Single-column cryogenic air separation: enabling efficient oxygen production with rapid startup and low capital costs—application to low-carbon fossil-fuel plants

Mao Cheng, Piyush Verma, Zhiwei Yang and Richard L. Axelbaum*

Mailing Address:

Department of Energy, Environmental and Chemical Engineering, Consortium for Clean Coal Utilization, Washington University in St. Louis, One Brookings Drive, St. Louis, Missouri 63130, USA

* Corresponding author: Dr. Richard L. Axelbaum; axelbaum@wustl.edu

Abstract

The rapid integration of intermittent renewable sources into the electricity grid is driving the need for more flexible, low-carbon fossil-fuel plants with lower capital costs. This then drives the need to improve the cryogenic air separation unit (ASU). To address this changing landscape, we explore a Praxair single-column ASU (PSC-ASU) design with the goal of reducing costs and improving flexibility, compared to a conventional double-column ASU. The PSC-ASU incorporates partial air condensation and air pre-separation in the bottom reboiler with a phase separator as well as N_2-enriched vapor condensation in the upper reboiler to decrease energy consumption, as compared to Linde's single-column ASU. All three of the above-mentioned ASU designs are simulated in Aspen Plus and analyzed. An economic analysis is applied to evaluate the relative cost savings of the PSC-ASU compared to the double-column ASU. Results suggest that the specific energy consumption of the PSC-ASU is significantly lower than that of Linde's single-column ASU due to a drastically improved oxygen recovery rate. Although this improved oxygen recovery rate is still lower than that of the double-column ASU, the required pressure ratio of the main air compressor is 21% lower than that of the double-column ASU. As a result, the specific energy consumption of the PSC-ASU is only 1.9% greater than that of the double-column ASU for producing 95.1 mol% O_2. However, the PSC-ASU reduces the hourly capital cost by 19% due to the elimination of a high-pressure column. This would effectively decrease the total hourly cost of the ASU, and thus the total hourly cost of low-carbon, fossil-fuel power plants that require oxygen.

Keywords: ASU, single-column, air separation, energy consumption, capital cost, rapid startup

1. Introduction

The push to incorporate greater amounts of intermittent sources, namely wind and solar, into the power grid has led to a drastic change in the desirable characteristics of modern power plants [1, 2]. These plants are no longer operating as base load plants, but rather at variable load, supplementing wind and solar when they are not available. Thus, there has been a significant emphasis on plants with lower capital costs and higher flexibility, along with low CO_2 emission [2-4]. Many low-carbon electricity generation technologies, such as the Allam cycle [5, 6], oxyfuel combustion [7, 8], and integrated gasification combined cycle (IGCC), require O_2-enriched gases that are typically supplied by cryogenic air separation units (ASUs) [9, 10]. However, the commonly used double-column ASU has a very high capital cost. For example, for a conventional oxyfuel power plant, the ASU accounts for about 14% of the total plant capital costs [9]. Moreover, typical ASU configurations are highly integrated, leading to low flexibility. The ramp rates and minimal load of the above-mentioned low-carbon power generation systems are often constrained by the flexibility of the ASU [4, 11]. Given the future market needs, it is of great interest to explore alternative ASU designs with lower capital cost and higher flexibility. Cryogenic air separation is also an energy-intensive process. In a low-carbon power plant, the ASU sometimes represents the single largest energy penalty for the power plant (10–15% of gross power output) [9, 10, 12].

Historically, research on O_2 production from an ASU has focused extensively on the efficiency of the process [13]. In 1902, Linde designed the first cryogenic distillation column to separate O_2 from air. The single-column design recovered only 67% of the O_2 from the compressed air flow and therefore was inefficient [14]. Since the liquid reflux of the single column is liquid air, the distillate vapor from the column top contains about 7% O_2, which results in a low O_2 recovery rate for the single column [14, 15]. In 1910, Linde modified the process, incorporating air pre-separation in another high-pressure column. The new double-column ASU simultaneously produced N_2 and O_2 products and achieved very high O_2 recovery rate, which revolutionized the industry and is still the workhorse of modern air cryogenic oxygen plants [14-16]. Figure 1 shows the history of energy consumption for O_2 production in cryogenic ASUs. With continuous improvement in ASU equipment and processes, the power consumption steadily decreased to nearly 250 kW·h/t (0.36 kW·h/Nm^3), a typical value for the double-column ASUs being used today [13, 17]. The improved designs with dual reboiler double-column, triple column, or multi-column have been proposed to decrease the necessary pressure or flow rate of compressed air for a more efficient low-carbon fossil-fuel plant [18, 19]. By making better use of the heat of compression and by improving the heat integration of the distillation columns, the efficiency of a double-column ASU that is part of an IGCC can be increased significantly [20]. Although these developments have increased the efficiency of the ASU, they have resulted in more complicated processes with higher capital costs and lower flexibility [10].

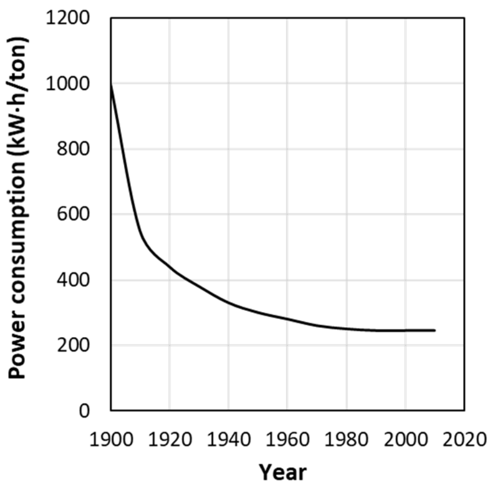


Figure 1. Historical power consumption for O_2 production in cryogenic ASUs [13, 17].

Although the single-column ASU design is less efficient, it holds several advantages over the double-column design. As the column is the most expensive component in an ASU [21, 22], a single-column ASU with just one low-pressure column can have a much lower capital cost than a double-column ASU [23]. Additionally, a single-column ASU enables a faster startup, which is critical to the flexibility of ASU in low-carbon, fossil-fuel power plants [2, 3]. ASU startup requires air liquefaction to obtain enough liquid holdup, and thermodynamic equilibrium in the column, which is a time-consuming procedure [11, 24]. A fundamental characteristic of a single-column ASU is that it has a significantly smaller column metal mass and liquid hold up than that of a double-column ASU. Therefore, a single-column ASU has an inherent advantage in startup speed. According to a dynamic process analysis for ASU startup, the necessary startup time can be reduced by about 20% without a high-pressure column [24]. Moreover, since air, instead of N_2-enriched vapor, is employed to boil the O_2-enriched liquid in the reboiler of Linde's single column, the required air pressure from the main air compressor (MAC) is lower than that of a double-column ASU [14], which reduces energy demand. For a gas supplier, a major advantage of the double-column ASU is the improved energy efficiency of the process, and the simultaneous production of high purity N_2, which can be used in different industries, such as the fertilizer industry. However, for many low-carbon fossil-fuel power generation systems, the desired product is just a modest purity O_2 (95-97%) [10]. This represents a significant opportunity for the application of a single-column ASU.

Taniguchi et al. [17] analyzed a single-column ASU with a N_2 cryogenic compressor, which does not compress the air feed but compresses a recycle N_2 vapor to produce liquid for reflux. Based on the exergy analysis, this nitrogen-refluxed single-column ASU can reduce power demand by as much as 30% compared to the conventional double-column ASU. However, the use of the cryogenic compressor for the ASU was not technically or economically feasible for many decades [25]. Nonetheless, many inventions followed from this concept, and in 2011 Kansha et al. proposed the recuperative vapor recompression (RVRC) distillation [26]. RVRC distillation avoids a cryogenic N_2 compressor by replacing it with a standard compressor operated at above ambient temperature. Several promising cryogenic separation processes based on RVRC have been discussed by Fu and Gundersen [27]. The reported simulation results showed a single-column ASU with RVRC can decrease energy consumption by about 7.9% with an enhanced O_2 recovery rate (98.4%) for producing 95 mol% O_2. As Fu and Gundersen have explained, low pressure improves the relative volatility between nitrogen and oxygen, and the air feed does not need to be compressed to high pressure. Since the flowrate of nitrogen used in vapor recompression is smaller than that of air, less compression work is needed [28]. Therefore, the nitrogen-refluxed single-column ASUs are considered to be more efficient than the conventional double-column ASU [25-27, 29, 30].

