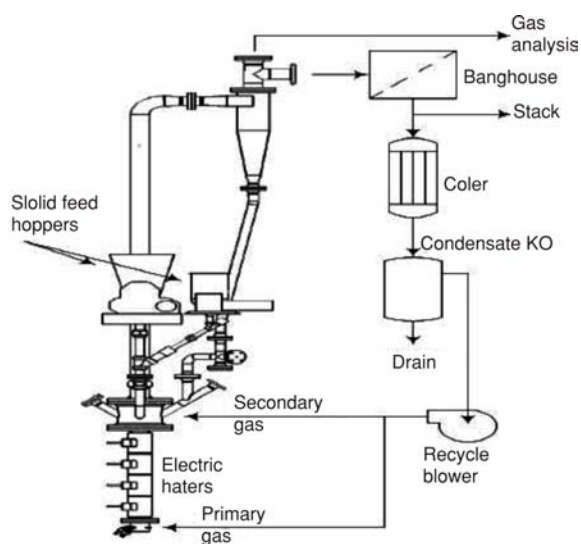


Figure 1. Schematic of CETC-O's mini-circulating fluidized bed facility

## Experiments

The test facility has been described in detail elsewhere [5] and will only be briefly described here. The mini-CFBC contains a 0.1 m ID stainless steel riser covered with 100 mm of insulation. Independent feed augers can supply multiple fuel types and a sorbent, with solid fuel feed rates of up to 15 kg/h. Oxygen,  $\text{CO}_2$ , and recycled flue gas flow rates are controlled by a combination of mass flow controllers and rotameters. Bed temperature is in the range of 750-950 °C. Superficial gas velocity is up to 8 m/s. The mini-CFBC has been extensively

modified for oxyfuel CFB combustion. Modifications include the addition of an oxygen supply line and a flue gas recycle train. A schematic of the unit appears in fig. 1.

*Fuel, limestone, and bed material*

Initially, eastern bituminous (EB) coal and Highvale coal (a sub-bituminous coal) were used. Table 1 gives the analysis of the two coals. Both coals were crushed to 5 mm. For sulphur capture tests, Havelock limestone was used when Eastern bituminous coal was fired.

**Table 1. Analysis of coals**

	Eastern bituminous	Highvale	Kentucky	Pet coke
<i>Proximate analysis [wt.%) (dry)</i>				
Moisture [wt.%) (as analyzed)	1.08	10.39	2.01	0.66
Ash	8.86	19.17	11.31	1.00
Volatile matter	35.78	33.76	37.35	11.46
Fixed carbon	55.56	47.07	51.34	86.97
<i>Ultimate analysis [wt.%) (dry)</i>				
Carbon	77.81	59.78	74.05	86.91
Hydrogen	5.05	3.49	5.06	3.22
Nitrogen	1.49	0.76	1.62	1.83
Sulphur	0.95	0.22	1.56	5.88
Ash	8.86	19.17	11.31	1.00
Oxygen (by difference)	6.04	16.58	6.40	1.16
<i>Heating value [MJ/kg]</i>	32.51	23.27	30.93	34.71

Table 2 gives the analysis of the Havelock limestone. The particle size of the limestone was in the range of 0-0.5 mm. In all cases, sand of size 0.15-0.35 mm was used as initial bed material. Subsequently, the work was extended to use both petroleum coke and Kentucky bituminous coal (analyses in tab. 1).

*Operating conditions*

The mini-CFBC was started in air-firing mode. When the bed temperature reached the desired level the air supply was shut off. The flue gas recycle blower was turned on. Oxygen was then supplied to the mini-CFBC. The transition from air-firing to oxyfuel-firing mode occurred easily and took only a few minutes. The mini-CFBC was operated at a

**Table 2. Analysis of Havelock limestone**

CaO	53.99	Cr <sub>2</sub> O <sub>3</sub>	<0.01
MgO	0.59	P <sub>2</sub> O <sub>5</sub>	<0.02
SiO <sub>2</sub>	1.23	SO <sub>3</sub>	0.20
Al <sub>2</sub> O <sub>3</sub>	<0.38	V <sub>2</sub> O <sub>5</sub>	<0.02
Fe <sub>2</sub> O <sub>3</sub>	<0.55	SrO	0.02
Na <sub>2</sub> O	<0.17	BaO	0.02
K <sub>2</sub> O	<0.08	NiO	0.01
MnO	0.08	LOF	43.34
TiO <sub>2</sub>	<0.04	SUM	99.48

nominal bed temperature of 850 °C. Superficial gas velocity was about 4 m/s. Global oxygen concentration during the oxyfuel combustion periods was 28-30%.

