

# New Process Flowsheet for CO<sub>2</sub> Compression and Purification Unit; Dynamic Investigation and Control

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**ABSTRACT:** *The present study provides a novel process flowsheet for CO<sub>2</sub> compression and purification unit (CPU) in order to improve its product quality and control performance. Unlike the previous process flowsheet, the number of cold-boxes has been reduced to one, which in turn decreases investment costs and improves energy integration. The performance of the proposed flowsheet was compared with two recently suggested ones for a given feed. The results showed that, compared with the other process flowsheet, the new one not only can operate at lower operating pressure but also needs a significantly smaller heat-transfer area. Also the dynamic behavior and controllability of the proposed process flowsheet are analyzed to ensure the proper functioning. The control loops used in the new flowsheet were simpler than those used in the previous flowsheet, and controllability was achieved using proportional (P) and proportional-integral (PI) controllers, which offers a performance advantage over the other process flowsheet. Using step changes, the effects of disturbances in feed temperature, flow rate, and composition on the final product specifications were also investigated. The proposed flowsheet process proved to be robust against the disturbances and the control structure was able to handle them appropriately. The proposed process flowsheet was also able to maintain purity and recovery rates of 96.74% and 90.08%, respectively, in the face of disturbance.*

**KEYWORDS:** *CO<sub>2</sub>-CPU; Process control; Oxy-fuel combustion; CO<sub>2</sub> capture; Dynamic modeling.*

## INTRODUCTION

In recent years raising the Earth's average surface temperature has become the major concern of many researchers [1]. A reason for this problem is increasing greenhouse gases specifically carbon dioxide (CO<sub>2</sub>) [2-4]. Therefore separation of this pollutant from flue gases has taken on great importance, both environmentally [5, 6] and economically [7, 8]. Combustion is one of the main sources of producing CO<sub>2</sub>. CO<sub>2</sub> capture approaches are generally

classified into pre-combustion, post-combustion, and oxy-fuel combustion [9] while each of them utilizes various technologies. Compared with other approaches, oxyfuel combustion capture has attracted more attention [10]. This is because of producing CO<sub>2</sub> in high concentrations that can be separated easily through compression and refrigeration systems [11]. The most widely used technology in this approach is the use of CO<sub>2</sub> Compression

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1021-9986/2021/2/593-604

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and Purification Unit (CPU) suggested by International Energy Agency Greenhouse Gas (IEAGHG) R&D Programme [12]. A number of studies have been conducted with the purpose of modeling the process [13], operating optimization of the process [14], parameter sensitivity analysis [15], identifying dynamics and developing control structure [16, 17] for CPU process.

*Jin et al.* [18], proposed and examined a systematic dynamic exergy method for identifying exergy performance dynamically to achieve efficient operation. They showed that dynamic exergy method would be a strong methodology to identify important information [18]. They reported that CPU is more sensitive to flue gas composition compared to flow rate as well as flow gas composition ramp up process runs more effectively than that of flow rate ramp down and composition ramp down processes [18]. *Fu and Gundersen* [19] conducted a techno-economic analysis of CO<sub>2</sub>-CPU structures with one, two, and three separators and found that, with increased separation stages in a given CO<sub>2</sub> purity, the CO<sub>2</sub> recovery rate increases, energy consumption in the process decreases and the total cost increases significantly. Based on economic analysis, they concluded that the CO<sub>2</sub>-CPU process with two separators was the most cost-effective option [19]. *Posch and Haider* [14] optimized and compared two types of CO<sub>2</sub>-CPU, namely, a double flash separation unit and a separation unit based on rectification. Their results showed that CO<sub>2</sub>-CPU with distillation column would achieve higher purity, but it required greater power and cooling duty (~30%) compared with the double flash unit [14]. They also reported that separation efficiency increased as pressure increased, although *Koohestanian et al.* [15] showed that pressures greater than 35 bar would result in contamination, thereby decreasing purity. *Chansomwong et al.* [13] performed a dynamic modeling of the behavior of a CO<sub>2</sub>-CPU process and found that the behavior of the system was highly nonlinear. Their results showed that the operating conditions of the first separator played a key role in performance of the process [13]. They also reported that CO<sub>2</sub> recovery and purity depended on operating conditions and feed composition, respectively. In an interesting study, *Jin et al.* [16] designed a control system and performed a process optimization analysis for CO<sub>2</sub>-CPU using Aspen Plus® and Aspen Dynamics® and proposed optimal operating conditions for the process. However, Luyben stated