In 1998, Praxair proposed an alternative to the nitrogen-refluxed single-column ASUs - a pre-separated air refluxed single-column ASU that applies multiple reboilers with phase separators to produce N_2-enriched liquid reflux, which increases O_2 recovery rate [23]. This technology has not been evaluated in the open literature, and the goal of this study is to do so in the context of the present needs of ASUs for low-carbon power applications. The configuration of the dual reboiler in a double-column ASU effectively decreases the energy consumption by reducing the air compression pressure [27, 31]. Compared with the nitrogen-refluxed single-column ASUs, the Praxair single-column ASU (PSC-ASU) is more like the conventional double-column ASU in that both are driven by air compression and have very similar heat integrations without recycling N_2 enthalpy flow. Most commercial ASUs are of this type and, thus, the risks associated with development of the PSC-ASU might be considered lower compared with that of the nitrogen-refluxed single column ASU.

This work studies the performance of the PSC-ASU with the specific goal of producing O_2 at 15 bar for a flexible carbon capture and storage (CCS) plant [32]. The CCS plant considered is the staged, pressurized oxyfuel combustion (SPOC) process, which shows promise for carbon capture with high operational flexibility and low energy penalty. The PSC-ASU design is then compared with Linde's conventional single-column design and a state-of-the-art double-column design. All three of the above-mentioned ASU designs are simulated in Aspen Plus. The ASU critical parameters are identified theoretically and calculated by the process models. The specific energy consumptions and O_2 recovery rates in producing O_2 at various purities are determined for the three processes. An approximate cost estimation is carried to understand the economic improvements of the PSC-ASU compared with the conventional double-column ASU. The PSC-ASU is introduced and analyzed in Section 2. In Section 3, the simulation models for the three ASUs mentioned above are established, evaluated and validated. In Section 4, the simulation results and cost analysis are presented and discussed.

2. The PSC-ASU with partial air condensation and pre-separation for producing O_2

An ASU primarily involves air compression, air cooling and purification, cold production and internal compression, and cryogenic rectification of air [14, 16]. The single-column ASU has only one low-pressure column and produces O_2, in contradistinction to Linde's state-of-the-art commercial ASU, which has four columns and simultaneously produces several pure products (N_2, O_2, Ar) [16]. Figure 2 shows the basic cryogenic air separation process of the PSC-ASU. The single column is based on Praxair's patented design [23]. The basic process, excluding the column, is based on the state-of-the-art double-column ASU [14].

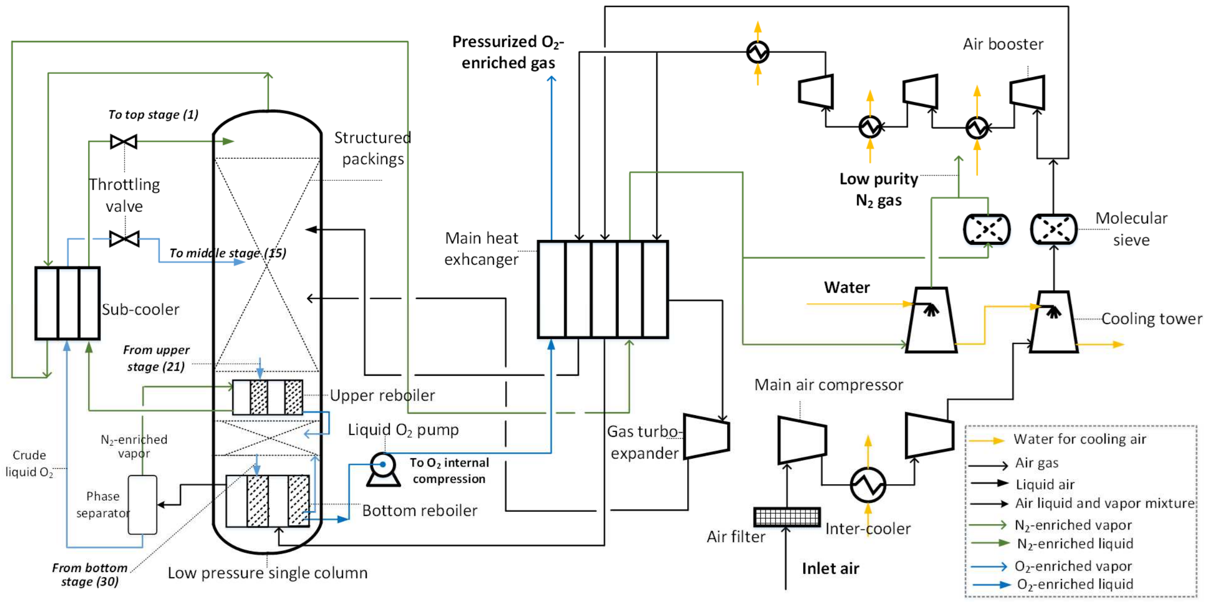


Figure 2. The PSC-ASU process for producing pressurized O_2.

In this ASU, the vapor-liquid counter-current flows in the columns are the critical flow structures for cryogenic air separation, determining the O_2 product purity and O_2 recovery rate. Heat integration in the ASU, which occurs primarily in the main heat exchanger, reboiler and sub-cooler, establishes the counter-current flow structure in the column. The gas turbo-expander, which provides the ASU process with the necessary cooling, is critical to maintain the low temperature and cryogenic liquid streams in the ASU. The air compression with air cooling drives the refrigeration cycle in the ASU. As Figure 2 shows, the reboiler/condenser (simply noted as a reboiler) of the low-pressure column is divided into two parts, an upper and bottom reboiler, or dual reboiler. The bottom reboiler is designed for heat transfer and partial air condensation. As air flows into the hot-side of the bottom reboiler, a fraction condenses on the cold wall and transfers heat to the O_2-enriched liquid flowing on the cold-side because of a small temperature difference. The heat transfer also leads to the partial evaporation of the liquid on the cold side. Around 65 – 85% of the O_2-enriched liquid from the bottom stage of the column (30^{th}

stage) evaporates on the cold-side of the bottom reboiler, which provides the single column with vapor reflux. The remaining O_2-enriched liquid is pumped into the main heat exchanger for O_2 internal compression [16]. The air liquid and vapor mixtures are separated in a phase separator, resulting in a crude liquid O_2 at the bottom and N_2-enriched vapor at the top of the phase separator.

As Figure 2 shows, crude liquid O_2 flows into the column's middle stage (15^{th} stage) and is distilled in the single column. N_2-enriched vapor flows into the upper reboiler and completely condenses to N_2-enriched liquid by transferring heat to the column liquid from a stage (note as an upper stage, 18^{th}-21^{th} stage) above the bottom stage. The heat vaporizes a fraction of this column liquid to the vapor reflux of the column. Since the liquid temperature of the upper stage is significantly lower than that of the bottom stage, the upper reboiler can operate with lower air pressure compared with a single bottom reboiler configuration. Moreover, as shown in Figure 3, partial air condensation (i.e., the vapor mole fraction on the hot-side outlet of the bottom reboiler is 0.47) requires lower gas pressure than other approaches. Compared with full condensation of high-purity N_2 in the reboiler of the double column, the necessary pressure for partial air condensation can be reduced about 0.8 bar. Through partial air condensation and pre-separation in the bottom reboiler with a phase separator, and N_2-enriched vapor condensation in the upper reboiler, the single column is able to 1) provide N_2-enriched liquid reflux for increasing O_2 recovery rate and 2) decrease the air pressure from the MAC, which can effectively reduce the energy consumption of ASU.