## Results and discussion

The first combustion tests used Highvale coal, since it is a low-sulphur fuel (0.22%) and does not require limestone addition. Experimental results during periods of stable operation under air firing and oxyfuel firing conditions are given in tab. 3. The data in tab. 3 show that, during oxyfuel firing, temperatures across

the mini-CFBC were very similar to those seen during air firing. Gas velocities in the dense bed zone and in the riser were slightly lower under oxyfuel firing conditions. Oxygen concentrations in the primary gas and secondary gas were 34.0% and 67.1% by volume, respectively. Global oxygen concentration was 28.8%. Primary and secondary combustion gas fractions were 0.684 and 0.056, respectively. The remainder of the gas was supplied through the return leg and the coal feed port. Errors associated with these estimates were about 5-10%.

The combustion of Highvale coal was excellent under oxyfuel firing conditions. The CO concentration of 39.4 ppm in the flue gas also provides a good indication of the excellent combustion performance, and was slightly lower than that achieved by air firing.

The SO<sub>2</sub> concentration was significantly lower during the oxyfuel-firing period. One possible explanation was that Highvale coal has an inherent Ca/S ratio of 6. Since the mini-CFBC was fired first in the air-firing mode before oxyfuel firing was started, fuel-derived ash might have been accumulated in the system as the test progressed. This may have resulted in an increase of calcium in the mini-CFBC for sulphur retention in the subsequent oxyfuel-firing period.

NO<sub>x</sub> concentration in the flue gas during the oxyfuel-firing period was twice as high as under the air-firing mode. Since temperatures across the mini-CFBC were very similar, the increase appears to be the result of flue gas recirculation. There are no data on oxyfuel CFB combustion with flue gas recirculation in the open literature, so we cannot evaluate our results with comparable test data reported elsewhere. Tan *et al.* [6] reported oxy-fuel pulverized fuel combustion with flue gas recirculation in a 0.3 MW<sub>t</sub> test facility. Using Highvale coal (the same coal as used here), results indicated that NO<sub>x</sub> concentration increased to 1183 ppm from 707 ppm in the air-firing mode. The magnitude of increase in NO<sub>x</sub> concentration was lower than in the current study (67% increase compared to 200%), albeit the NO<sub>x</sub> level from the pulverized coal combustor was much higher than for CFBC operation.

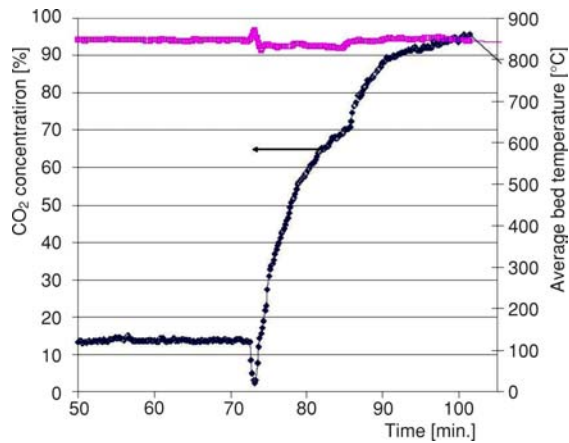
**Table 3. Oxy-fuel mini-CFBC combustion tests with Highvale and EB coals**

	Highvale coal				Eastern bituminous coal			
	Air firing		Oxyfuel		Air firing		Oxyfuel	
O <sub>2</sub> [%]	4.95	0.5	6.67	1.5	3.66	0.2	4.55	1.3
CO <sub>2</sub> [%]	13.4	0.4	80.5	1.3	13.9	0.13	83.5	
CO [ppm]	53	11	39.4	9	180		195	48
SO <sub>2</sub> [ppm]	169	29	100	13	295	20	1262	
NO <sub>x</sub> [ppm]	123	6	257	31	188	7	214	
Fuel mixture feed rate [kg h <sup>-1</sup> ]	4.0	0.4	4.0	0.4	3.5	0.35	4.0	0.4
Gas velocity in the bed [ms <sup>-1</sup> ]	2.94	0.06	2.60	0.07	2.74		2.33	0.10
Gas velocity in the riser [ms <sup>-1</sup> ]	3.72	0.09	3.56	0.11	3.72		3.37	0.15
Average bed temperature [°C]	849	4	845	10	848	6	851	14
Average riser temperature [°C]	824	19	823	11	827	22	868	18
Average cyclone temperature [C]	651	7	722	6	756		770	6
Average baghouse inlet temperature [°C]	227	4	219	3	258	5	216	6
Ca/S molar ratio	–		–		2.0		2.0	
Sulphur capture efficiency [%]	–		–		68.4		7	
Fuel N to NO <sub>x</sub> [%]	6.61		6.64		5.61		0.6	
Recycle ratio [%]	–		58.03		3.3		–	
Initial bed charge (sand) [kg]	5.0		5.0		5.0		5.0	

