A pilot-scale demonstration of oxy-combustion with flue gas recirculation in a pulverized coal-fired boiler

Fabienne Châtel-Pélage, Ovidiu Marin and Nicolas Perrin
Air Liquide
Chicago Research Center, Countryside, IL

Ronald Carty
Illinois Clean Coal Institute
5776 Coal Drive, Suite 200, Carterville, IL

Gary R. Philo
Illinois Department of Commerce and Economic Opportunity
Office of Coal Development, 620 E. Adams St., Springfield IL

Hamid Farzan and Stanley J. Vecci
Babcock & Wilcox Company
B&W Research Center, Alliance, OH

ABSTRACT

Over the past years, environmental concerns for various combustion-generated emissions have grown rapidly: acid rain precursors (NO_x, SO_x) were initially targeted, followed by mercury emissions and more recently by carbon dioxide emissions, brought into focus as a greenhouse gas. As a consequence, governments have strengthened regulations, imposing SO_2, NO_x and mercury limits in a growing number of areas, and encouraging CO_2 reductions. Since the power generation industry and more specifically coal-fired power plants are responsible for a large amount of these pollutants, new combustion processes have to be developed in order to provide coal-fired power plants with a solution to comply with existing and future regulations.

In this context, a new combustion process based on oxygen-enriched flue gas recirculation for pulverized coal-fired power plants is being proposed to provide an easy-to-implement option for multi-pollutant control, including CO_2 capture. To date, theoretical analyses and lab-scale experiments have been reported, with no large-scale implementations.

This paper presents the experimental results obtained from a pilot-scale demonstration of the oxygen-enriched flue gas recirculation combustion process on a pulverized coal-boiler. This demonstration was performed on a 1.5MW boiler simulator, using Illinois coal.

Measurements from the pilot boiler demonstrate the technical feasibility of the process for larger scale-boilers, highlighting the favorable impact on NO_x, SO_x and mercury emissions, while concentrating the carbon dioxide in the flue gas. The results also show a significant reduction in fly ash unburned combustible content resulting in improvement in boiler efficiency due to the use of oxygen. The paper concludes that oxygen enrichment represents a technically sound technology for multi-pollutant control. A consequent paper presented during this conference discusses the economical aspects related to the technology, indicating that oxygen enrichment is an attractive solution for ultra-clean coal-based power generation.

INTRODUCTION

Over the past years, environmental concerns for various combustion-generated pollutants have grown. In addition to acid rain precursors (NO_x and SO_x), which were the first ones to be targeted, and remain a short-term issue, carbon dioxide emissions have became a major concern due to its associated greenhouse effect. As a consequence, the governments have tightened the regulations on SO_2, NO_x and mercury emission limits in a growing number of areas, and have started to regulate CO_2 release into the atmosphere and/or encourage the development of new technologies aimed at controlling this release.

The power generation industry and more specifically coal-fired units account for a large amount of above-mentioned pollutants – for instance around 10% of the worldwide CO_2 emissions result from coal-fired power plants. Therefore, it is imperative that new combustion concepts be developed and customized for power-plant needs, so that existing and new coal-fired units may utilize available options to comply with existing and future emissions regulations.

The traditional pollutant control method consists of a post-combustion flue gas treatment system comprising as many treatment devices as regulated pollutants. Currently, a conventional post-treatment line includes: a wet- or dry-FGD for SO_x removal, an ESP for particulate removal and a SCR or SNCR for NO_x reduction (when regulated). Future regulations on mercury and CO_2 emissions will necessitate the addition of an activated carbon injection device (Hg removal) and CO_2 separation installation (amine sorption and regeneration technique). Such a pollutant control method comes along with several drawbacks:

- Most of these installations are flue gas flow rate dependant. Therefore, they are very expensive when applied to conventional air-combustion systems, the inert nitrogen playing a “ballast” role in the process.
- In general these installations control one independent pollutant, thus requiring the addition of a new device (and financial burden) each time the authorities impose the emission of any new specie to be controlled.

It is thus easy to imagine that a nitrogen-free process would benefit from a highly reduced flue gas flow rate. This can be achieved by replacing the combustion air with pure oxygen in the combustion process. The resulting five-fold decrease in oxidant volume into the boiler leads to reduced flue gas treatment costs. In addition, by removing the nitrogen from the process, the flue gas is highly enriched in carbon dioxide. This characteristic allows its relatively straightforward capture for eventual sequestration without any need for expensive and energy consuming separation systems.