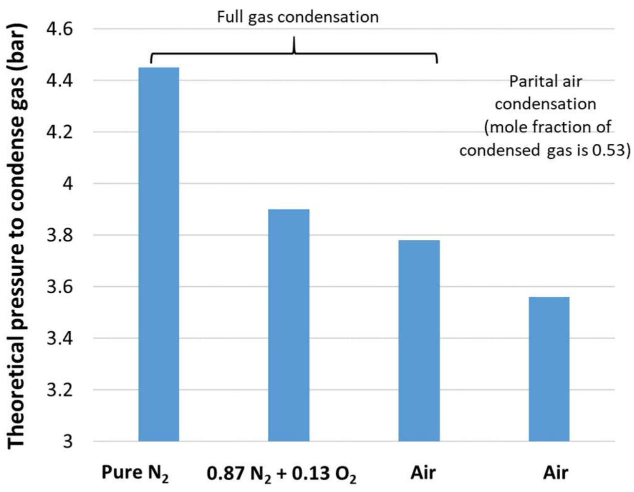
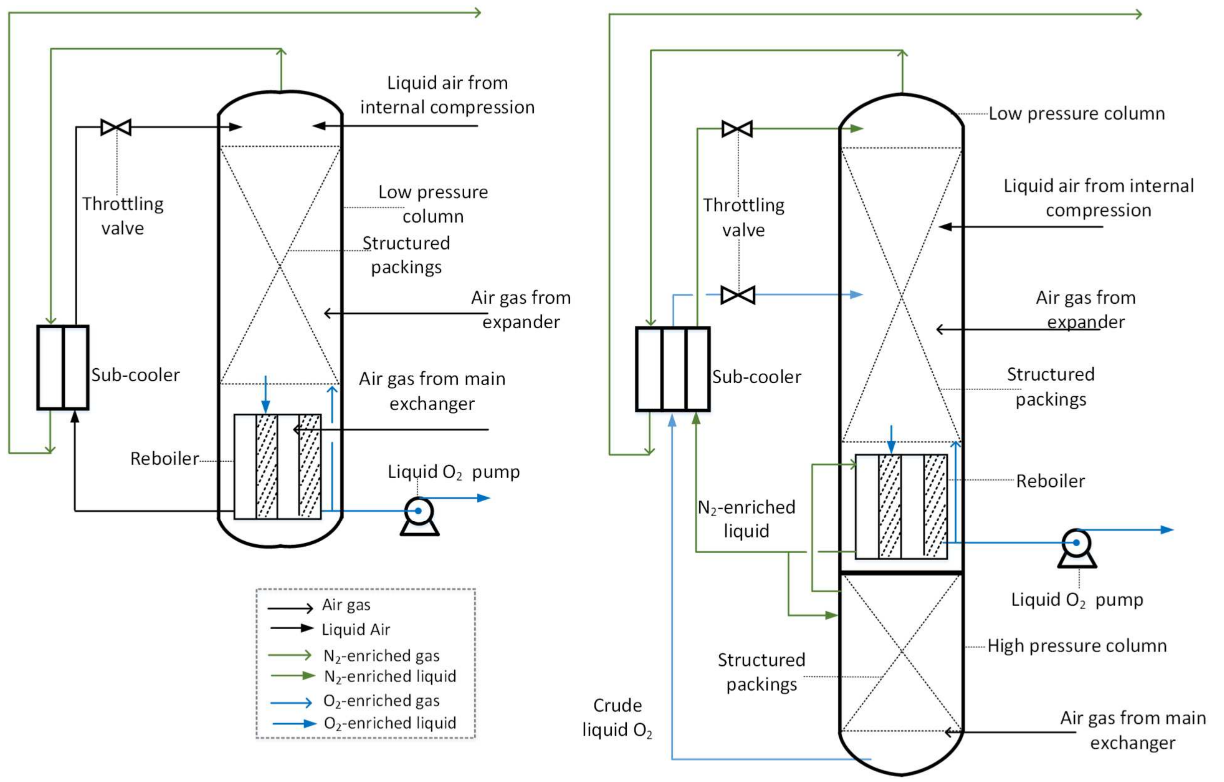


Figure 3. Theoretical pressures to fully condense gases of different compositions and partially condense air at a temperature of -180.5 ^{o}C.

In addition to energy consumption, capital cost is also a critical consideration in ASU design for low-carbon fossil-fuel plants. Since the cost of a heat exchanger is much lower than that of a high-pressure column [21], the cost of the additional upper reboiler in the PSC should be marginal. The lower air pressure from the MAC in the PSC design would increase the size of several downstream units (main heat exchanger, reboiler, etc.), which adds a modest capital cost. However, the total capital cost of a single-column ASU is expected to be much smaller than that of a double-column ASU. For comparison, Linde's single column and conventional double column are presented in Figure 4, with the other parts of the process remaining effectively the same as those presented in Figure 2. As these two columns are thoroughly discussed in the literature [14, 15], they will not be described here, other than to note that in Linde's single column (Figure 4a) there is nearly zero pre-separation; in the double column (Figure 4b) the air has a high pre-separation; and in the PSC (Figure 2) there is moderate pre-separation. The level of pre-separation significantly influences the O_2 recovery rate (see Section 4).



(a) **(b)**

Figure 4. (a) Linde's single column and (b) the conventional double column ASU [14, 15].

3. Process simulation and evaluation

3.1 Process model

Aspen Plus V10 is employed to simulate the three ASU designs. The Peng-Robinson property method is used for thermodynamics calculations. A two-stage centrifugal compressor with an intercooler is used as the MAC to compress air to 3.5~5.5 bar for driving the process of cryogenic air separation. After the first stage compression, the air is assumed to be cooled to 30°C in the intercooler using boiler feedwater from a fossil-fuel plant. After the second stage compression, the hot air is assumed to be cooled to 16ºC in a cooling tower by the cold water that is initially cooled by the cold N_2-enriched gas in another cooling tower. The impurities of air (CO_2, H_2O, hydrocarbons, etc.) are not considered in this work. The total pressure drop of the cooling tower and molecular sieve is assumed to be about 20 kPa.

About 20-35% of the low-pressure air (3.5-5.5 bar) from the cooling tower is further compressed to about 35 bar by the air booster compressor (ABC), which is a three-stage centrifugal compressor with intercoolers. For O_2 internal compression, the pressure of the ABC is chosen to easily condense air through the heat exchanger while vaporizing the liquid O_2 stream. Generally the pressure of the condensing air stream is much greater than the liquid O_2 stream [14, 16]. During internal O_2 compression, the main fraction (70-80%) of this high-pressure (35 bar) air is condensed to vaporize pumped O_2-enriched liquid in the main heat exchanger to produce O_2-enriched gas at 15 bar. Additionally, a fraction of this high-pressure air (20-30%) is cooled down in the main heat exchanger by the cold flows from the column and then expanded in a centrifugal turbo-expander, which reduces the temperature of this air stream to near the dew point. Isentropic efficiencies of the centrifugal compressors and expanders are assumed to be 80% and 85%, respectively.

The balance of the low-pressure air from the cooling tower flows into the main heat exchanger in order to recover the cold from the gas product streams. A high-efficiency brazed plate-fin main heat exchanger is simulated by using the multi-stream heat-exchanger model (*MHeatX*) in Aspen Plus, which is capable of calculating heat transfer between the three hot streams and two cold streams shown in Figure 2. The heat transfer in the sub-coolers is calculated by using the countercurrent two-stream heat-exchanger model (*HeatX*) in Aspen Plus. The design parameters used in the models are provided in Tables A1 and A2 in the supplementary file.

All the columns in the three ASU designs are modeled as packed bed columns, which have much higher flexibility than tray columns [16]. The details of the model are provided in Table A3 in the supplementary file. The packed columns are simulated by using the Aspen *RadFrac* model, which is a rigorous model for simulating all types of multistage vapor-liquid fractionation operations. The low-pressure column in each of the ASU designs contains 30 theoretical stages. The number of theoretical stages was chosen to obtain the best O_2 recovery rate, as Figure A1 presents. The high-pressure column in the double-column design contains 20 theoretical stages. Identical design parameters are used for the low-pressure columns in each of the processes.