that the proposed control system, apart from being overly complicated, would not be robust [17]. Luyben proposed a new structure for CO<sub>2</sub>-CPU process [17]. In the proposed flowsheet, the degree of freedom was increased by increasing the number of cold-boxes to three, providing a simple control structure for CPU process. Although the use of simpler control structures has always been appealing from a practical point, the increase in the number of cold-boxes will inevitably increase the investment cost. Furthermore, Luyben had not considered thermal stabilization of the first separator in his work, which could result in saturation of control valves in the long term. According to API RP 521 [20], in a series of columns where one column's output is fed to the next one, regulation of thermal load of individual columns is of great importance because temperature decline in a column could disturb the performance of the next one. Failure to regulate the temperature of the first separator will lead to very little, but constant, decreases in liquid level within the first separator, which in turn will increase the liquid level in the second separator. Considering that these conditions have to be adjusted to avoid heating up or freezing, the control valves will be saturated in the long run, resulting in a compromised process control. *Jin et al.* [16] had not taken into account the interactions among parameters in process optimization. Therefore, *Koohestanian et al.* [15], performed a sensitivity analysis and process optimization for CO<sub>2</sub>-CPU using response surface methodology and presented a new optimal operating condition. Statistical analysis showed that the first separator temperature played a key role in the total work and heat duty of the process [15]. Considering the interactions among parameters, and using statistical analysis, they proposed new operating conditions where the operating pressure was reduced to 29 bar, decreasing operational and the investment costs [15]. However, the size of cold-boxes, as a key contributor to the decreased costs, was not investigated in that study.

While there have been a number of studies conducted on CPU processes issue, no research has focused on the modification of the CPU flowsheet. Therefore, based on the optimal operating conditions proposed in literature [15], we aim in this study to produce improvements in the structure of the process and its practical view of controllability. The steady-state simulation of the new process flowsheet was performed and tested using Aspen Plus®. To ensure the stability and controllability

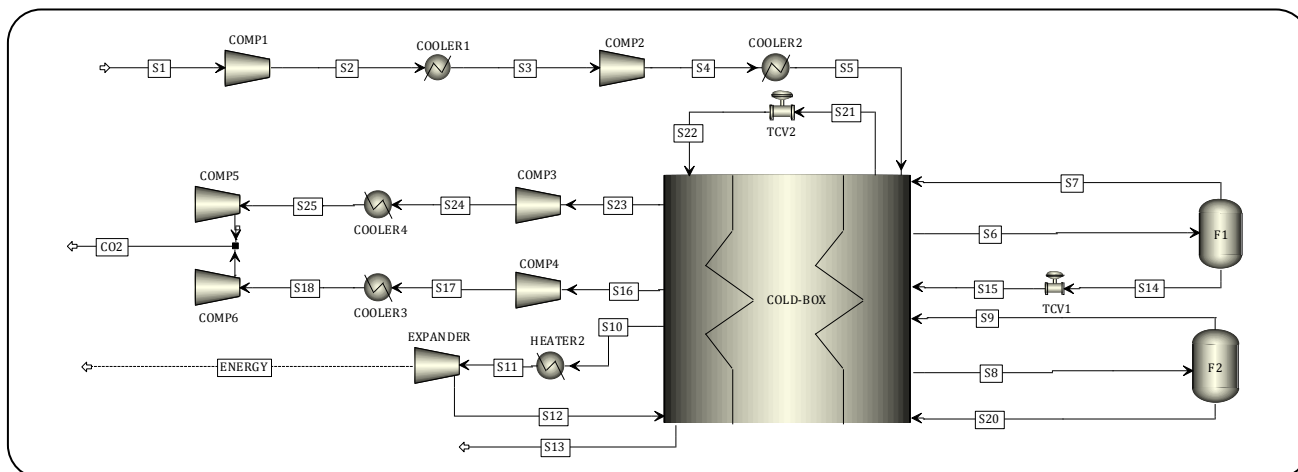


Fig. 1. PFD of the proposed flowsheet.