The oxy-combustion strategy may result in two main technologies. For new plants, the oxy-combustion strategy would include advanced, compact oxy-fired boilers, resulting in significant reductions in boiler dimensions (Horbaniuc *et al.* 2001). To develop a retrofit technology applicable to existing boilers, the solution consists of recirculating a portion of the flue gas, so that the nitrogen is basically replaced with CO_2 and the boiler characteristics remain close to the air-fired base-case.

Figure 1 represents a schematic of the proposed technology for an existing boiler. The combustion process includes the different streams of oxidizer, namely the primary oxidant (PO), secondary oxidant (SO), and tertiary oxidant (TO), respectively. All these streams, traditionally air, could be replaced, in part or in totality with oxygen and recycled flue gas. This way the volume of flue gas exiting the boiler is greatly reduced, allowing a reduction in the size of the flue gas post-treatment, namely the particulate removal (ESP), and NO_x, SO_x, Hg, etc. reduction devices. Finally, the cleaned flue gas is highly concentrated in carbon dioxide, allowing its capture and/or sequestration. The whole process may make the stack optional, leading to a zero-emission process.

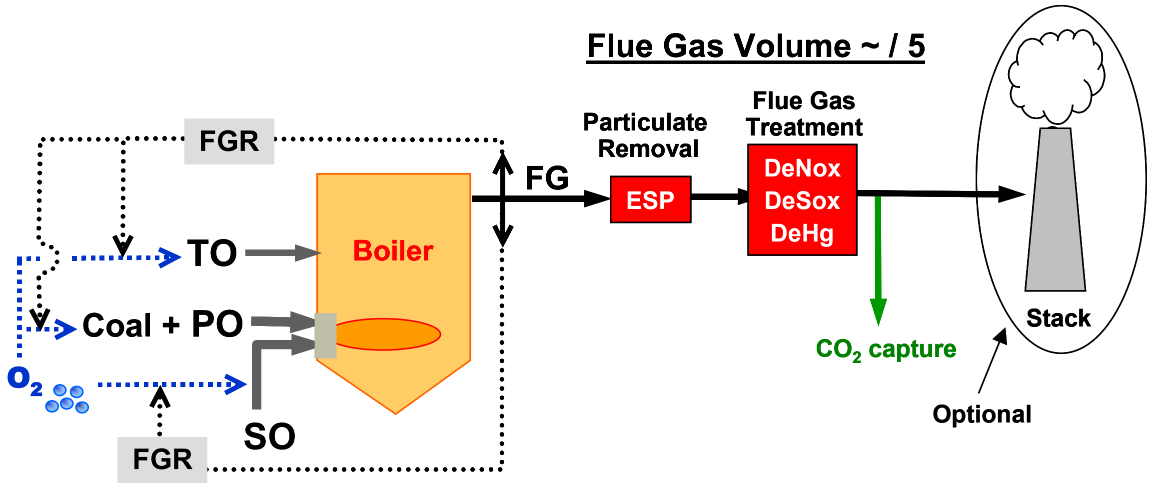

Figure 1. Retrofitted oxy-fired boiler with Flue Gas recirculation and flue gas post-treatment, including carbon dioxide capture.

This paper presents experimental results of oxy-combustion in a coal-fired pilot boiler, including flue gas recirculation. A companion paper presented during this conference shows the economical advantages of oxy-combustion in boiler and flue gas post-treatment costs.

PREVIOUS WORK

Preliminary, lab-scale experiments have been conducted by Abele *et al.* (1987), Miyamae *et al.* (1994), Kimura *et al.* (1995), Croiset and Thambimuthu (1998), highlighting the main characteristics of coal-combustion in O_2/CO_2 atmospheres, and concluding in the technical feasibility of such operating conditions.

Several simulation studies have been performed to assess the impact of parameters like air infiltration or oxygen purity on combustion characteristics such as: furnace outlet temperature or CO_2 concentration in flue gas. The HYSIS simulations of Zhang *et al.* (1998) indicate that the conversion to O_2-firing of a typical 400MWe PC air-fired power plant is possible with minimal impact on boiler characteristics. Such a conversion could be easily implemented, at a minimal cost while retaining the option of transition back to air-firing.