The reboilers in the ASUs are simulated with the *HeatX*, which is capable of calculating heat transfer between two streams with phase change. Figure 5 shows the basic connections between the models of the reboilers, columns and phase separators in the three ASU designs. A separator model (Aspen *Flash2*) that can simulate vapor-liquid phase equilibrium is used to represent the air (vapor and liquid mixture) pre-separation in the phase separator for the PSC, as shown in Figure 5c. The minimum temperature differences at the cold end of the reboilers are assumed to be 1°C [33], which is critical to determine the air pressure from the MAC. The vapor mole fraction of the hot exit stream leaving the bottom reboiler is specified to be 0.47, which is optimized for highest O_2 recovery rate, as shown in Figure A2. Pressure drop in each heat exchanger in the ASUs is specified to be about 5-10 kPa, mainly determined by material flow rate and heat duty. The air pressure from the MAC in each process is chosen to obtain the lowest energy consumption and stable vapor liquefaction in the reboiler. The detailed stream parameters and process specifications in the steady-state PSC-ASU are shown in Figure A3. Table A4 presents the balance of material and energy in this process. The validation of the process models can be found in Appendix B.

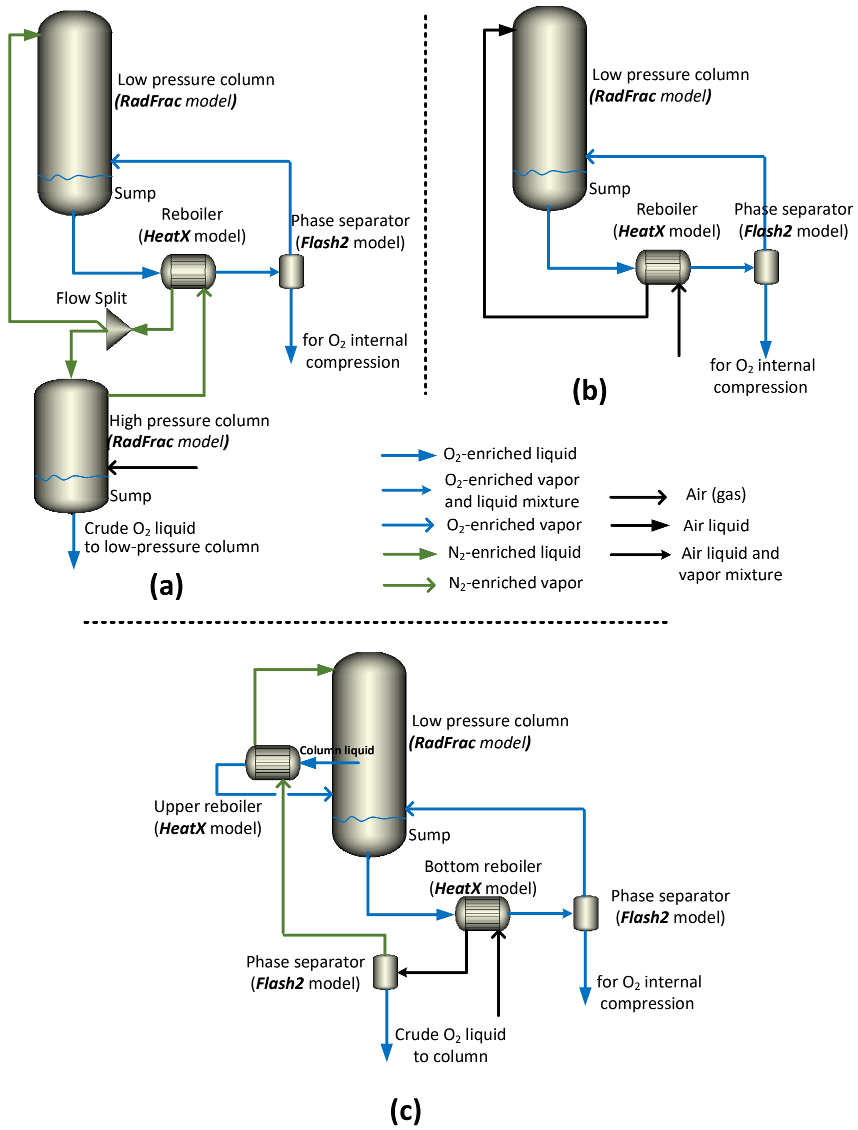


Figure 5. Aspen models of the reboilers with their connected unit models in (a) the conventional double column, (b) Linde's single column, and (c) Praxair single column.

3.2 Process evaluation

The specific energy consumption for producing a unit amount of O_2 (E_O) is a critical criterion in evaluating ASU performance. It is calculated by dividing the total energy consumed by the ASU (E) with the normal volumetric flow rate of the pure O_2 (V_O):

$$E_{\mathrm{O}} = \frac{E}{V_{\mathrm{O}}} \tag{1}$$

The O_2 recovery rate (η) of the ASU is obtained by Eq. (2):

$$\eta = \frac{V_{\mathrm{O}}}{V_{\mathrm{F}} \cdot c_{\mathrm{O}}} \tag{2}$$

where V_{F} is the air feed rate (normal volumetric flow rate), and c_{O} is obtained by dividing the total O_2 flows in the products with V_{F}. Similarly, we define the internal compression ratio (φ) as the ratio of the normal volumetric flow rate of the high-pressure air from the ABC (V_{B}) to $\mathrm{V_O}$:

$$\varphi = \frac{V_{\mathrm{B}}}{V_{\mathrm{O}}} \tag{3}$$

The necessary flow rates of high-pressure air (35 bar) and low-pressure air (3.5-5.5 bar) for producing a unit amount of pressurized O_2 gas (15 bar) are evaluated by φ and the reciprocal of η, respectively. Most of the energy in ASU is consumed by the air compression in the MAC (E_{C}) and ABC (E_{B}):

$$E = E_{\mathrm{C}} + E_{\mathrm{B}} \tag{4}$$

Assuming the process in each stage of the MAC and ABC is compression with a constant isentropic efficiency (η_{e}), E_{C} and E_{B} are calculated by Eqs. (5) and (6) that are from Aspen help file:

$$E_{\mathrm{C}} = N_{\mathrm{C}} \cdot n_{\mathrm{F}} C_{\mathrm{P}} T_{\mathrm{I}} \cdot \left[\left(\frac{p_{\mathrm{C}}}{p_{\mathrm{I}}}\right)^{\frac{k-1}{N_{\mathrm{C}} \cdot k}} - 1 \right] \cdot \frac{1}{\eta_{\mathrm{e}}} \tag{5}$$

$$E_{\mathrm{B}} = N_{\mathrm{B}} \cdot n_{\mathrm{B}} C_{\mathrm{P}} T_{\mathrm{I}} \cdot \left[\left(\frac{p_{\mathrm{B}}}{p_{\mathrm{C}}}\right)^{\frac{k-1}{N_{\mathrm{B}} \cdot k}} - 1 \right] \cdot \frac{1}{\eta_{\mathrm{e}}} \tag{6}$$

where N_C and N_B are, respectively, the number of compression stages in the MAC and ABC, n_F and n_B are, respectively, the mole flow rates of the air feed and high-pressure air, T_I is the air temperature at the inlets of the MAC and ABC, p_I, p_C and p_B are, respectively, the atmospheric air pressure, the outlet pressures of the MAC and ABC, and k is heat capacity ratio (C_P/C_V). Using Eqs. (1)-(6) with the ideal gas equation, E_O can be obtained from:

$$E_O = \left[N_C \cdot \frac{1}{c_O \cdot \boldsymbol{\eta}} \cdot \left((\frac{p_C}{p_I})^{\frac{k-1}{N_C \cdot k}} - 1\right) + N_B \cdot \boldsymbol{\varphi} \cdot \left((\frac{p_B}{p_C})^{\frac{k-1}{N_B \cdot k}} - 1\right)\right] \cdot p_I \cdot \frac{k}{k-1} \cdot \frac{1}{\eta_e} \quad (7)$$

Here N_C, N_B, η_e, p_I, p_B, c_O, k are constant parameters, which are not changed with the different column designs and O_2 product purities, while η, φ and p_C are varied parameters that are determined by the column designs and O_2 product purities (see Section 4). The simulation models in Aspen Plus determine all the three varied parameters and calculate E_O based on Eq. (7).

