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Air Separation, flue gas compression and purification units for oxy-coal combustion systems

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Abstract

Air Liquide (AL) has been actively involved in the development of oxy-coal technologies for CO2 capture from power plants for almost 10 years. Large systems for oxygen production and flue gas purification are required for this technology. Air Liquide has been a leader in building large Air Separation Units (ASUs) and more developments have been performed to customize the air separation process for coal-fired power plants. Air Liquide is also actively involved in developing processes for purification of flue gas from oxy-coal combustion systems for enhanced oil recovery applications as well as sequestration in saline aquifers. Through optimization of the overall oxy-coal combustion system, it has been possible to identify key advantages of this solution: minimal efficiency loss associated with CO2 capture (less than 6 pts penalty on HHV efficiency compared to no capture), near zero emission, energy storage, high CO2 purity and high CO2 recovery capability.

Keywords: oxy-combustion; coal; air separation unit; CO2 capture; clean coal ; CO2 purification unit.

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1. Introduction

Oxy-combustion of pulverized coal is one of several proposed technologies for clean coal power generation. The core concept of oxy-combustion is the use of a high purity oxidant stream for the combustion process so that the combustion products are highly concentrated in CO_2, thus simplifying the CO_2 capture process. In 2007, Air Liquide contributed together with Babcock & Wilcox (B&W) to a US Department of Energy (DOE) report[1]: "Pulverized Coal Oxy-combustion Power Plants, Vol. I: Bituminous Coal to Electricity", DOE/NETL Report 2007/1291. AL and B&W provided performances and costs for an oxy-boiler, an Air Separation Unit (ASU) and a CO2 compression and purification unit (CO2 CPU) for Case 5, 5A, 5B, 5C, 6 and 6A. After analysis of the results of this report, AL and B&W decided to do a joint study to reoptimize Case 5 (Pulverized Coal – Supercritical Steam) and 6 (Pulverized Coal – Ultrasupercritical Steam).

Figure 1 show the results of this joint study compared to the original results of the DOE/NETL report:

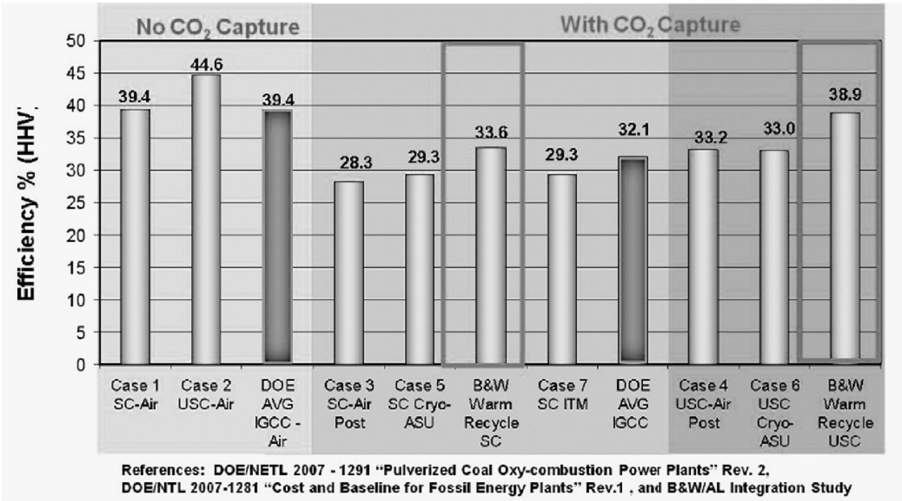

Figure 1: HHV efficiency of different technologies for CO2 capture

Without capture, the HHV efficiency of the state-of-the art Pulverized Coal power plant with Supercritical Steam was 39.4% (Case 1) identical to the average of the IGCC cases without capture. With CO2 capture, the HHV efficiency dropped to 28.3% with post-combustion capture with amines (Case 3), to 32.1% for the average of the IGCC cases and originally to 29.3% with oxy-combustion capture (Case 5). After reoptimizing this case with B&W, it was possible to achieve 33.6% with today technology.