At the same time, engineering feasibility studies have been carried out to evaluate the economics of converting an existing steam and/or power plant to oxygen-firing with flue gas recirculation. Such studies consider the entire plant from fuel and oxidant handling to post-treatment requirements. The feasibility study conducted by Wilkinson *et al.* (2000 and 2001) on an existing refinery power station boiler, mentioned some related concerns including the impact of O_2 firing on boiler efficiency (higher radiative power of CO_2 as compared to N_2), air infiltration into the boiler and/or oxygen purity requirements. Simbeck (2001) developed a general spreadsheet to calculate, under customized assumptions, the cost of electricity produced by any retrofit boiler with a CO_2 capture option and applied it to a hypothetical 300MWe plant. Similar comparative analyses have been performed by Nsakala *et al.* (2001) in order to compare different retrofit concepts for CO_2 capture, such as air-fired coal combustion followed by amine-based CO_2 separation, or oxy-firing with flue gas recycle. These studies indicate that oxygen-enriched combustion is technically and economically feasible compared to alternative CO_2 control processes.

In addition to the theoretical studies such as those presented above, to-date there have been few reported experimental efforts at a relevant scale. Extensive experimental lab-scale work is reported by Thambimuthu and Croiset (1998) on a 0.3MWth oxy-coal cylindrical combustor, including flue gas recycle. The results show that oxy-combustion with flue gas recycle is a feasible retrofit for coal-fired burners.

The next step from an experimental standpoint is to apply the technology on a pilot boiler, as described below.

OXY-COMBUSTION DEMONSTRATION IN A COAL-FIRED PILOT BOILER

A team consisting of Air Liquide (AL), Babcock and Wilcox (B&W), the Illinois State Geological Survey (ISGS) and sponsored by the Illinois Department of Commerce and Economic Opportunity through the Office of Coal Development and the Illinois Clean Coal Institute has conducted a series of experimental tests on the B&W pilot boiler, using Illinois coal. Both low- and high-oxygen enrichment tests have been conducted (Marin *et al*. 2002).

The pilot boiler, presented in Fig. 2, is a 1.5MW (5 MMBtu/hr) pilot-scale boiler simulating a utility boiler. Both the radiative (furnace) and the convective sections of the pilot are representative in terms of geometry and heat exchangers equipments of a utility boiler.

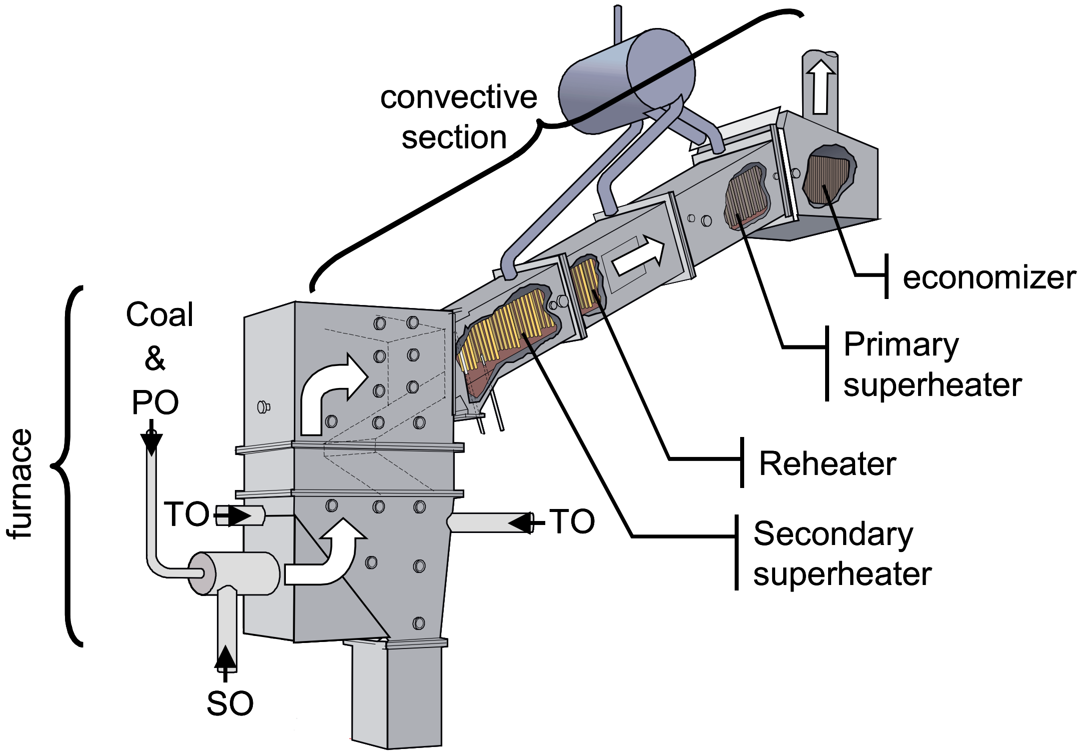

Figure 2. 1.5MW Pilot-Boiler.