To simplify the cost analysis of the single-column ASUs (Linde's and Praxair), the conventional double-column ASU is used as a reference. E_O represents the hourly operational cost (energy cost) of the ASU [22, 30]. Therefore, the increase or decrease in the hourly operation cost of the ASU ($\Delta OPEX\%$) compared to that of the double-column ASU, is almost equal to the relative change in energy consumption, or:

$$\Delta OPEX\% \approx \frac{E_{O,S} - E_{O,D}}{E_{O,D}} \quad (8)$$

where $E_{O,S}$ and $E_{O,D}$ are the specific energy consumptions of the single-column ASUs and the conventional double-column ASU, respectively.

The capital costs of the low-pressure and high-pressure columns account for about 60% of the total capital cost of ASU [22]. Elimination of the high-pressure column reduces about 40% of the capital cost of columns [34]. Therefore, the hourly capital cost of a single-column ASU reduces to $1-60\%\times 40\%$. However, a decrease in O_2 recovery rate of a single-column ASU would result in a larger ASU scale, which can be represented by:

$$\frac{SC_S}{SC}=\frac{\eta_D}{\eta_S} \tag{9}$$

where η_D and η_S are the O_2 recovery rates of the double-column and single-column ASUs, respectively. SC_S and SC are the scales of the single-column ASU at O_2 recovery rates of η_S and η_D, respectively. The hourly capital cost of the ASU increases by the scale of the ASU with a scaling exponent of 0.5 [21]. Therefore, the relative difference in the hourly capital cost of the single-column ASU ($\Delta CPEX\%$) compared with the double-column ASU can be estimated by:

$$\Delta CPEX\% \approx (1-60\%\times 40\%)\cdot\left(\frac{SC_S}{SC}\right)^{0.5}-1 \tag{10}$$

The relatively difference in the total hourly cost of the single-column ASUs ($\Delta TEX\%$) compared to that of the double-column ASU can be roughly estimated with $\Delta CPEX\%$ and $\Delta OPEX\%$ by:

$$\Delta TEX\% \approx \theta\cdot\Delta CPEX\% + (1-\theta)\cdot\Delta OPEX\% \tag{11}$$

where θ is the fraction of the hourly capital cost in the total hourly cost of the conventional double-column ASU, while $1-\theta$ is the fraction of the hourly operation cost in the total hourly cost of the conventional double-column ASU. θ is usually about 20% for a typical conventional double-column ASU [22]. However, for the ASU of a low-carbon power plant, θ could be higher than 20%, because of the lower operation hours and average load. Using Eq. (8)-(11), $\Delta TEX\%$ can be obtained from:

$$\Delta TEX\% \approx \theta\cdot\left[0.76\left(\frac{\eta_D}{\eta_S}\right)^{0.5}-1\right]+(1-\theta)\cdot\frac{E_{O,S}-E_{O,D}}{E_{O,D}} \tag{12}$$

4. Results and discussion

4.1 Specific energy consumption

From Eq. (7), the O_2 recovery rate, the air pressure from the MAC and the internal compression ratio are three critical parameters determining the specific energy consumption for O_2 production in the different ASU designs. Figure 6 shows the impact of the O_2 product purity on O_2 recovery rates for all three ASU designs. The PSC-ASU has a significantly higher O_2 recovery rate than Linde's single-column ASU, but lower than the double column. By partially condensing and pre-separating air in the bottom reboiler and phase separator, and condensing N_2-enriched vapor in the upper boiler, the PSC-ASU receives N_2-enriched liquid reflux, which effectively increases O_2 recovery rate. Table 1 lists the purities in some critical components for all three ASU designs. With about 95 mol% O_2 product purity, the N_2-enriched liquid reflux (86.7 mol% N_2) decreases the O_2 concentration in the N_2-enriched vapor to 4.6 mol%, as opposed to 7.4 mol% in Linde's single-column design. A lower O_2 concentration in the N_2-enriched vapor means a higher O_2 recovery rate. Table 1 also presents the O_2 purities of the crude liquid O_2 in the ASUs. These purities, along with the O_2 recovery rates, demonstrate the value of the moderate air pre-separation in PSC.

As shown in Figure 6, for all ASU designs, the change in O_2 recovery rate is marginal when the O_2 product purity is varied from 90 to 96 mol%. However, as O_2 product purity increases above 96 mol%, the O_2 recovery rate drops drastically for all designs. This sharp change is caused by the increase in vapor reflux from the reboiler, which originates from the need to separate argon from O_2. Argon and O_2 have similar boiling points, therefore, separating argon from O_2 will increase the specific energy consumption and/or capital costs.

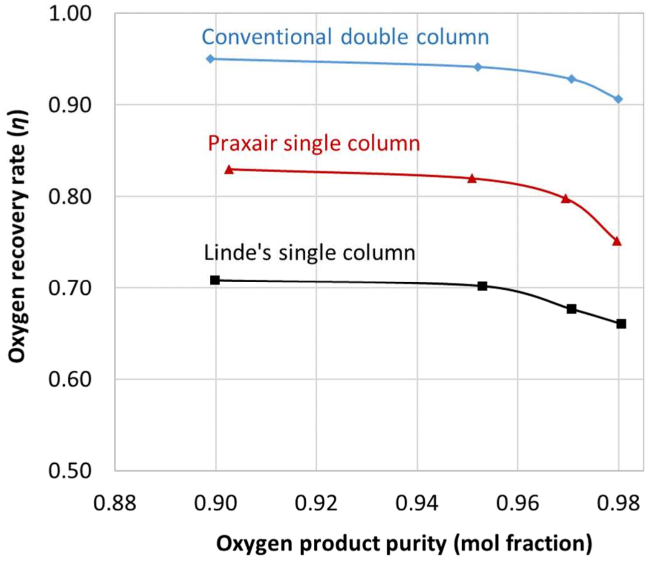


Figure 6. Impact of O_2 product purity on the O_2 recovery rates for the three different ASU designs (conventional double column, Linde's single column, and Praxair single column)

Table 1. Purities of critical components in three different ASU designs with 95 mol% O_2 product purity.

	The double column	Linde's single column	Praxair single column
N_2 purity in N_2-enriched vapor	0.986	0.921	0.951
O_2 purity in N_2-enriched vapor	**0.012**	**0.074**	**0.046**
Ar purity in N_2-enriched vapor	0.002	0.005	0.004
N_2 purity in O_2-enriched vapor	0.011	0.012	0.014
O_2 purity in O_2-enriched vapor	0.952	0.953	0.951
Ar purity in O_2-enriched vapor	0.038	0.035	0.035
N_2 purity of N_2-enriched liquid reflux	**0.999**	**0.781**	**0.868**
O_2 purity of crude liquid O_2	0.340	0.210	0.284

The O_2 recovery rate of the PSC-ASU is lower than that of the double-column ASU because of the reduced air pre-separation in the PSC-ASU. However, because of the partial air condensation in the bottom reboiler, the PSC-ASU operates with a significantly lower air pressure from the MAC (p_C) compared with the double-column ASU, as evident in Figure 7. The p_C can be lower than 4 bar in PSC-ASU if the O_2 product purity is lower than 95 mol%. All three ASU designs show a similar impact of O_2 product purity on p_C, as shown in Figure 7. Higher p_C with increased O_2 product purity is due to a higher liquid bubble point at the cold-side inlet of the bottom reboiler (T_{bu}). The impact of O_2 product purity on the T_{bu} of the PSC-ASU is also shown in Figure 7. T_{bu} increases from -184 to -180.5 °C when the O_2 product purity increases from 90 to 98 mol%. This temperature increase results in a necessary increase of 0.8 bar in p_C.