of the process against potential disturbances, the effects of variation in flowrate, feed temperature and composition on process dynamics, purity, and recovery rate, explored using Aspen Dynamics®. To achieve that, the controllers structure were designed and applied. The controllers were adjusted using process reaction curve and integral of Time-wWighted Absolute error (ITAE) criterion [21]. Dynamics of the process against step change in feed temperature, flowrate and feed composition were then investigated.

## SIMULATION DETAILS AND DESIGN

### Flowsheet description

Fig. 1 illustrates the structure of CO<sub>2</sub>-CPU designed in this study. Data of the flue gas were extracted from literature [17]. The dehydrated flue gas (S1) compressed through a two-stage compressor and enters the cold-box at 80°C (stream S5). A pressure drop of 0.21 bar has been assumed through the cold-box. Stream S5 is cooled down to -35°C to separate the liquefied CO<sub>2</sub>, and then enters the first separator F1. The separators are assumed adiabatic and the pressure drop inside them has been neglected. The output flow from the bottom of this separator (S14) is expanded through a valve and cools down to -55°C due to Joule-Thomson effect. Although greater cooling is achievable through greater pressure drops in the valves, we cautiously chose this temperature to prevent CO<sub>2</sub> from freezing (which occurs at -56.6°C) and avoid hydrate formation and erosion of the pipes. The stream from the first separator (S7) is cooled to -54°C and fed to the second separator (S8). The purity of CO<sub>2</sub> in the final product needs to be greater than 95% [22, 23]. In the operating pressure of 29 bar, CO<sub>2</sub>

occurs in liquid state, therefore eliminating the concerns about the purity of the final CO<sub>2</sub> product. The results for CO<sub>2</sub> purity in the final product confirms this (96.74%).

The high-pressure stream from the second separator (S9) enters the cold-box and leaves it at 75°C (S10). In exchange for this reduced pressure, power is generated in a turbine. Since turbines are expensive equipment, and to decrease fluctuations in the stream fed to the turbine, the stream is heated to 100°C. This increased temperature will also help increase the power generated. The stream leaves the turbine (S12) at 2.2 bar and -37°C and reenters the cold-box for its cooling capacity to be used, leaving the cold-box at 28°C (S13). This stream can be fed to an amine or ammonia-based CO<sub>2</sub> capture unit.

The stream from the bottom of the second separator (S20), which contains almost pure CO<sub>2</sub>, is fed to the cold-box and heated to -35°C and then is flashed through the valve TCV2 to reach pressure and temperature of 7.3 bar and -55°C. The stream, then, is fed back to the cold-box to leave it at 50°C (S23). The CO<sub>2</sub> streams from cold-boxes (namely, S16 and S23) merge together after being pressurized to 97 bar and leave the process as the final CO<sub>2</sub> product and as a possible feed to urea plant.

This process was simulated with Aspen Plus® using Peng-Robinson equation of state recommended for nonpolar or mildly polar mixtures by AspenTech company [24]. Furthermore, experimental data were used [25] to improve thermodynamic coefficients to increase the accuracy of simulation.