As shown in Figs. 2 and 3, the system is equipped with the following oxidant streams:

- The primary oxidant (PO) conveys the pulverized coal from the mill (pulverizer) to the boiler, through the "core" of the burner. 15 to 20% of the overall O_2 needed to complete the combustion is contained in the PO in a conventional air-fired configuration.
- The secondary oxidant (SO) also enters the boiler at the burner level, and usually provides 50% to 85% of the O_2 requirements. The ratio between the oxygen provided by both PO and SO and the oxygen needed to complete the combustion is referred to as the Burner Stoichiometric Ratio (BSR).
- The tertiary oxidant (TO) is used for staged combustion (sub-stoichiometric combustion at the burner level). TO is injected into the pilot boiler through overfire air ports located on the same side with the burner (front ports), or on the opposite wall (rear ports). TO may represent up to 30% of the overall O_2 requirements and is currently implemented in low-NOx utility boilers. In this study, the Overall Stoichiometric Ratio (OSR) refers to the ratio between the O_2 contained in PO+SO+TO and the O_2 needed for complete combustion.

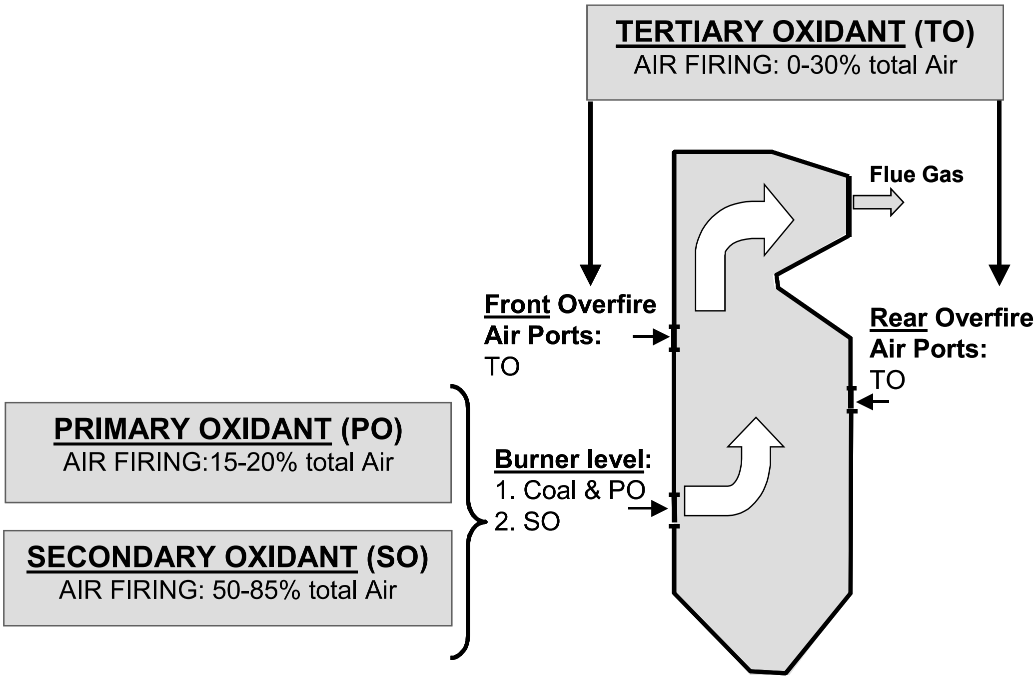

Figure 3. Oxidant streams in the pilot boiler.

Modifications were carried out to allow the pilot to be operated with O_2-enriched flue gas, including the oxygen delivery system. Measurements on oxidant and coal streams as well as on streams leaving the furnace provided accurate values of flowrates, gas temperature and gas composition (O_2, CO_2, CO, NO_x, SO_2). Fly Ash was sampled across the convection pass section exit and representative samples at each test condition were collected on a glass fiber filter and analyzed to determine the "unburned carbon in ash" (UBC) as a percentage of dry ash in the flue gases. It is noted that during the tests, no more than 80% of the overall air streams were replaced with oxygen and recycled flue gas.

Several tests were performed during the campaign, including the oxy-fired/flue gas recycle tests reported here. They include, as shown in Table 1, different degrees of recirculation, as well as the option of staging the combustion process. It is noted that for each state steady-state was achieved.