The flue gas recirculation ratio was estimated at 58.03–3.3%, which was significantly lower than for pulverized fuel oxyfuel firing. One of the reasons for the low recycle ratio was that, in the mini-CFBC, the solid particles were cooled in the return leg (all insulation material was removed from the return leg for these runs). The temperature of the solids entering the combustor from the return leg was 514–4 °C. If an external solid heat exchanger were available to cool the solids to a lower temperature, the flue gas recirculation ratio could be further reduced. During stable periods of oxyfuel CFBC firing with flue gas recirculation, the average CO<sub>2</sub> concentration in the flue gas was 80.5–1.3%.

The transition from air firing to oxyfuel firing was smooth in CETC-O's mini-CFBC. As shown in fig. 2, the amount of time required from the start of the transition to stable oxyfuel firing with flue gas recirculation was about 20 minutes.

To assess sulphur capture performance under oxyfuel CFBC firing conditions, EB coal was burned with Havelock limestone addition at a Ca/S molar ratio of 2. Table 3 gives the experimental results for these tests. Temperatures in the riser were higher during oxyfuel firing compared to air firing by about 40 °C. However, the average bed temperature was essentially the same for the two modes of combustion. Superficial gas velocities were slightly lower during the oxyfuel-firing period (–10%). The oxygen concentrations in primary and secondary combustion gases were 40.1 and 73.1%, respectively. Primary and secondary gas fractions were 0.588 and 0.062. The global oxygen



**Figure 2. Transition from air firing to oxy-fuel firing in CETC-O's mini-CFBC during Highvale coal test**

concentration was 32.9%, higher than in the tests performed with Highvale coal, which may explain the higher combustor temperatures measured in the EB coal tests.

The flue gas recycle ratio was very similar to that in the Highvale coal tests, 55.03%. The average temperature of the solids entering the CFBC riser from the return leg was 439 ± 15 °C. These results confirmed that recycle ratio for oxyfuel CFBC can be much lower than in oxyfuel pulverized fuel combustion, which represents a major advantage of CFBC, allowing one to reduce operational

cost, improve cycle efficiency and reduce the amount of CO<sub>2</sub> generated per unit electric power produced.

Concentrations of CO were basically the same during air firing and oxyfuel-firing. The trend was the same as in the Highvale coal tests, although absolute concentrations were higher at 180 to 195 ppm. NO<sub>x</sub> concentration was only slightly higher during the oxyfuel-firing period. The different behaviour in NO<sub>x</sub> concentration variation compared to Highvale coal tests (tab. 3) was likely the result of the characteristics of the coal. In this case, the ratio of fuel nitrogen converted to NO<sub>x</sub> was decreased by almost 50%.

Sulphur capture efficiency was 68.4% during air firing. During the oxyfuel-firing period, the sulphur capture efficiency decreased to 40.1%. These values were lower than expected as up to 90% sulphur capture is typically achievable in utility/industrial CFBC units with Ca/S ratios of 2 to 2.5 [7]. However, the current tests were short runs (7-8 hours), and steady-state of ash composition in the mini-CFBC had not been achieved.

To check that the limestone remained fully calcined, CFBC bed ash generated during the oxy-fuel-firing period was subjected to thermogravimetric analysis (TGA), and it was clear from the TGA results that added limestone did not calcine in the CFBC. Therefore, sulphur capture was via direct sulphation of CaCO<sub>3</sub>.

#### *Longer duration sulphation tests*

Longer duration runs were additionally performed with a Kentucky bituminous coal and a petroleum coke. For these runs Ca/S molar ratios were maintained at a value of 3, and the temperature studied ranged from 850 to 950 °C, *i. e.*, to allow a change from direct to indirect sulphation. The results for the petcoke test are shown in fig. 3.