Finally, the performance of these three flowsheets were compared using given data on flue gas composition [17]

at a given flowrate (596069 kg/h). The proposed control structure was also tested with different feed compositions, and its controllability was investigated. Maximum operating pressure (MOP) for cryogenic process as a factor affecting compressor power consumption and the thickness of process equipment was also compared among structures. Heat-transfer area, CO<sub>2</sub> purity, and CO<sub>2</sub> recovery of these three processes were compared. To increase the accuracy of the calculations and ensure the prevention of temperature cross, a minimum of 120 zones for the proposed cold-box is considered before performing the energy analysis. Since most streams were in gas state, the overall heat-transfer coefficient is assumed to be  $U=170 \text{ W/m}^2\text{K}$  [26]. The cold-box heat-transfer area ( $A$ ) can be obtained using Eq. 1:

$$A = \frac{\text{Duty}}{U.L.M.T.D} \quad (1)$$

Details on calculation of volumes of each stream can be found in the literature [17]. For determining the size of separators (Eq. (3)), the maximum velocity of the vapor stream was used based on Eq. (2) [27]. In this equation,  $F_{\text{Factor}}$  is equal to 1.0 in English Engineering measurement system and 0.6 in SI system [27].

$$F_{\text{Factor}} = V_{\text{max}} \sqrt{\rho_v} \quad (2)$$

where  $V_{\text{max}}$  is the maximum vapor velocity in m/s and  $\rho_v$  is the vapor density in kg/m<sup>3</sup>.

$$D = 2 \sqrt{\frac{Q}{\pi V_{\text{max}}}} \quad (3)$$

In Eq. (3),  $D$  is the diameter of the separator and  $Q$  is the vapor volumetric flowrate in m<sup>3</sup>/s. The aspect ratio  $L/D$  was considered 2. Also, the recovery rate in each structure was calculated using Eq. 4.

$$\text{Recovery} = \frac{\text{Total CO}_2 \text{ in product stream}}{\text{Total CO}_2 \text{ in feed stream}} \quad (4)$$

### Dynamic model

To ensure the proper performance of the process against potential disturbances, we conducted a dynamic simulation of the process using Aspen Dynamics®. To this end, the steady-state simulation in Aspen Plus® was exported into Aspen Dynamics® and the dynamic modeling was carried out using pressure-driven approach to accommodate more

rigorous dynamic performance [28, 29]. Fig. 2 shows the Process Flow Diagram (PFD) and the control structure. The feed flowrate is controlled by break-horse power of the compressors COMP1 and COMP2. These two stages will keep compressor discharge temperature below the maximum allowable temperature. The maximum allowable temperature can be set either by characteristics of the compressor cylinder or by the gas specifications such as temperature decomposition, or auto ignition [30]. According to CO<sub>2</sub> physical properties, the maximum allowable temperature can be considered as 260°C [31]. As the cold box is a heat exchanger with zero net duty, control of input temperatures can guarantee its performance. However, the temperature fluctuations during controlling the unit make reduces the robustness of the control performance. The more robust way to overcome this problem is using manipulation of input stream enthalpy to the cold box. Therefore, the proposed control structure to control drum F1 temperature (as output control variable) with duty of COOLER2 (as manipulation) were considered. Therefore, COOLER1 controls inter-stage temperature, and COOLER2 regulates the flash drum F1 temperature.

According to previous research [13, 15] as well as the results of the present study, precise control of the temperature of the first separator (F1), achieved by COOLER2, is of great significance. The liquid level in the drums is controlled by valves TCV1 and TCV2. To avoid freezing of the S15 and S22 streams due to Joule-Thomson effect, the stream temperature was controlled by compressors COMP3 and COMP4.

### Design and control limitations

An open-loop test system was performed for the regulation of control parameters. Design and control limitation are as bellow:

- The feed flowrate is set upstream. The unit must therefore be able to handle disturbances in feed composition and flowrate.
- The temperature of the first separator as the most important part of the process should be controlled precisely.
- The temperature in the process should always be above the freezing point of CO<sub>2</sub> (-56.6°C).
- S22 and S15 streams contribute greatly to the cooling of the process. Therefore, their temperatures need to be controlled precisely.