RECIRCULATION RATE / STAGING	0	LOW (1)	HIGH (2)
NO (a)	AIR-CASE a	OXY-CASE 1a	OXY-CASE 2a
YES (b)	AIR-CASE b	OXY-CASE 1b	OXY-CASE 2b

Table 1. Test Series characteristics.

The recirculation rate refers to the ratio between the flue gas recycled and flue gas leaving the furnace. Two levels of recirculation rate were tested during these tests, referred to as "LOW" and "HIGH" recirculation rate.

For each "recirculation rate" (Low or High), an unstaged combustion case (OXY-CASE 1a and OXY-CASE 2a) and a staged case (OXY-CASE 1b and OXY-CASE 2b) are tested. The baselines are provided by two air-fired cases, with and without combustion staging (AIR-CASE a and b).

All the reported experiments are performed with an identical excess of oxygen in the flue gas (around 3% on a dry volume basis) and with similar amounts of air infiltration into the boiler.

EXPERIMENTAL RESULTS

For the cases presented in Table 1, the impact of oxy-combustion on pilot operation, NO_x emissions and unburned fuel is determined in order to identify optimum conditions of the O_2/FGR combustion process.

Oxy-Combustion and Flue-Gas Recirculation Impact on NO_x, SO_x and Mercury Emissions

As shown in Table 2, the impact of oxygen-enriched flue gas was to lower NO_x emissions in both unstaged (a) and staged (b) cases: 53% and 69% NO_x reductions are measured as compared to the unstaged air-case a, in the low and high unstaged oxy-cases respectively. Similar effects were observed regarding the staged cases since 54% and 62% NO_x reductions were measured as compared to the staged air-case (b), in the low and high staged oxy-cases respectively.

Staging the combustion further magnifies the NO_x emission reduction, achieving 38% and 23% NO_x reductions with respect to the corresponding unstaged oxy-case, which corresponds to 71 to 76% NO_x emission decrease with respect to the regular unstaged air-baseline.

STAGING \ RECIRCULATION RATE	0	LOW (1)	HIGH (2)
		- 69%	
NO (a)	100	- 53% 47	31
	- 37%	- 38%	- 23%
YES (b)	63	- 54% 29	24
		- 62%	

Table 2. NO_x emissions assuming the baseline value in AIR-CASE (a) is 100.

These results are displayed in Fig. 4, indicating that three of the four measured NO_x emission rates are below the expected future US regulations of 0.15lb/MMBtu (65ng/J).

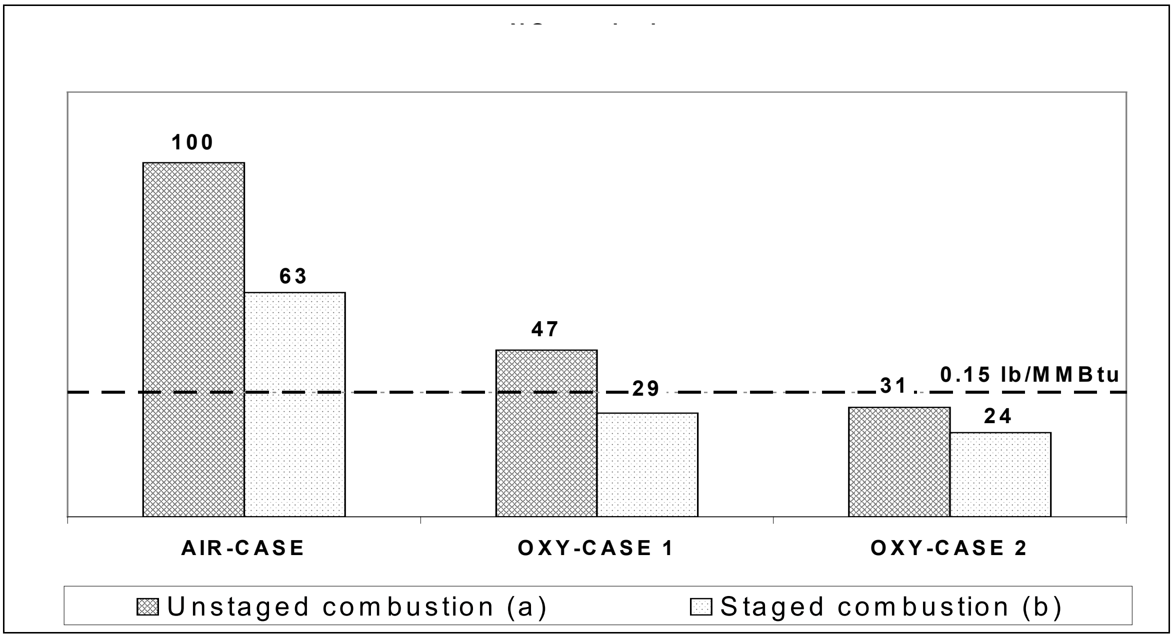

Figure 4. NOx emissions assuming the baseline value in AIR-CASE (a) is 100.

