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## Air separation and 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 CO<sub>2</sub> capture from power plants for the past 5 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 cryogenic air separation process for oxy-coal combustion, a 20% reduction in the separation energy requirement of the ASU has been achieved. Several CO<sub>2</sub> capture schemes have been developed to study the energy requirement to achieve CO<sub>2</sub> product streams meeting different specifications. Utilization of waste heat from both the ASU and CO<sub>2</sub> capture unit for heating boiler feed water has been shown to significantly improve the energy efficiency of the overall process.

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Keywords: oxy-combustion; coal; cryogenic air separation unit; CO<sub>2</sub> capture; clean coal.

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### 1. Introduction

To produce electricity from coal in an environmentally responsible manner, it is critical to capture CO<sub>2</sub> from coal power plants. 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<sub>2</sub>, thus simplifying the CO<sub>2</sub> capture process. A recent US Department of Energy (DOE) report [1] has indicated that oxy-combustion processes are comparable to air-combustion-based CO<sub>2</sub> capture processes such as amine absorption, with respect to the overall cost of clean power production. The report also indicated that further improvements are required for all current clean-coal technologies to reach DOE's goal of 90% CO<sub>2</sub> capture with no more than 20% increase in the cost of electricity.

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This study presents results of Air Liquide's efforts in further improving the viability of coal oxy-combustion through technological advances in the main components of the oxy-combustion process: the air separation unit (ASU) for oxygen production and the CO<sub>2</sub> compression and purification unit (CO<sub>2</sub> CPU) for CO<sub>2</sub> capture.

## 2. Air Separation Unit (ASU)

### 2.1. Cryogenic ASU Technology

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<sub>2</sub> economically and at high purity. Other air separation technologies like pressure swing adsorption (PSA), vacuum swing adsorption (VSA) or polymeric membranes cannot compete economically for such quantities, especially to produce >95% purity oxygen. 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. Nevertheless, the industry has been able to achieve great success over the last 3 decades in improving this technology. Figure 1 shows the magnitude of these improvements both in terms of productivity of the distillation columns and in their energy efficiency. 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.

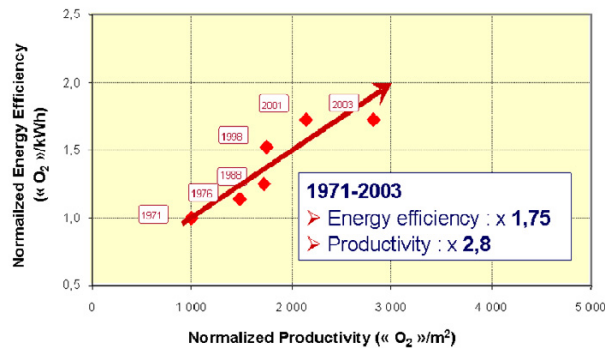


Figure 1: Improvements in productivity and energy efficiency of cryogenic ASU.

### 2.2. ASU for Oxy-coal Combustion

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% O<sub>2</sub> compared to the typical 99.5-99.6% O<sub>2</sub> of the high purity units. This allows significant savings in power consumption in the ASU as shown in Figure 2.

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 O<sub>2</sub> when the cost of power was high. 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 a gaseous oxygen stream at a given oxygen purity at atmospheric pressure

(101325 Pa) under ISO conditions (15°C, 60% relative humidity). The efficiency of the compressor motors, heat of regeneration of driers and power consumption of the cooling system are not considered in this definition.

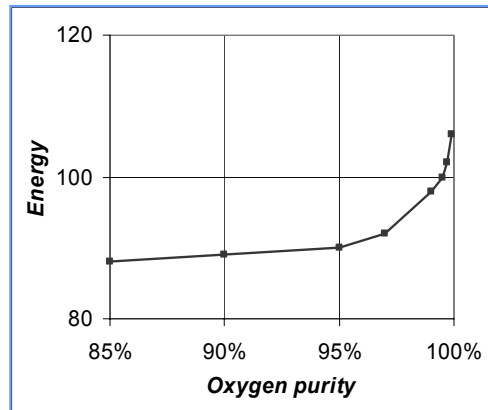


Figure 2: Power requirement of cryogenic ASU.

The cycles developed in the 1990s were not fully adapted for oxy-combustion. For example, they were optimized to produce relatively high pressure oxygen (5-80 bar abs) and in some cases to perform co-production of nitrogen. In 2007, Air Liquide launched an ASU development program 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 for other Air Separation Units since the 1990s. Thus far in this project, ASU designs requiring as low as 160 kWh/t have been developed.