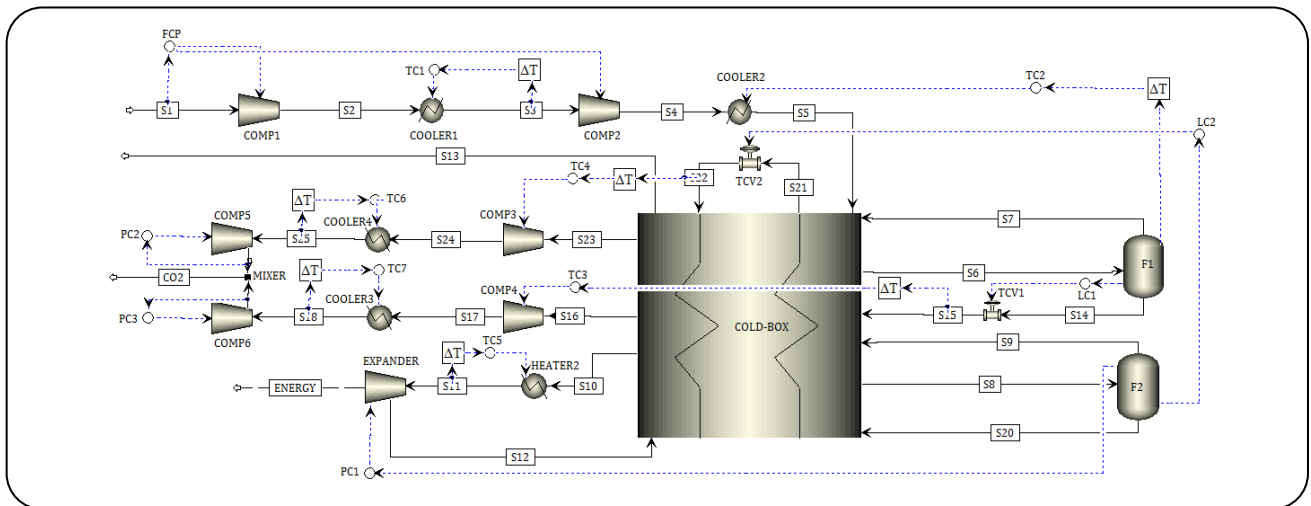


Fig. 2: The control structure of new proposed design.

- The minimum CO<sub>2</sub> purity in the product gas must be 95 mole % [22, 23]. Considering the operating temperatures of the separators, namely, -55°C and -35°C, respectively, at which CO<sub>2</sub> will occur in liquid phase, there is no concern regarding the violation of this condition. Therefore, no explicit controller has been set for CO<sub>2</sub> purity as this will be achieved by precise controlling of other operating conditions.

- The recovery rate should not be less than 90%.
- The liquid levels in the separators must be sustained between maximum and minimum limits.
- The product gas should be delivered at 97 bar.
- The output flow pressure from the turbine should be approximately 2.2 bar, as it is assumed here that the stream from turbine enters a capture unit for further recovery. Otherwise, the pressure can be reduced to atmospheric pressure, which will increase power generated and will improve energy integration.
- Temperature measurements have a one minute delay.

## RESULTS AND DISCUSSION

### Steady-state results

Table 1 presents the results of mass-energy balance analysis for the proposed flowsheet based on feed composition. The CO<sub>2</sub> purity and recovery in the product were about 97%, and 90% respectively which were quite desirable.

The results of steady-state process output based on the feed composition in the study of *Jin et al.* [16] have been summarized in Appendix A. The results point to

the stability of the process against changes in feed specification.

The composite curves for the streams within the cold-box (Fig. 3) confirms that no temperature cross has occurred. Except for the beginning and end points of the curves, where the difference is 5°C and 1°C, respectively, the curves are in appropriate distances from each other, ensuring the absence of temperature cross. Therefore, the proposed flowsheet will remain robust against disturbances if the beginning and end of the process are controlled properly.

The main equipment sizing was carried out based on Eq. 1–3, results of which are presented in Table 2.