This test, including preheating (not shown on the graph), lasted for about 12 hours. It started with combustion in air and then changed to oxyfuel firing without having to stop the fuel feed. The bed temperature was kept at 850 °C during air firing and the first part of the oxyfuel test, and was then increased to 950 °C for the second period of oxyfuel testing. It can be seen that, as the combustion mode switched from air firing to oxyfuel firing, the concentration of CO<sub>2</sub> rapidly increased and stabilized at about 85%. The concentration of O<sub>2</sub> stayed steady at 3-5%. The concentration of CO decreased from 100 ppm at 850 °C to 20 ppm at 950 °C due to higher O<sub>2</sub> concentration, which leads to better fuel burnout. However, NO<sub>x</sub> concentration stayed relatively steady.

The concentration of SO<sub>2</sub> decreased significantly when the bed temperature increased from 850 to 950 °C. This can be seen more clearly in fig. 4, which shows that as the bed temperature increased, SO<sub>2</sub> concentration quickly decreased from about 6000 ppm at 850 °C to about 2000 ppm at 950 °C, a reduction of more than 65%.

Figure 5 (EB coal) shows, once again, that a steady CO<sub>2</sub> concentration ( 90%) was achieved during the entire period of oxygen- -firing. As in the case for petcoke, concentration of CO decreased slightly when bed temperature increased from 850 to 950 °C (from 290+ min.). We also note that NO<sub>x</sub> concentration increased slightly with

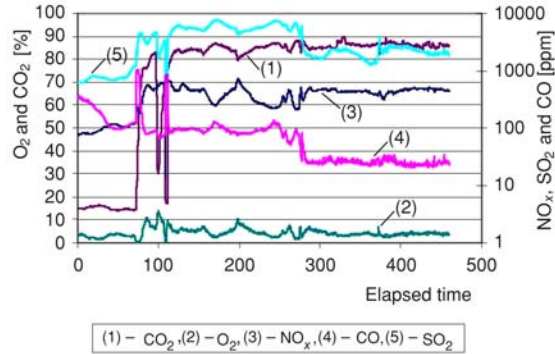


Figure 3. Concentration profiles of major species as a function of time for petcoke

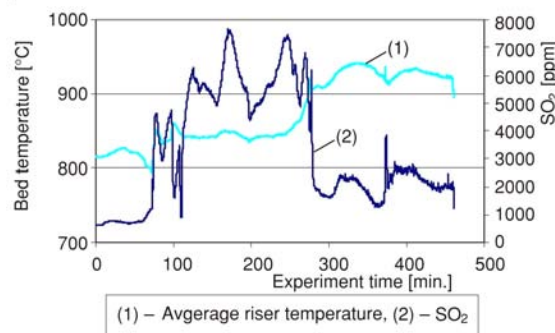


Figure 4. Profiles of SO<sub>2</sub> concentration and average bed temperature for petcoke

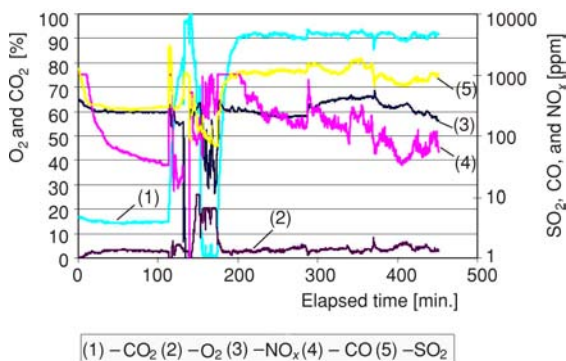
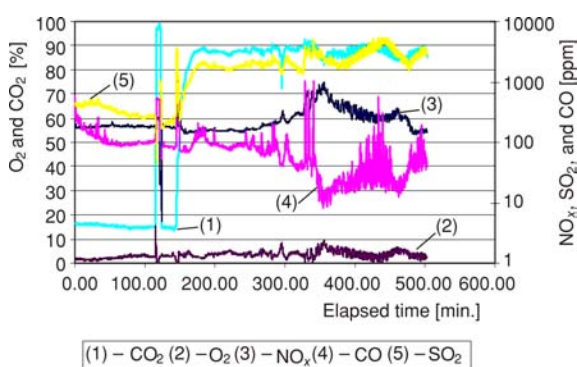


Figure 5. Concentration profiles of major species vs. time for EB coal

higher temperature. The effect of temperature on the concentration of  $\text{SO}_2$  was much less pronounced for this fuel than for petcoke. The data showed a decrease from about 1500 to 900 ppm, about 40% compared to 65% for petcoke.