It is concluded that the tested technology represents an efficient way to not only meet but to achieve levels lower than the NO_x emission limits, making unnecessary the presence of flue gas, de-NO_x post-treatment.

The observed NO_x emission reductions using oxy-firing with flue gas recycle are in good agreement with Miyamae's (1994) and Okasaki's (1997) observations, explaining this drop as resulting from the chemical reduction of NO to molecular nitrogen N_2 by the reburning mechanism in the boiler. Not only is NO_x partially destroyed due to the "reburning" effect, but NO contributes also to decreasing the NO_x formation rate by promoting the reduction of fuel-bound nitrogen N to N_2 in the near burner region. Croiset and Thambimuthu (1999) further suggest that recirculated CO may also inhibit NO formation in the near burner region via a char catalyzed reaction.

However, though this NO_x reduction effect in O_2/FGR systems has already been described, none of the above-mentioned previous work achieved NO_x emissions below 0.15lb/MMBtu. In that regard, the experiments presented in this paper are the a clear demonstration of the O_2/FGR technology as a low NO_x retrofit technology enabling any power plant to comply with present and future Ozone Transport Region NO_x emission regulations.

The observed very low NO_x emissions are attributed to several phenomena:

- the "reburning" process promotes NO_x destruction, which is even more effective in fuel rich conditions, thus while staging the flame
- the fuel-N "reduction" process limits the fuel-NO_x formation, which is even more effective in fuel rich conditions, including staging the combustion process
- the decrease in flame temperature resulting from the combined phenomena of ① staged combustion and ② N_2 - CO_2 replacement, thereby limiting thermal-NO_x formation.

Additional experimental/analytical work is necessary to quantify the impact of each of these phenomena on the NO_x reduction mechanisms, in order to further optimize the process.

In addition to NO_x measurements, the experimental work investigated the impact of the tested technology on SO_x emissions. For this purpose, a number of tests included a wet FGD prior to flue gas release in the atmosphere. While more detailed results will be made public in a future publication, the oxy-fired case showed better SO_x removal characteristics than the air-fired case. The results are attributed in part to an increased residence time of the flue gas in the de-sulfurization equipment.

Finally, the tests included Mercury (Hg) measurements, for both the air-case and the oxy-fired case. The results currently being analyzed show a significant reduction in Hg in the oxy-fired case, of the order of around 50%, increasing the confidence that the oxy-firing may potentially constitute an efficient method to control mercury emissions in coal-fired boilers. A future publication will expand on this topic as well.

Oxy-Combustion and Flue-Gas Recirculation Impact on Boiler Efficiency

Unburned carbon in ash, also known as Loss on Ignition (LOI) provides an indication of the quality of the combustion process. The results presented here include LOI measurements for the base-case (air-fired operation), as well as for the low recirculation cases.

Table 3 and Fig. 5 compare the base-case - air operation without staging (defined as 100%), with several other cases. With air staging alone, the LOI increases by 20%, due to a deterioration of the combustion process in the boiler – this outcome is not unusual in industrial operations, where, instead of being a marketable product, ash becomes a cost to the operator. On the other hand, by using oxy-firing with flue gas recycle, the LOI decreases by 22% without staging, and 48% with staging, therefore the trend is reversed when compared to air firing. The explanation for these results resides in a good control of the combustion process in the case without staging. When staging, the oxygen-rich tertiary oxidant stream enhances the char combustion process, leading to low LOI. This phenomenon is presented in the low-oxygen enrichment paper presented during this conference.

STAGING \ RECIRCULATION RATE	0		LOW (1)
NO (a)	100	- 22%	78
	+20%		-33%
YES (b)	120	- 56%	52

Table 3. Unburned Carbon in Ash assuming the baseline value in AIR-CASE (a) is 100.