### 2.3. Process scheme

An ASU consists of the following equipment:

- Main air compressor
- Precooling system
- Purification unit to remove water and CO<sub>2</sub> prior to entering the cryogenic section
- Heat exchangers
- Distillation columns
- Vaporizers/condensers

For ASU's up to 5,000 tons/day, Air Liquide proposes a process with a double column scheme with no duplication of equipment: one purification unit for water and CO<sub>2</sub> removal with its proprietary radial bed design, one high pressure column and one low pressure column.

### 2.4. Tradeoff between capital and operating costs in ASU design

Figure 3 illustrates the tradeoff between capital and operating expenditures of an Air Liquide ASU unit. The capital expenditure for the previous design of 200 kWh/t has been normalized to 100, and its tradeoff curve is indicated by the red line. The tradeoff for the improved ASU design for oxy-combustion is indicated by the dotted line. From Figure 3, the optimization of the ASU design has led to a significant reduction in the specific energy of separation of the ASU at any given capital cost. For example, at a fixed normalized capital expenditure of 100, the improved ASU requires 20% less specific energy (*i.e.* 160 kWh/t vs. 200 kWh/t) than the previous ASU. The trade-

off between capital expenses (CAPEX) and operating expenses (OPEX) for the improved ASU is ~4000 \$/kW or 3000 €/kW.

### 2.5. 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 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. In some cases, a reduction of ~10% in power consumption of the ASU 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.*

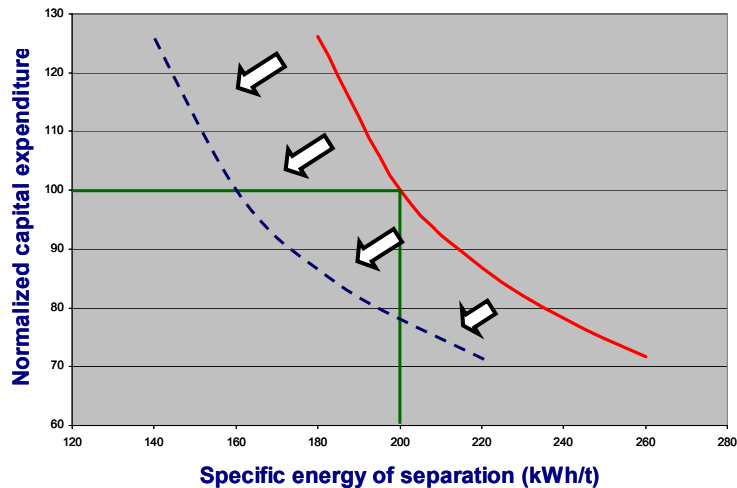


Figure 3: Tradeoff between capital and operating expenses in ASU design.

## 3. CO<sub>2</sub> Compression and Purification Unit (CO<sub>2</sub> CPU)

### 3.1. Considerations for CO<sub>2</sub> CPU design

The role of the CO<sub>2</sub> CPU is to capture CO<sub>2</sub> from combustion flue gases and purify it to the required specifications. Thus, both the composition of flue gases and CO<sub>2</sub> product specifications have a strong influence on the design and cost of the CO<sub>2</sub> CPU. In addition, the desired product pressure, the tradeoff between CAPEX and OPEX, and any value attached to the CO<sub>2</sub> product influence the overall design.

The composition of flue gases can be significantly influenced by the amount of air infiltration in the boiler and associated equipment such as fans, filters, piping and the desulphurization unit. Existing old plants are likely to have significant air infiltration. In contrast, new oxy-combustion plants can be designed to have minimal air infiltration. Thus, CO<sub>2</sub> capture units designed for retrofitting existing power plants would have to treat a flue gas stream that is

diluted in CO<sub>2</sub> relative to a stream from a new oxy-combustion power plant boiler (*cf.* Table 1). New plants also provide greater flexibility in integrating the CPU with the rest of the plant and realizing savings through exchange of process streams for heating, cooling *etc.*

Table 1: Estimated flue gas compositions (mole fractions) from new & retrofit oxy-combustion coal power plants using Illinois No. 6 coal [1,2].