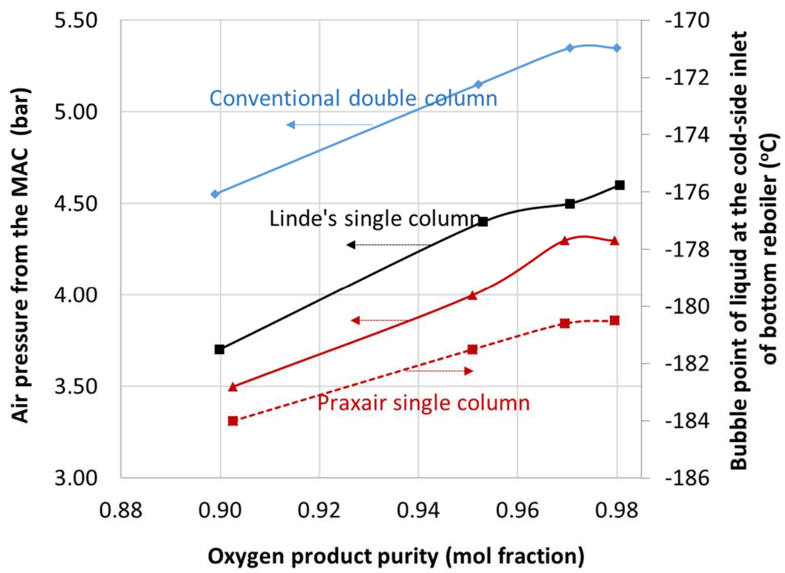


Figure 7. Impact of O_2 product purity on the air pressure from the MAC of three different ASU designs (the double column, Linde's single column, and Praxair single column) and the liquid temperature at the cold-side inlet of the bottom reboiler of the Praxair single column.

As Figure 6 and 7 reveal, the O_2 recovery rate and p_C depend not only on the O_2 product purity but also the column design. However, the differences of the internal compression ratios (φ) for the three ASU designs are much smaller, as shown in Figure 8, since the basic processes of O_2 internal compression and cold production that determine φ are the same, as presented in Section 2. All three ASUs necessarily need about 1.6-1.7 moles of high-pressure air for producing one mole of O_2 at a pressure of 15 bar when O_2 product purity is higher than 95 mol%. In Figure 8, φ increases with decreasing O_2 product purity (<97 mol%) because for each mole of O_2 compressed, a higher amount of N_2 and inert gases are compressed.

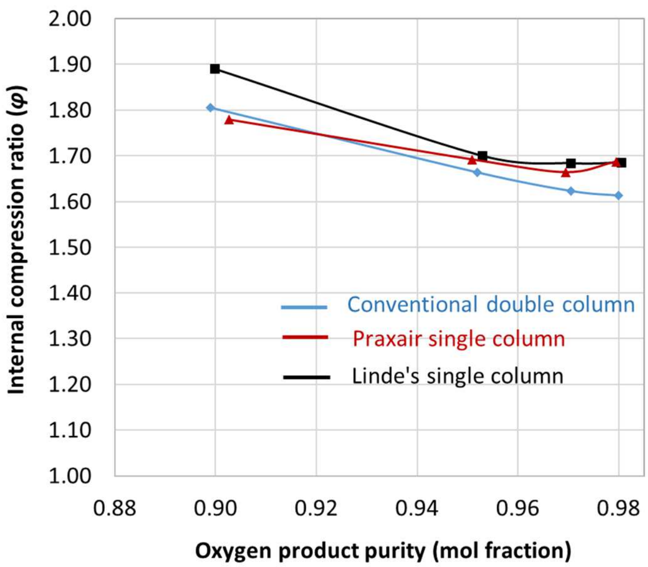


Figure 8. Impact of O_2 product purity on the internal compression ratio of three different ASU designs (the double column, Linde's single-column, and Praxair single column) when producing oxygen at a pressure of 15 bar.

The above results show that, when the O_2 product purity is lower than 97 mol%, the PSC-ASU has improved O_2 recovery rate by about 16% compared with Linde's single-column ASU; and p_C is about 21% less than that for the double-column ASU. Both changes are critical to the performance of this single-column ASU. Figure 9 shows a comparison of the specific energy consumptions for all three ASU designs. The PSC-ASU has significantly lower specific energy consumption (about 14.5%), compared with Linde's single-column ASU, when producing relatively low-purity O_2 (< 97 mol%). Compared with the double-column ASU, the PSC-ASU has comparable specific energy consumption when producing 90 mol% O_2 and only 1.9% higher specific energy consumption when producing 95 mol% O_2. For the PSC-ASU, around 70-80% of the energy is consumed in the MAC.

As shown in Figure 9, when the O_2 product purity is lower than 97%, the specific energy consumption of the PSC-ASU increases slowly with O_2 product purity because p_C increases. However, when the O_2 product purity is higher than 97%, the specific energy consumption for the PSC-ASU increases significantly due to the drastic decrease in O_2 recovery rate. Since 95 mol% O_2 can be used for most low-carbon, fossil-fuel power plants [10], the PSC-ASU would avoid this significant power loss. It should be noted that, the Allam cycle requires a minimum O_2 purity of 98 mol% [6], which would lead to over 7% more power requirement of the PSC-ASU, compared with the double-column ASU.

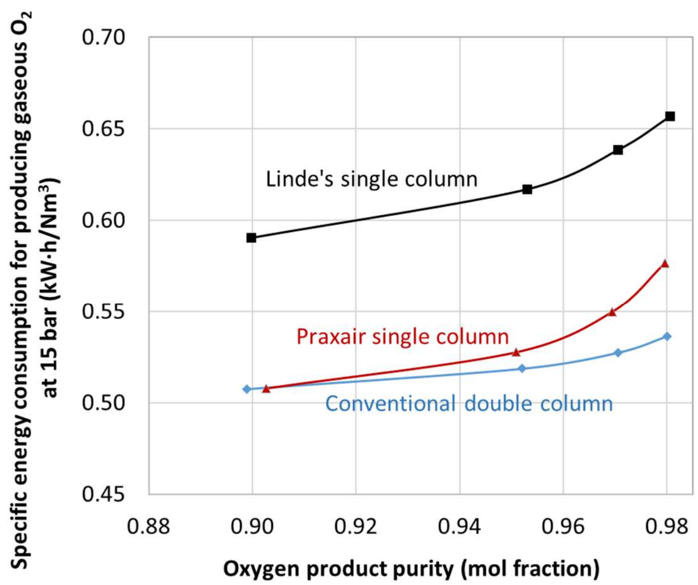


Figure 9. Impact of O_2 product purity on the specific energy consumptions of three different ASU designs (the double column, Linde's single column, and Praxair single column) when producing oxygen at a pressure of 15 bar.

4.2 Cost analysis

With known energy consumption and O_2 recovery rate, we can analyze the cost of the PSC-ASU from Eqs. (8-12), with the double-column ASU (95 mol% O_2) as reference. The PSC-ASU has a 1.9% increase in the hourly operation cost based on Eq. (8). However, based on Eq. (10), the PSC-ASU reduces about 19% of the hourly capital cost by eliminating a high-pressure column. As a result, with a θ of 20% the PSC-ASU decreases the total hourly cost about 2.2% compared with the double-column ASU. In contradistinction, the total hourly cost of Linde's single-column ASU has an increase of 13% because of the low O_2 recovery rate. Figure 10 presents the $\Delta TEX\%$ and indicates the decrease in total hourly cost of the PSC-ASU when producing 95 mol% O_2. When producing 98 mol% O_2 for the Allam cycle, $\Delta TEX\%$ is about 2.7% for the PSC-ASU with the typical θ (20%), which means a higher cost compared to the double-column ASU. Therefore, the PSC-ASU is more suitable for applications which require only a lower O_2 purity (90-95 mol%). It should be noted that a nitrogen-refluxed single-column ASU can potentially save up to 10% on the total hourly cost because of its lower energy consumption [27, 30] compared with the double-column ASU, making it more cost-effective than the PSC-ASU.