With an Ultrasupercritical Steam cycle, the no capture Case 2 was at 44.6% efficiency, dropping to 33.2% with post-combustion capture with amines (Case 4) and originally to 33.0% with oxy-combustion capture (Case 6). After reoptimization, the HHV efficiency was improved to 38.9% (very close to the efficiency of Case 1).

This paper presents Air Liquide's efforts in improving the viability of coal oxy-combustion through technological advances in the ASU and CO_2 CPU for CO_2 capture.

2. Air Separation Unit

2.1. ASU for coal oxy-combustion

A commercial-scale coal-fired oxy-combustion power plant would require thousands of tons of oxygen each day. Cryogenic distillation is the only commercially available technology today to produce such large quantities of O_2 economically. Other air separation technologies like pressure swing adsorption (PSA), vacuum swing adsorption (VSA) or polymeric membranes cannot compete economically for such quantities. Ceramic membranes (oxygen ion transport membranes) are not yet commercially available for large-scale oxygen production and so it is hard to compare them to cryogenic distillation both in terms of investment and performance.

Cryogenic ASU is considered to be a mature technology. However, the industry has been able to achieve great improvements over the last 3 decades in improving this technology.

The main characteristics of an ASU for oxy-coal combustion are: large size (typically beyond 8000 tpd for industrial-scale plants), low pressure (between 1.3 and 1.7 bar abs) and possible low oxygen purity. Low oxygen purity means a value in the range of 85-98% O2 content compared to the typical 99.5-99.8% O2 content of the high purity units. This allows significant savings in power consumption in the ASU.

The cycles for the production of low purity oxygen at 95% were extensively developed at the beginning of the 1990s essentially for 2 applications: gasification (including IGCC) and oxygen enrichment of blast furnace vent streams. At that time, Air Liquide designed several plants for these applications and could demonstrate specific energy of separation around 200 kWh/t of pure O2 when the cost of power was high (kWh/t is written as SI units meaning kilowatt-hours per metric ton). Air Liquide is currently operating several plants in Italy with this specific energy of separation. Energy of separation is defined as the power required to produce 1 metric ton of pure oxygen contained in a gaseous oxygen stream at a given oxygen purity at atmospheric pressure (101325 Pa) under ISO conditions (15°C, 60% relative humidity). Compressors driver efficiency (electrical motor, steam or gas turbine), heat of regeneration of driers and power consumption of the cooling system are not considered in this definition.

The cycles developed in the 1990s were not fully adapted for oxy-combustion. For example, they were optimized to produce relatively high pressure oxygen (from 5 bar abs to 80 bar abs) and in some cases to perform co-production of nitrogen. In 2007, Air Liquide launched an ASU development program in order to develop an Air Separation Unit optimized for oxy-combustion. The idea was not to fully redesign an Air Separation Unit but just to adapt the process cycle to the specific requirements of oxy-combustion (i.e. low oxygen pressure, no nitrogen requirement) and also to include technology improvements that have been demonstrated in other Air Separation Units since the 1990s. Thus far in this project, the energy requirement of the ASU has been improved from 200 kWh/t to less than 160 kWh/t with heat integration.

Further optimizations of the Air Separation Unit have been identified and are currently under development and it is expected to achieve separation energy around 140 kWh/t with heat integration in 2015.

Figure 2 shows the magnitude of these improvements. This trend is expected to continue in the future, since the overall energy of separation is still significantly greater than the theoretically required separation energy.

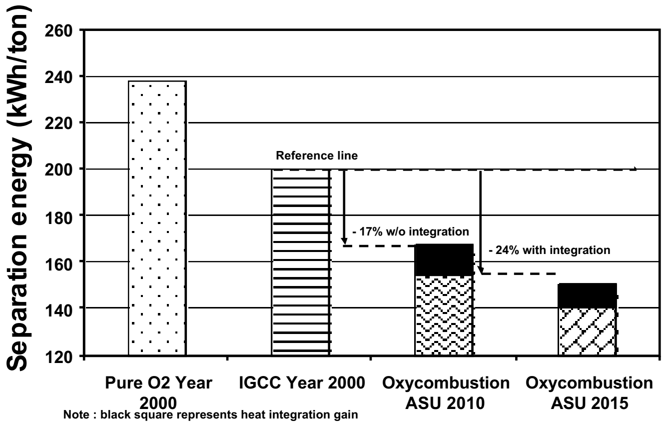

Figure 2: Improvements in energy efficiency of cryogenic ASU.