Finally, before addressing the controllability of the process, the results of the comparison of the three flowsheet have been presented in Table 3. The proposed flowsheet in the present study provides a greater purity as well as having a decreased MOP and A. Decreased MOP and A will contribute to reduce operational and investment costs. Although the recovery rate in our structure was slightly smaller than those of the other two, the difference is negligible.

A sensitivity analysis was performed to evaluate the changes in recovery and purity response to pressure changes (Fig. 4). CO<sub>2</sub> recovery increased with increasing pressure; however, the product purity would be decreased as a result of contamination by other particles. According to Table 3, CO<sub>2</sub> recovery in the *Jin's* research is more and the pinch point's in his research was 0.37°C, while it considered 1°C at the present study. Although the pinch

Table 1: Details of the streams and mass-energy balance.

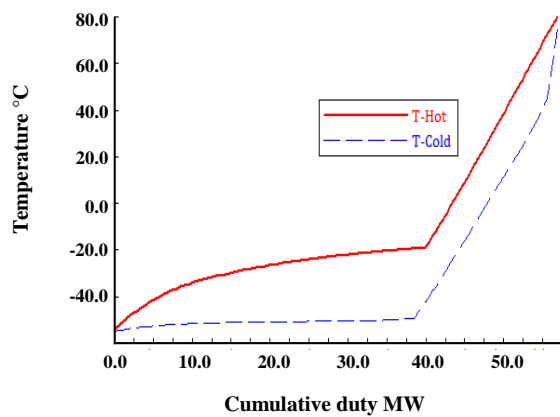
Stream no.	S1	S2	S3	S4	S5	S6
Composition Mole %						
CO <sub>2</sub>	75.96	75.96	75.96	75.96	75.96	75.96
O <sub>2</sub>	6.27	6.27	6.27	6.27	6.27	6.27
N <sub>2</sub>	15.30	15.30	15.30	15.30	15.30	15.30
Ar	2.46	2.46	2.46	2.46	2.46	2.46
Flowrate, kg/hr	596069.0	596069.0	596069.0	596069.0	596069.0	596069.0
Temperature, °C	20.0	203.0	25.0	207.7	80.0	-35.0
Pressure, Bar	1.10	5.90	5.55	29.0	28.65	28.44
Stream no.	S7	S8	S9	S10	S11	S12
Composition Mole %						
CO <sub>2</sub>	48.89	48.89	25.75	25.75	25.75	25.75
O <sub>2</sub>	13.14	13.14	18.97	18.97	18.97	18.97
N <sub>2</sub>	32.78	32.78	47.77	47.77	47.77	47.77
Ar	5.18	5.18	7.51	7.51	7.51	7.51
Flowrate, kg/hr	236347.9	236347.9	144793.2	144793.2	144793.2	144793.2
Temperature, °C	-35.0	-54.0	-54.0	75.0	100.0	-36.9
Pressure, Bar	28.44	28.23	28.23	28.02	27.68	2.21
Stream no.	S13	S14	S15	S16	S17	S18
Composition Mole %						
CO <sub>2</sub>	25.75	96.93	96.93	96.93	96.93	96.93
O <sub>2</sub>	18.97	0.95	0.95	0.95	0.95	0.95
N <sub>2</sub>	47.77	1.76	1.76	1.76	1.76	1.76
Ar	7.51	0.35	0.35	0.35	0.35	0.35
Flowrate, kg/hr	144793.2	359721.1	359721.1	359721.1	359721.1	359721.1
Temperature, °C	28.0	-35.0	-55.0	43.9	156.5	25.0
Pressure, Bar	2.00	28.44	6.95	6.74	20.0	19.65
Stream no.	S19	S20	S21	S22	S23	S24
Composition Mole %						
CO <sub>2</sub>	96.93	95.97	95.97	95.97	95.97	95.97
O <sub>2</sub>	0.95	1.28	1.28	1.28	1.28	1.28
N <sub>2</sub>	1.76	2.29	2.29	2.29	2.29	2.29
Ar	0.35	0.46	0.46	0.46	0.46	0.46
Flowrate, kg/hr	359721.1	91554.7	91554.7	91554.7	91554.7	91554.7
Temperature, °C	192.2	-54.0	-35.0	-55.0	50.0	158.5
Pressure, Bar	97.0	28.23	28.02	7.31	7.10	20.0
Stream no.	S25	S26	CO2			
Composition Mole %						
CO <sub>2</sub>	95.97	95.97	96.74			
O <sub>2</sub>	1.28	1.28	1.02			
N <sub>2</sub>	2.29	2.29	1.87			
Ar	0.46	0.46	0.37			
Flowrate, kg/hr	91554.7	91554.7	451275.8			
Temperature, °C	25.0	192.6	192.3			
Pressure, Bar	19.65	97.0	97.0			