As shown in Figure 10, there is a more significant reduction in hourly total cost for the PSC-ASU when θ increases. A higher θ is possible when the ASU is part of a low-carbon, fossil-fuel power plant because the plant would not necessarily be operating at base load due to the penetration of intermitent power sources like wind and solar [2, 3, 35]. This would decrease the capacity factor of the plant [2], leading to a situation where capital-costs begin to dominant decision making. For example, in the north of China, where integration of solar and wind energy is high, many coal-fired power plants operate at partial load (<50%) for most of the operation time (<4000 hours per year), and operate at high load only occasionally, during peak demand. For the ASU in such a power plant, θ might be above 50%. Based on Figure 10, the PSC-ASU can save up to 8% and 4% of the total hourly cost for producing 95 mol% and 98 mol% O_2, respectively, compared with a double-column ASU.

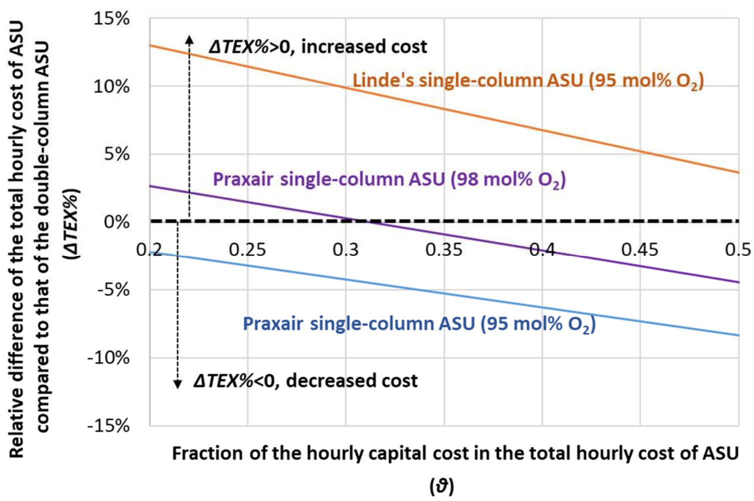


Figure 10. The relative difference in the hourly total cost of the single-column ASU (Praxair and Linde's), compared to that of the conventional double-column ASU, vary with the fraction of the hourly capital cost in the total hourly cost of ASU, when producing 95 mol% and 98 mol% O_2.

4.3 Limitations and future work

Based on the present configuration, the PSC-ASU is not competitive with the nitrogen-refluxed single-column ASUs in terms of total hourly cost. This is because the PSC has a lower O_2 recovery rate compared with the nitrogen-refluxed column. However, in the present work the PSC-ASU was not systematically optimized, while the nitrogen-refluxed single-column ASUs has been improved over past decade. Therefore, future work should focus on optimization of the PSC-ASU to systematically lower total hourly cost. This optimization should also consider producing O_2 at higher purity (>98 mol%) for the Allam cycel. Based on Luyben's study, a control problem or a dynamic issue may occur on a column with a series of reboilers [36], which would decrease the ASU flexibility. To understand this issue, a dynamics study of the PSC-ASU should be conducted, with a focus on the operation of the multi-reboiler. Moreover, to explore the optimum application of the single-column ASU for low-carbon, fossil fuel power plants, a detailed economic analysis on the hourly capital cost and hourly operation cost of the ASU should be conducted.

5. Conclusion

In this work, we studied the Praxair single column ASU for producing O_2. The PSC-ASU is characterized with partial air condensation and air pre-separation in the bottom reboiler. Three ASU designs were modeled in Aspen Plus and compared in this study: 1) the state-of-the-art double-column ASU, 2) Linde's conventional single-column ASU, and 3) the PSC-ASU. Simulation results suggest that, compared with Linde's conventional single-column ASU, the PSC-ASU enhances the O_2 recovery rate to 82% (for an O_2 product purity of 95 mol%) by providing enriched-N_2 liquid reflux (87 mol% N_2). Moreover, the air pressure from the MAC in the PSC-ASU can be reduced to below 4 bar when producing relatively low purity O_2 ($\leqslant$95.1 mol%), leading to less energy consumption. The PCS-ASU with the internal O_2 compression can produce 95.1 mol% O_2 product at a pressure of 15 bar with an energy consumption of 0.53 kW·h/Nm^3, which is 14.5% lower than that of Linde's single-column ASU, and only 1.9% higher than that of the conventional double-column ASU (0.52 kW·h/Nm^3). However, the PSC-ASU reduces the hourly capital cost by about 19% by eliminating a high-pressure column, and enables a faster startup, compared with the conventional double-column ASU. Considering that the future market needs for low-carbon fossil-fuel plants will be driven towards lower capital cost and high flexibility, the PSC-ASU could be very attractive.

Acknowledgment

This material is based upon works supported by the Department of Energy, USA, under Award Number DE-FE0009702 and the US-China Clean Energy Research Center - Advanced Coal Technology Collaboration, and by the Consortium for Clean Coal Utilization at Washington University in St. Louis. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Appendix B. Model validation

Nomenclature

Abbreviations

ABC	Air Booster Compressor
ASU	Air Separation Unit
CCS	Carbon Capture and Storage
IGCC	Integrated Gasification Combined Cycle
MAC	Main Air Compressor
PSC	Praxair Single Column
PSC-ASU	Praxair Single-Column ASU
RVRC	Recuperative Vapor Recompression

Symbols

c_O	O_2 purity of air feed (obtained by material balance) (mole fraction)
E	Total energy consumed by ASU (kW·h)
E_B	Energy consumed by ABC (kW·h)
E_C	Energy consumed by MAC (kW·h)

E_O	Specific energy consumption for producing a unit amount of O_2 (kW·h/Nm^3)
$E_{O,D}$	Specific energy consumption of double-column ASU (kW·h/Nm^3)
$E_{O,S}$	Specific energy consumption of single-column ASU (kW·h/Nm^3)
k	Heat capacity ratio (C_P/C_V)
N_B	Number of compression stages in ABC
N_C	Number of compression stages in MAC
n_B	Mole flow rate of the boosted air (kmol/h)
n_F	Mole flow rate of the air feed (kmol/h)
p_B	Air pressure at ABC outlet (bar)
p_C	Air pressure at MAC outlet (bar)
p_I	Air pressure at process inlet (atmosphere pressure) (bar)
SC	Scale of single-column ASU at O_2 recovery rate of η_D
SC_S	Scale of single-column ASU at O_2 recovery rate of η_S
T_{bu}	Liquid bubble point at cold-side inlet of bottom reboiler (°C)
T_I	Air temperature at the inlet of MAC and ABC (°C)
V_F	Air feed rate (Nm^3/h)
V_O	Normal volumetric flow rate of the pure O_2 (Nm^3/h)
ΔCPEX%	Relative difference in hourly **capital** cost of single-column ASU, compared with that of the double-column ASU
ΔOPEX%	Relative difference in hourly **operation** cost of single-column ASU, compared with that of the double-column ASU
ΔTEX%	Relative difference in **total** hourly cost of single-column ASU, compared with that of the double-column ASU

Greeks

η	O_2 recovery rate of ASU
η_D	O_2 recovery rate of double-column ASU
η_S	O_2 recovery rate of single-column ASU
η_e	Isentropic efficiency of each stage in MAC and ABC
φ	Internal compression ratio
θ	Fraction of hourly capital cost in total hourly cost of the conventional double-column ASU

Subscript

B	ABC or boosted air
bu	Bubble
C	MAC
D	Double column
e	Efficiency
F	Air feed
I	Process inlet or compressor inlet
O	Oxygen or a unit amount of oxygen
S	Single column

References

[1] International Energy Agency. Status of Power System Transformation 2019. Paris: IEA; 2019. https://www.iea.org/reports/status-of-power-system-transformation-2019

[2] Gonzalez-Salazar MA, Kirsten T, Prchlik L. Review of the operational flexibility and emissions of gas-and coal-fired power plants in a future with growing renewables. Renewable and Sustainable Energy Reviews 2018;82:1497-513. https://doi.org/10.1016/j.rser.2017.05.278

[3] International Renewable Energy Agency. Innovation landscape brief: Flexibility in conventional power plants. Abu Dhabi: IRENA; 2019. https://www.irena.org/-/media/Files/IRENA/Agency/Publication/2019/Sep/IRENA_Flexibility_in_CPPs_2019.pdf?la=en&hash=AF60106EA083E492638D8FA9ADF7FD099259F5A1