As bed temperature started to increase from 850 to 950 °C (at about 300 min elapsed time), the major species exhibited significant fluctuations in concentrations. Although similar fluctuations were observed for EB coal, in this case the fluctuations were very pronounced, especially after 350 min. when bed temperature rose above 930 °C.

Fluctuations in both the bed temperature and  $\text{SO}_2$  concentration were too large to draw any conclusion concerning the effect of increasing temperature on  $\text{SO}_2$  emissions. During the coal tests, we observed that these fluctuations seemed to be closely correlated to the limestone calcination temperature. As bed temperature approached 930 °C, we would observe a sudden drop in the lower bed pressure accompanied by poor solid recirculation, which led to a rapid rise in bed temperature. In the case of petcoke, this effect was not as obvious. Further tests are being planned to elucidate this observation.



**Figure 6. Concentration profiles of major species vs. time for Kentucky coal**

For the Kentucky coal, the test results are shown in fig. 6.

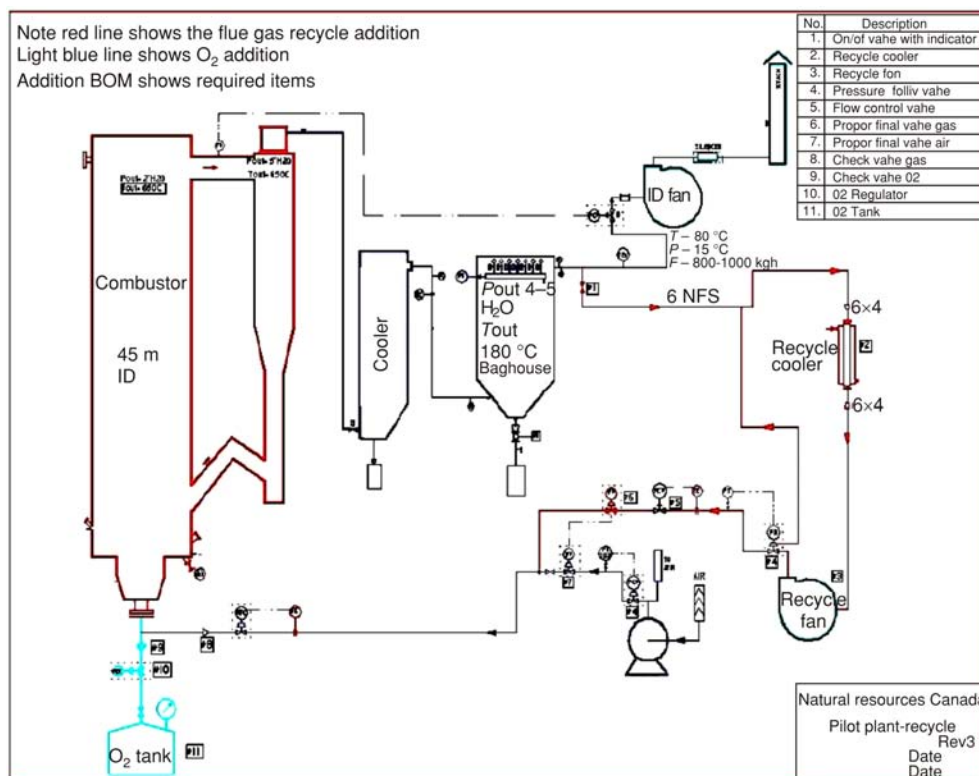
For petroleum coke the sulphur capture was about 55% at less than 900 °C, but increased to 85% as the bed temperature increased above 930 °C. For EB coal the sulphur capture was around 50-60% regardless of temperature. A possible explanation for the differences in these results may be the concentration of water in the combustion gases. For indirect sulphation the literature suggests no effect of water at least up to a concentration of 6% [7], but for direct sulphation water is

known to have an effect [8]. For petroleum coke, which is a low volatile fuel, and for which the moisture levels here are also very low, any effect of water might be expected to be minimal. For coal, however, water can be present in fairly high concentrations. Currently, TGA work is being carried out with these limestones to sort out these effects, and a test is also planned with water injection for the petroleum coke. A general result of these tests is to confirm relatively low sulphation levels for both coal and petroleum coke with direct sulphation, which is in agreement with the literature for direct sulphation with high  $\text{CO}_2$  partial pressures.