2.2. Heat integration

Heat integration consists in transferring heat from the ASU compressor(s) to the steam cycle. Two benefits can be achieved through this integration:

- Energy losses associated with compression can be reduced
- Energy losses associated with condensates or boiler feed water preheating can also be reduced

This transfer of heat can be direct (feed water preheating) or indirect (oxygen preheating, coal drying, heating of any fluid of the oxy-combustion cycle). Air Liquide has performed several studies on heat integration with Babcock & Wilcox. In some cases, a reduction of ~10% in power consumption of the air separation unit could be achieved. One of the conclusions of these studies is that these gains are very dependent on the design of the overall plant: ambient conditions, efficiency of the steam cycle, cooling system (dry versus wet), coal type (water and sulfur content) *etc.*

2.3. Energy storage and flexibility

Energy storage is another area of development for AL as it is possible to associate it with the technology of cryogenic ASU which means that, at one point in the process, oxygen is produced in a liquid form which can be easily stored. The idea behind energy storage is very simple:

- store liquid oxygen at off-peak hours (typically at night or when power from wind is available in great quantities)
- unstore liquid oxygen at peak hours

As a result, it is possible to produce extra power at peak hours corresponding for example to 50% of the power requirements of the ASU. At off-peak hours, the liquid oxygen could for example be stored by running the ASU at 110% capacity.

Table 1 shows the benefits of such a system for a 550 MW plant:

	Average	**Peak**	**Off-peak**
Gross power from steam (MW)	**724.1**	**721.3**	**724.6**
Gross power for ASU (MW)	75.8	37.9	83.4
Gross power for CO2 CPU (MW)	62.4	62.4	62.4
Primary, FD & ID fans & miscellaneous (MW)	35.9	35.9	35.9
Sub-total Auxiliary load (MW)	**174.1**	**136.2**	**181.7**
Net plant power (MW)	**550.0**	**585.1**	**542.9**
HHV efficiency	**33.6%**	**35.7%**	**33.2%**

Table 1: Energy storage

As can be seen in this table, 35.1 MW additional power can be produced at peak hours for a 550 MW plant i.e. 6.4% additional power.

In term of flexibility, AL has demonstrated in ASUs for IGCC that at least 5% per minute capacity change can be achieved. In other words, the ASU can be ramped down from 100% to 50% in less than 10 minutes (or ramped up from 50% to 100% in the same time).

3. CO_2 Compression and Purification Unit

The role of the CO_2 CPU is primarily to compress CO_2 at the required pressure for transport and storage and to purify the CO_2 rich flue gas to the required specifications.

When designing such a unit, two main considerations are to be taken into account:

- Ability to deal with all the impurities contained in the flue gas to be processed: water, SOx, NOx, Hg, Particulates, N_2, Ar, O_2, CO…while delivering the required CO_2 stream quality and controlling emissions
- Ability to balance performances (specific power and CO_2 recovery) and cost of the processing unit

Please refer to "Air separation and flue gas compression and purification units for oxy-coal combustion systems" paper[2] for more details on CO_2 CPU process schemes.

3.1. Impurities contained in the flue gas

As all the flue gas produced by the plant is compressed, it is possible to achieve a near zero emission plant. In particular, conventional emissions i.e. NOx, SOx, Particulate and Mercury are expected to be below detectable limit.

In addition, it is possible to remove the impurities from the flue gas at the most economical location between low pressure (close to atmospheric pressure) and high pressure. Extensive work has been performed by Air Liquide regarding the removal of impurities from the flue gas either at lab test level or at pilot plant level demonstrating the feasibility of very high CO_2 purity (99.99%+) at a reasonable cost.

3.1.1. Sulfur

Sulfur is the key impurity to manage, with the highest stakes in term of capital and operating expenditure.