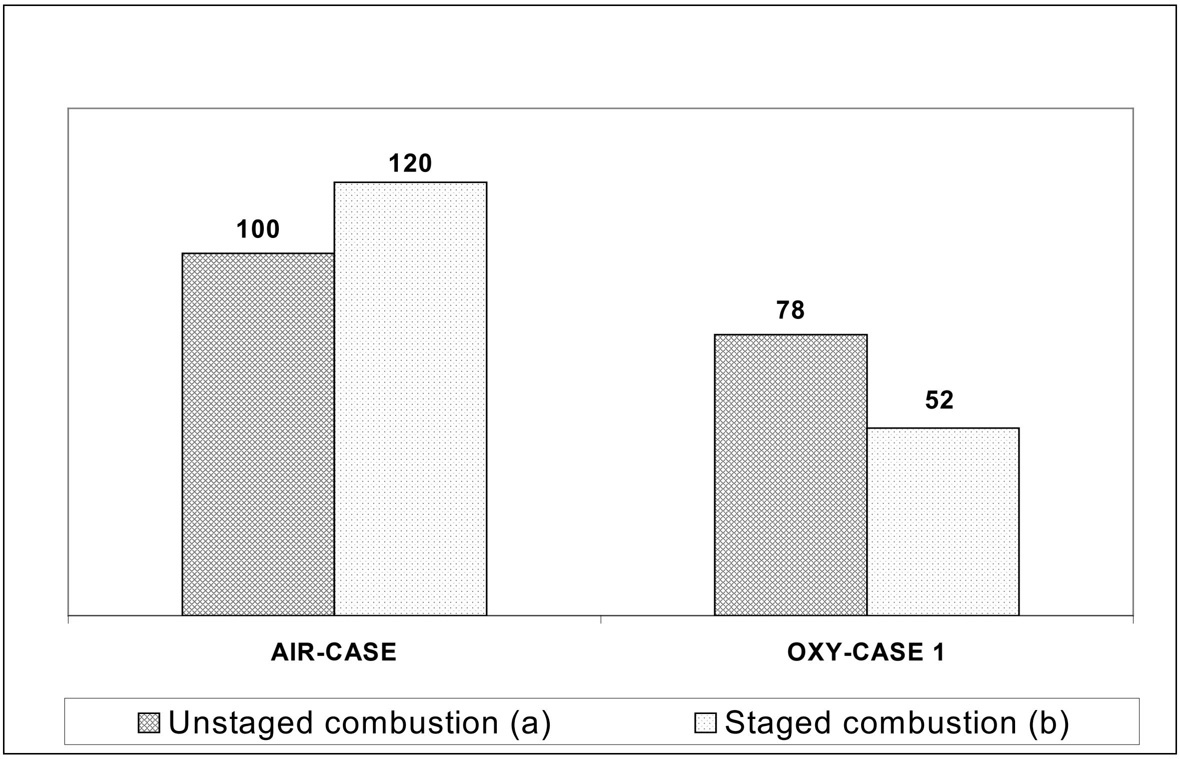

Figure 5. Unburned Carbon in Ash assuming the baseline value in AIR-CASE (a) is 100.

In conclusion, oxygen enrichment is an efficient tool in reducing the LOI content of the ash. The resulting benefit is that, with sufficiently low LOI levels, the boiler fly ash can be marketed as a valuable by-product rather than being landfilled. In addition, reduced LOI equates into a higher combustion efficiency and better utilization of the fuel (fuel savings or increased plant productivity).

The results obtained during these tests increase the confidence that future industrial implementations of ultra-low emission oxy-coal fired boilers can be achieved with limited flue gas post-treatment. Figure 6 shows such a possible scenario, where the compact particulate removal system will provide high-quality ash, and the only other post-treatment phase will include a SO_x removal system, and possibly a CO_2 capture process. The stack will be optional, in case of carbon sequestration, resulting in a zero-emission process.