Component	New plant [1]	Retrofit plant [2]
Ar	0.0305	0.0250
CO <sub>2</sub>	0.6963	0.5818
H <sub>2</sub> O	0.1661	0.1743
N <sub>2</sub>	0.0818	0.1763
O <sub>2</sub>	0.0253	0.0411
SO <sub>2</sub>	0.0001	0.0010
NO <sub>x</sub>	0.0000	0.0004
CO	0.0000	0.0001

Today there is no commonly agreed upon CO<sub>2</sub> product specification for CO<sub>2</sub> sequestration. Several specifications exist for CO<sub>2</sub> transport through pipelines. [3] However, these specifications are usually a result of considerations of both pipeline transport as well as the requirements of the end application, and are hence not always directly applicable to CO<sub>2</sub> sequestration in saline aquifers. Also, these specifications are for CO<sub>2</sub> derived from natural sources or from reducing environments such as gasification plants and hence do not include components like SO<sub>x</sub> and NO<sub>x</sub> that are present in boiler flue gas. In this study, all cases produced a dry product with at least 90% CO<sub>2</sub> recovery. Other component specifications were treated as parameters for study.

### 3.2. CO<sub>2</sub> CPU schemes

The basic process for processing flue gas to capture CO<sub>2</sub> is as follows:

1. Compression of wet flue gas.
2. Drying of the flue gas at the outlet of the “wet compression” step.
3. Flue gas purification (if considered).
4. Compression of the dry product gas to a pressure at which it condenses at 20°C.
5. Pumping of the condensed product to pipeline pressure.

The combination of compression, condensation and pumping minimizes power consumption of the process. However, condensation and pumping processes can be used only in cases where the condensation pressure is lower than the final product pressure. The flue gas purification step can contain several unit operations to meet the specifications on different gas components.

For this study, the following flue gas purification schemes were considered:

**Case 1: No purification:** In this scheme, the flue gas purification step is skipped. Thus, the entire flue gas is compressed, dried and compressed again (pumped, if possible) to the final product pressure of 175 bar, resulting in 100% CO<sub>2</sub> recovery. However, the CO<sub>2</sub> purity of this product is quite similar to that in the flue gas, on a dry basis. Hence this scheme is useful only for plants where the flue gas purity is quite close to the desired product purity. In addition, this scheme does not target reduction in any other gas components and hence cannot meet any specifications on minor components in the product stream. For example, such a process would typically have O<sub>2</sub> in the single percentage range, which is not acceptable for enhanced oil recovery applications.

**Case 2: Partial condensation (Cold box):** In this scheme, the compressed and dried flue gas is cooled to a very low temperature to condense out at least 90% of the CO<sub>2</sub>. CO<sub>2</sub> purity in the condensed phase is a function of the pressure and composition of the inlet gas to the partial condenser system, the number of stages of partial

condensation in the cold box and the condensation temperature(s); 95% CO<sub>2</sub> purity is usually achieved quite easily for typical flue gas compositions. This scheme can deliver product with O<sub>2</sub> in the thousands of ppm range.

**Case 3: Cold box including distillation:** This is an extension of case 2 in which a distillation column is used to further purify the condensed CO<sub>2</sub> stream in the cold box. This scheme also targets 90% CO<sub>2</sub> recovery. CO<sub>2</sub> purity in excess of 99% is typically achieved in this scheme. The distillation column also helps to reduce the O<sub>2</sub> content to the low ppm range.

Figure 4 shows the specific power consumption of the above schemes as a function of flue gas composition for a CO<sub>2</sub> product pressure of 175 bar. From Figure 4, with increasing CO<sub>2</sub> concentration in the inlet flue gas, the power requirement of the CPU unit decreases. This is because a lower volume of total gas has to be compressed and treated to capture the same quantity of CO<sub>2</sub>. At about 72% inlet CO<sub>2</sub> (dry basis), which is in the range for a retrofit plant, just compression of the gas to the final pressure requires nearly 170 kWh/t of energy. In contrast, compressing the gas to an intermediate pressure and purifying it in a cold box, with or without a distillation column, requires only about 140 kWh/t of energy and produces a much higher purity product (~95-99%+ CO<sub>2</sub>).

As the CO<sub>2</sub> concentration in the inlet flue gas increases, less energy is required for all processes to meet product specifications. If the inlet CO<sub>2</sub> concentration is ~ 93%, the 3 processes take nearly the same amount of specific power. However, the CO<sub>2</sub> purity of the product is significantly above 95% for the purification schemes, while it is ~93% in case of no purification. The data point at 100% CO<sub>2</sub> in the inlet provides an indication of the theoretical minimum power requirement for the CPU, since it represents the power consumption for compression of a pure CO<sub>2</sub> stream from the feed pressure to the product pressure.