For the first generation of CPU, Air Liquide has chosen a two-step proven solution for sulfur removal:

- Classical FGD (Flue Gas Desulphurization) to go from 1000 ppm to 50 ppm, using calcium reagent (limestone $CaCO_3$ and/or lime CaO).
- Polishing with sodium reagent to go from 50 ppm to 1 ppm, using trona (a natural sodium carbonate), caustic soda (NaOH) or soda ash (Na_2CO_3).

For the 2^{nd} generation of CPU, two strategies are currently evaluated:

- Low pressure drier to avoid/limit sulphuric acid formation during the compression process and production of liquid SO_2 and possibly NO_2 e.g. for co-sequestration with CO_2
- Use NO_2 as reagent for SO_2 conversion to sulphuric acid at low pressure before the flue gas compressor

3.1.2. Particulate matter

Two strategies are possible for the management of particulate matter (dust):

- Allow fouling with related consequences for maintenance
- Remove particulates to the same level as the one used for centrifugal compressors in ASU

Air Liquide is working on the 2 strategies in order to propose the best solution for any specific requirement. In particular, pilot tests of a dust abatement system have been performed on real flue gas to qualify the feasibility of the second option.

3.1.3. Mercury

Two strategies are possible for the management of mercury:

- No specific mercury removal unit because equipment such as scrubbers, flue gas condenser, coolers may remove enough mercury to have a level compatible with the use of brazed aluminum heat exchangers especially in the presence of strong acids like H_2SO_4 and HNO_3.
- Adsorption of mercury in a guard bed

3.1.4. Water

Adsorption is a well proven technology for water removal but the adsorbent needs to be carefully selected in order to resist to acids like H_2SO_4 and HNO_3.

3.2. Performance

3.2.1. Energy efficiency

Heat integration like in the ASU is a first step to decrease the specific energy of the CO2 CPU. Further improvements in process are also expected to enable further reduction in the power consumption.

3.2.2. CO2 recovery

Although liquid condensation at low temperature assures a CO2 recovery of 90% at a reasonable specific energy, it is easily achievable to increase CO2 recovery without major increase and in some cases with slight decrease in the marginal CO2 capture cost expressed in euro or dollar per ton.

It consists in adding a CO2 recovery system on the non-condensable stream of the CO2 CPU e.g. based on swing adsorption, absorption or permeation technology. With such addition, CO2 recoveries above 95% could be achieved.

4. Conclusions

Table 2 below summarizes AL targets in term of improvement of the efficiency of the oxy-coal combustion solution:

	2008 study	2015 target	Delta
Gross power from steam (MW)	**724.1**	**729.8**	**+0.8%**
Gross power for ASU (MW)	75.8	68.2	-10%
Gross power for CO2 CPU (MW)	62.4	56.2	-10%
Primary, FD & ID fans & miscellaneous (MW)	35.9	37.3	+3.9%
Sub-total Auxiliary load (MW)	174.1	161.7	-8%
Net plant power (MW)	**550.0**	**568.1**	**+3.3%**
HHV efficiency	**33.6%**	**34.7%**	**+1.1pt**

Table 2: Efficiency improvements

Therefore, by 2015, the objective is to achieve an HHV efficiency loss of only 4.7 points compared to the case without capture (34.7% versus 39.4%). In parallel, it is also intended to decrease the capital expenditure of the plant and in particular of the ASU and the CO2 CPU.

Air Liquide has made several technical advances to increase the attractiveness of the oxy-combustion process for clean electricity production from coal.

Through optimization of the overall oxy-coal combustion system, it has been possible to identify key advantages of this solution: minimal efficiency loss associated with CO2 capture (less than 6 pts penalty on HHV efficiency compared to no capture), near zero emission, energy storage, high CO2 purity and high CO2 recovery capability.

5. References

[1] "Pulverized Coal Oxy-combustion Power Plants, Vol. I: Bituminous Coal to Electricity", DOE/NETL Report 2007/1291, Rev 2, Aug. 2008.

[2] A. Darde et al., " Air separation and flue gas compression and purification units for oxy-coal combustion systems", Energy Procedia 1 (2009) 527-534.