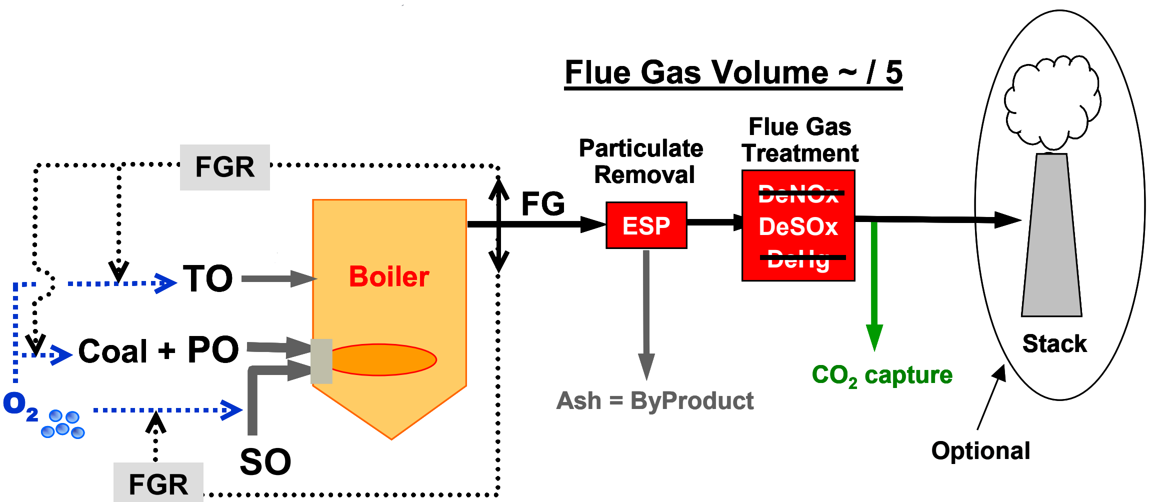

Figure 6. Oxy-firing Steam or Power Generation Plant including the new demonstrated benefits in addition to the reduced flue gas volume and the CO_2 capture option.

Technical Feasibility Demonstration for Oxy-firing retrofit technology

During the tests presented in this paper, a smooth transition from air to oxy-combustion has been demonstrated. No upsets were noted, and the control of the process was observed to be efficient and fast.

The exit temperatures measured for the air and oxy cases displayed no significant differences – the slightly higher temperatures for the oxy-case were attributed to increased combustion efficiency and the absence of N_2 to dilute and quench the flue gas. This supports the argument that the oxy-firing technique can be implemented as an effective retrofit technology, as it does not significantly alter the heat transfer patterns. It is also noted that for retrofit applications, engineering feasibility studies are recommended to evaluate the impact of oxy-fuel combustion on boiler temperatures and steam generation. In addition, optimizing the oxygen and flue gas injection locations in the boiler provides an easy way to optimize the oxygen concentration in different parts of the boiler. This flexibility can be used to enhance the flame stability, to control local temperatures in the furnace, to increase the local oxidizing characteristics of the furnace gases, or to enhance mixing.

It is concluded that this technology is likely to be easily implemented in existing coal-fired plants with minimal plant modifications, and enhanced operation control.

CONCLUSIONS

This paper presents the experimental results of a 1.5MW oxy-coal fired pilot boiler, including flue gas recirculation.

Following several theoretical studies and lab-scale experiments, these tests represent a major step in demonstrating an ultra-clean coal combustion process, targeting the utility boiler market. In the context of increased emissions regulations, including greenhouse gas containment, these experimental data increase the confidence in oxy-combustion processes

The tests show that the impact of oxy-combustion on NO_x emissions is very significant, with emissions lower than 0.15 lbs NO_x/MMBTU (65ng/J), exceeding future Ozone Transport Region regulation. This result is very significant, as it makes the expensive de-NO_x equipment unnecessary, and thus simplifies the process.

The tests also show effective removal of SO_x using conventional wet FGD equipment, and also a significant reduction in mercury emissions. These results are in the process of being analyzed and better understood.

In addition to emission reductions, easy conversion from air-firing operation to oxy-firing operation has been achieved, with favorable flame stability and heat transfer characteristics.

In conclusion, these experimental results demonstrate that oxy-combustion with flue gas recirculation represents an ultra clean combustion process, leading to the reduction in unburnt fuel, in addition to the lowered emissions and increased carbon dioxide concentration. The flexibility of the process, in terms of optimizing the amounts of oxidant in the various parts of the combustion process is considered to be an important feature of the technology.

Future work will continue to optimize the process. Thus, the entire amount of air will be replaced by flue gas recirculation and the more widely used PRB coal will be tested as well. In addition, as the economics of the process is affected by the size of the boiler, a new, significantly smaller full oxy-fired boiler design will be considered.

It is also noted that a full industrial demonstration of the process can be extremely beneficial in advancing the technology. It is strongly believed that the technical hurdles can be easily overcome, as proven in the tests presented here.

ACKNOWLEDGMENTS

The authors wish to thank the Office of Coal Development of the Illinois Department of Commerce and Economic Opportunity and the Illinois Clean Coal Institute for the support and guidance provided during the project.