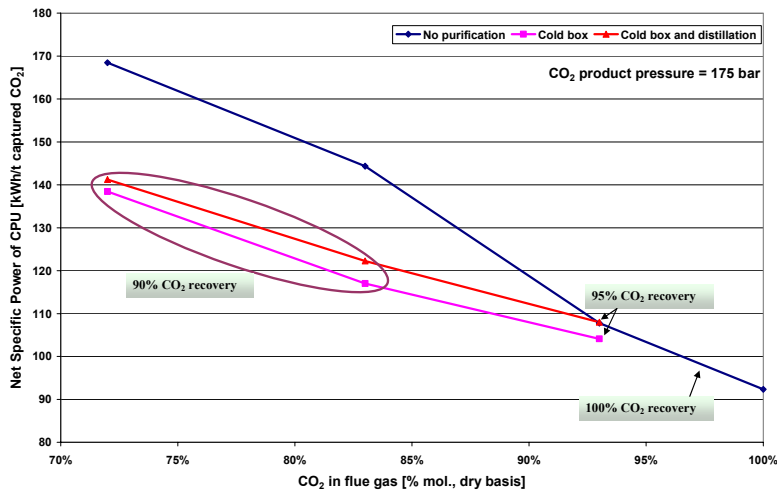


Figure 4: Specific energy consumption of different CO<sub>2</sub> CPU schemes (no integration) as a function of CO<sub>2</sub> purity in the inlet flue gas.

### 3.3. Influence of product pressure

Since the overall power requirement of the CPU is dependent upon the product pressure, it is instructive to examine whether the relative power requirements of the 3 schemes shown in Figure 4 are changed at different product pressures. Figures 5(a-b) show results of simulations of the 3 CPU schemes of Figure 4 at different product pressures for 2 inlet CO<sub>2</sub> compositions. From Figure 5a, for 83% inlet CO<sub>2</sub>, flue gas purification requires significantly less specific energy than simply compressing the entire flue gas to the product pressure, for the entire range of product pressures studied. In contrast, if the flue gas is more concentrated in CO<sub>2</sub>, the compression only case is more comparable to the purification cases. For example, at 93% inlet CO<sub>2</sub> (*cf.* Figure 5b), the ‘no

purification' process requires lower specific energy than the cold box with distillation process, up to a product pressure of 185 bar. The cold box only process though can provide a higher CO<sub>2</sub> purity in the product and lower O<sub>2</sub> content while consuming lower specific energy than the 'no purification' process. From figure 5b, the distillation process provides a nearly pure CO<sub>2</sub> stream with only about 1ppm O<sub>2</sub>. This level of product purity cannot be achieved by the other 2 schemes.