[4] Domenichini R, Mancuso L, Ferrari N, Davison J. Operating Flexibility of Power Plants with Carbon Capture and Storage (CCS). Energy Procedia 2013;37:2727-37. https://doi.org/10.1016/j.egypro.2013.06.157

[5] Allam R, Martin S, Forrest B, Fetvedt J, Lu X, Freed D, et al. Demonstration of the Allam Cycle: An Update on the Development Status of a High Efficiency Supercritical Carbon Dioxide Power Process Employing Full Carbon Capture. Energy Procedia 2017;114:5948-66. https://doi.org/10.1016/j.egypro.2017.03.1731

[6] Allam RJ, Palmer MR, Brown GW, Fetvedt J, Freed D, Nomoto H, et al. High Efficiency and Low Cost of Electricity Generation from Fossil Fuels While Eliminating Atmospheric Emissions, Including Carbon Dioxide. Energy Procedia 2013;37:1135-49. https://doi.org/10.1016/j.egypro.2013.05.211

[7] Skorek-Osikowska A, Bartela L, Kotowicz J, Job M. Thermodynamic and economic analysis of the different variants of a coal-fired, 460MW power plant using oxy-combustion technology. Energy Conversion and Management 2013;76:109-20. https://doi.org/10.1016/j.enconman.2013.07.032

[8] Scheffknecht G, Al-Makhadmeh L, Schnell U, Maier J. Oxy-fuel coal combustion—A review of the current state-of-the-art. International Journal of Greenhouse Gas Control 2011;5:S16-S35. https://doi.org/10.1016/j.ijggc.2011.05.020

[9] Wu F, Argyle MD, Dellenback PA, Fan M. Progress in O2 separation for oxy-fuel combustion–A promising way for cost-effective CO2 capture: A review. Progress in Energy and Combustion Science 2018;67:188-205. https://doi.org/10.1016/j.pecs.2018.01.004

[10] Lockwood T. Developments in oxyfuel combustion of coal. IEA Clean Coal Centre 2014;300:240.

[11] Seliger B, Hanke-Rauschenbach R, Hannemann F, Sundmacher K. Modelling and dynamics of an air separation rectification column as part of an IGCC power plant. Separation and Purification Technology 2006;49:136-48. https://doi.org/10.1016/j.seppur.2005.09.007

[12] Andersson K, Johnsson F. Process evaluation of an 865 MWe lignite fired O2/CO2 power plant. Energy Conversion and Management 2006;47:3487-98. https://doi.org/10.1016/j.enconman.2005.10.017

[13] Eyre D, Gorup I, Pawulski T. PRODUCTION OF OXYGEN-KEEPING PACE WITH METALLURGICAL DEMANDS. Proceedings of the Metallurgical Society of the Canadian Institute of Mining and Metallurgy. Elsevier, 1987. pp. 77-85.

[14] Agrawal R, Herron DM. AIR LIQUEFACTION: DISTILLATION. Encyclopedia of Separation Science, 2000.

[15] DAVIES M. THE PRINCIPLES AND PRACTICE OF AIR SEPARATION. Science Progress (1933-) 1952;40:419-35.

[16] Linde Engineering. Air separation plants. https://www.linde-engineering.com/en/images/Air-separation-plants-history-and-technological-progress-2019_tcm19-457349.pdf (accessed 2020)

[17] Taniguchi M, Asaoka H, Ayuhara T. Energy saving air-separation plant based on exergy analysis. Kobelco Technology Review 2015;33:34-8.

[18] Linde Engineering. Oxygen production for oxyfuel power plants: status of development. http://oxyfbc.eu-projects.de/Portals/29/OxyFBC/10_Goloubev_Linde%20-%20Oxygen%20Production%20for%20Oxyfuel%20Power%20Plants.pdf (accessed 2021)

[19] Higginbotham P, White V, Fogash K, Guvelioglu G. Oxygen supply for oxycoal CO2 capture. Energy Procedia 2011;4:884-91. https://doi.org/10.1016/j.egypro.2011.01.133

[20] van der Ham LV. Improving the exergy efficiency of a cryogenic air separation unit as part of an integrated gasification combined cycle. Energy Conversion and Management 2012;61:31-42. https://doi.org/10.1016/j.enconman.2012.03.004

[21] Ebrahimi A, Meratizaman M, Akbarpour Reyhani H, Pourali O, Amidpour M. Energetic, exergetic and economic assessment of oxygen production from two columns cryogenic air separation unit. Energy 2015;90:1298-316. https://doi.org/10.1016/j.energy.2015.06.083

[22] Tesch S, Morosuk T, Tsatsaronis G. Comparative evaluation of cryogenic air separation units from the exergetic and economic points of view. Low-temperature Technologies 2019;30:159. 10.5772/INTECHOPEN.85765

[23] Bonaquist DP. Single column cryogenic rectification system for lower purity oxygen production. USA Patent 5,832,748. 1998-11-10

[24] Kender R, Wunderlich B, Thomas I, Peschel A, Rehfeldt S, Klein H. Pressure-driven dynamic simulation of start up and shutdown procedures of distillation columns in air separation units. Chemical Engineering Research and Design 2019;147:98-112. https://doi.org/10.1016/j.cherd.2019.04.031

[25] Brigagão GV, de Medeiros JL, Araújo OdQF. A novel cryogenic vapor-recompression air separation unit integrated to oxyfuel combined-cycle gas-to-wire plant with carbon dioxide enhanced oil recovery: Energy and economic assessments. Energy Conversion and Management 2019;189:202-14. https://doi.org/10.1016/j.enconman.2019.03.088

[26] Kansha Y, Kishimoto A, Nakagawa T, Tsutsumi A. A novel cryogenic air separation process based on self-heat recuperation. Separation and Purification Technology 2011;77:389-96. https://doi.org/10.1016/j.seppur.2011.01.012

[27] Fu C, Gundersen T. Recuperative vapor recompression heat pumps in cryogenic air separation processes. Energy 2013;59:708-18. https://doi.org/10.1016/j.energy.2013.06.055

[28] Luyben WL. Control of a recuperative vapor- recompression air separation process. Journal of Process Control 2016;45:55-64. https://doi.org/10.1016/j.jprocont.2016.06.007

[29] Zhou H, Cai Y, Xiao Y, Mkhalel ZA, You B, Shi J, et al. Process Configurations and Simulations for a Novel Single-Column Cryogenic Air Separation Process. Industrial & Engineering Chemistry Research 2012;51:15431-9. 10.1021/ie3022225

[30] Singla R, Chowdhury K. Enhanced oxygen recovery and energy efficiency in a reconfigured single column air separation unit producing pure and impure oxygen simultaneously. Chemical Engineering and Processing - Process Intensification 2021;162:108354. https://doi.org/10.1016/j.cep.2021.108354

[31] Fu C, Gundersen T. Using exergy analysis to reduce power consumption in air separation units for oxy-combustion processes. Energy 2012;44:60-8. https://doi.org/10.1016/j.energy.2012.01.065

[32] Yang Z, Khatri D, Verma P, Li T, Adeosun A, Kumfer BM, et al. Experimental study and demonstration of pilot-scale, dry feed, oxy-coal combustion under pressure. Applied Energy 2021;285:116367. https://doi.org/10.1016/j.apenergy.2020.116367

[33] Linde Engineering. Plate-fin heat exchangers (PFHEs). https://www.linde-engineering.com/en/images/Aluminium-plate-fin-heat-exchanger-2019_tcm19-5772.pdf (accessed 2021)

[34] Loh HP , Lyons J , White CW. Process Equipment Cost Estimation Final Report. Morgantown: NETL; 2002. https://doi.org/10.2172/797810

[35] US Department of Energy. Coal FIRST. https://www.energy.gov/fe/coal-first (accessed 2021)

[36] Luyben WL. Series versus parallel reboilers in distillation columns. Chemical Engineering Research and Design 2018;133:294-302. https://doi.org/10.1016/j.cherd.2018.03.025