**CETC-O's retrofitted 0.8 MW<sub>t</sub> CFBC: platform for future oxyfuel combustion work**

CETC-O's 0.8 MW<sub>t</sub> CFBC, shown in fig. 7, was recently retrofitted for oxyfuel combustion. The main components are the riser, hot cyclone, return leg, flue gas cooler, baghouse, and feeders for fuel and sorbent. The riser has an internal diameter of 0.406 m, and an internal height of 6.6 m. Riser temperature is controlled with up to four wa-



**Figure 7. CETC-O's 0.8 MW<sub>t</sub> CFBC with flue gas recycle and oxygen supply**

ter-cooled bayonet tubes, which can be inserted or retracted during operation by a motorized winch system. An 1800 MJ per hour natural gas startup burner preheats the CFBC to the ignition temperature of the test fuel. During retrofitting, a flue gas recycle line was added, including a recycle blower, a flue gas condenser, and pressure control and safety equipment. The flue gas is drawn from the exit of the baghouse. Oxygen from a 3500 gal storage tank is mixed with the recycled flue gas to maintain combustion in the CFBC. Operating parameters remain basically the same as in the air-firing mode. However, the fa-

cility is run under slightly positive pressure to prevent air leakage, and oxygen level in the combustion gas can reach 29%. Flue gas from the CFBC is continuously analyzed for CO<sub>2</sub>, CO, O<sub>2</sub>, SO<sub>2</sub>, and NO<sub>x</sub>. The system can operate at temperatures up to 1000 °C and superficial velocity of 4-6 m/s.

## Conclusions

Oxyfuel CFBC tests were successfully conducted with flue gas recirculation for both sub-bituminous and bituminous coals and petroleum coke. Global oxygen concentrations in the combustion gas were 28.8 to 32.9%. Flue gas recycle ratio was in the range of 55 to 60%. The recycle ratio is significantly lower than that required for oxyfuel pulverized fuel combustion. NO<sub>x</sub> emissions were on a par with air firing with Highvale coal (sub-bituminous) and decreased by almost 50% for Eastern bituminous coal. CO concentration was basically the same for air firing and oxyfuel firing with flue gas recirculation under CFBC conditions. Sulphur capture was 40.1% with Ca/S = 2 during the oxyfuel-firing period, down from 68.4% in the air-firing mode. It was confirmed that sulphur capture under oxyfuel CFBC conditions was via direct sulphation of CaCO<sub>3</sub>. To further examine the issue of sulphur capture in oxyfuel-fired CFBC, tests of extended durations were carried out to ensure bed ash composition in the fluidized bed reached steady-state. Here, the tests were done using three kinds of fuel at bed temperature of 850 to 950 °C. The results of these tests showed that steady CO<sub>2</sub> concentrations were obtained at levels above 85% and that the bed temperatures were well controlled by adjusting the amount of recycled flue gas. Despite high local O<sub>2</sub> concentrations, no adverse effects were observed on ash quality. Test results showed that for petcoke, increasing bed temperature from 850 to 950 °C under oxyfuel conditions (to switch sulphur removal mechanism by limestone from direct sulphation to indirect sulphation) significantly increased limestone's sulphur removal efficiency. However, this effect was not conclusively observed for coal, which may suggest that water plays a significant effect on the sulphation process in oxyfuel combustion.

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## Апстракт

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## **Сагоревање у атмосфери обогаћеној кисеоником у циркулационом флуидизованом слоју Канадска искуства**

Истраживање сагоревања у атмосфери обогаћеној кисеоником је обављено на 100 kW-тном пилот постројењу са циркулационим флуидизованом слојем. Приказана технологија показује предности у односу на класичну технологију сагоревања у флуидизованом слоју када су у питању емисије гасова CO и NO<sub>x</sub>, као и могућности веома лаког преласка са класичне технологије сагоревања у ваздуху на сагоревање у атмосфери обогаћеној кисеоником чак до 60-70% уз рецикулацију димног гаса до 50-60%. Када је у питању одсумпоравање приказани резултати су у колизији са истраживањима других аутора и у овом делу су потребна додатна испитивања. Дуготрајнија испитивања овог сагоревања су потврдила тврдње из експерименталних тестова уз коришћење две врсте угља и коксом из петрохемијске индустрије. Такође, у случају кокса је потврђено ефикасније одсумпоравање при преласку са директног на индиректни метод, што се није могло потвдити на узорцима угља.

Кључне речи: *сагоревање у атмосфери обогаћеној кисеоником, циркулациони флуидизовани слој, смањење емисије, издвајање сумпора*

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