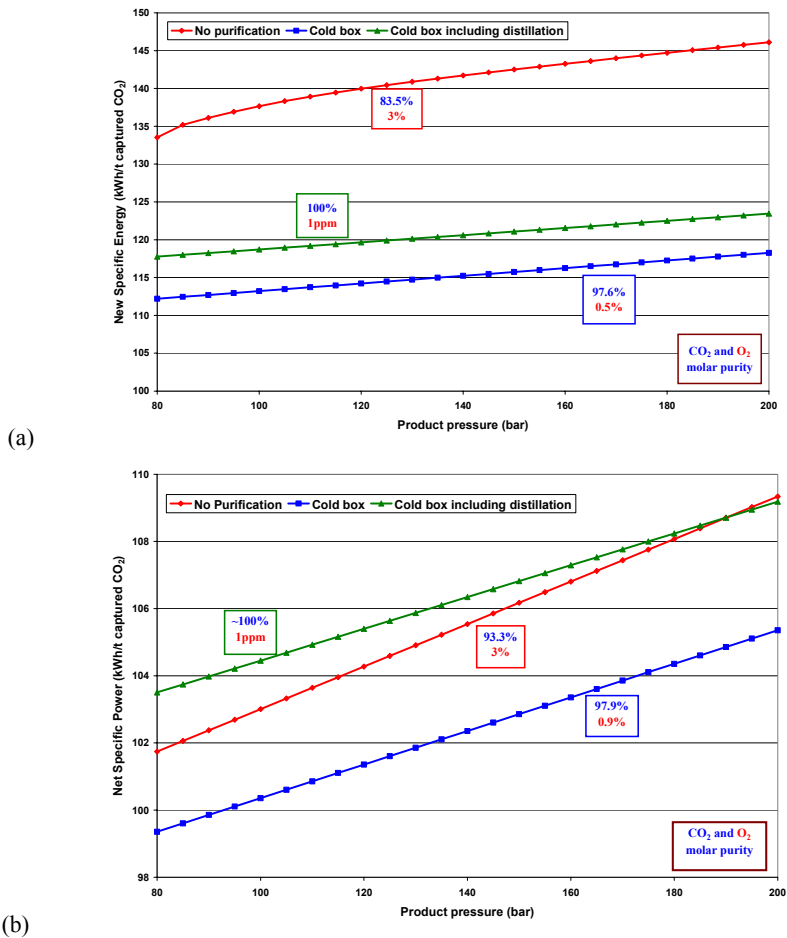


Figure 5: Specific energy consumption as a function of product pressure for (a) 83%, and (b) 93% inlet CO<sub>2</sub> (dry basis) and no integration.

### 3.4. Heat Integration

As explained in section 2.5, heat integration aims to improve overall process efficiency by transferring waste heat from various processes to the steam cycle. Considerable heat is generated by the compressors in the CO<sub>2</sub> CPU. In the schemes above, this heat energy is removed by cooling water and is thus eliminated from the process as waste. If

this energy can be used in other parts of the power plants for heating process streams, then the overall energy requirement for the plant can be reduced. Also, the requirement for cooling water is lessened.

Figure 6 shows the results of simulations of the 3 cases, as a function of inlet CO<sub>2</sub> content, wherein the heat from both wet and dry compressors is used to heat boiler feed water (full heat integration). To assess the energy savings from heat integration without having to calculate the energy consumption for the entire power plant, the energy transferred to the boiler feed water in each case has been taken as a credit in calculating the net specific energy of the CO<sub>2</sub> CPU process.

From Figures 4 and 6, heat integration can significantly reduce specific energy consumption. For example, at 72% inlet CO<sub>2</sub>, the specific energy required by the ‘no purification’ process is reduced from ~170 kWh/t to ~140 kWh/t by using heat from the wet and dry compressors for heating boiler feed water (full integration). Similarly, the energy requirement of the cold box process is reduced from ~140 kWh/t to ~120 kWh/t by incorporating heat integration into the process. Again, as for the ASU, the improvements shown here are specific to the conditions of the current study and may change if the design conditions are changed.

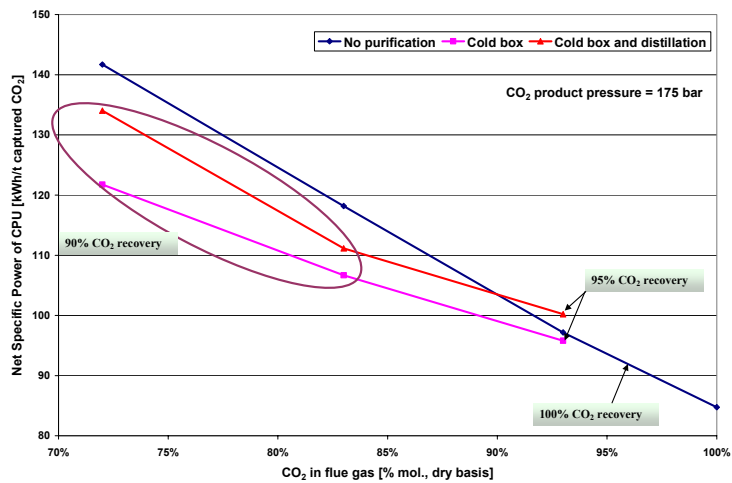


Figure 6: Specific energy consumption of different CO<sub>2</sub> CPU schemes with full heat integration as a function of CO<sub>2</sub> purity in the inlet flue gas.

#### 4. Conclusions

Air Liquide has made several technical advances to increase the attractiveness of the oxy-combustion process for clean electricity production from coal. By tailoring the cryogenic ASU scheme for oxy-combustion, a 20% reduction in specific energy has been achieved. Several CO<sub>2</sub> CPU designs have been developed to cover the requirements to treat different flue gas compositions and produce CO<sub>2</sub> product meeting different specifications over a range of pressures. Heat integration of the CPU design with the steam cycle shows considerable promise in reducing the overall operating cost of the oxy-combustion process.

Air Liquide has worked extensively with Babcock and Wilcox to optimize the overall oxy-combustion process. The results of this study are presented in another paper at this conference. [4]

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