**Table 2: Sizing of the main equipment.**

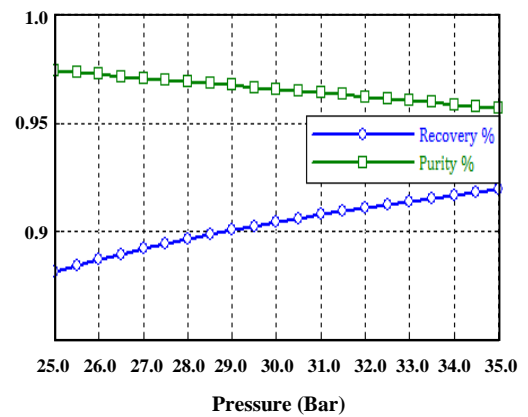
Equipment	Volume (m <sup>3</sup> )	Parameter	Value
F1	143.5	Length	9 m
		Diameter	4.5 m
F2	101.0	Length	8.0 m
		Diameter	4.0 m
		Stream volume vs. m <sup>3</sup>	
COLD-BOX	254	S5	213.0
		S7	41.0
		S9	23.0
		S12	10.5
		S15	172
		S20	4.5
		S22	44.0

**Table 3: Comparison of the three flowsheet.**

		Luyben research [17]	Jin research [16]	This research
Cold-Box 1	Duty (MW)	5.108	27.525	56.858
	LMTD (K)	11.04	4.94	17.00
	A (m <sup>2</sup> )	2721	32786	19674
Cold-Box 2	Duty (MW)	25.910	22.007	-
	LMTD (K)	4.71	4.1	-
	A(m <sup>2</sup> )	32320	31750	-
Cold-Box 3	Duty (MW)	21.400	-	-
	LMTD (K)	5.71	-	-
	A(m <sup>2</sup> )	22000	-	-
Total area (m <sup>2</sup> )		57041	64536	19674
Purity %		95.17	96.65	96.74
Recovery %		90.29	90.82	90.08
MOP (Bar)		30	30	29



**Fig. 3: The composite curve for the cold-box in the new flowsheet.**



**Fig. 4: Purity and recovery rate vs., pressure.**

Table 4: Controller parameters.

Name	Controller type	Controller action	Gain	Integral time
FPC	PI	Reverse	1.6431	1.0158
TC1	PI	Reverse	0.0934	1.5022
TC2	PI	Reverse	6.7216	3.7345
TC3	PI	Direct	2.7102	3.1932
TC4	PI	Direct	3.7667	1.6435
TC5	PI	Reverse	0.6760	1.6187
TC6	PI	Reverse	0.0581	1.6401
TC7	PI	Reverse	0.2313	1.9438
PC1	PI	Reverse	3.2042	3.7009
PC2	PI	Reverse	0.3	0.5
PC3	PI	Reverse	0.3	0.5
LC1	P	Direct	0.1	-
LC2	P	Direct	2.6525	-

point difference of  $0.37^{\circ}\text{C}$  increase recovery, but the proposed control become more complicated and it also increase the A of cold-box. Furthermore, at the present study, the pressure drop of streams within the cold-box is considered as 0.21 bar that according to Fig. 4, increased pressure drop, it will reduce recovery. However, at a pinch temperature difference and a uniform pressure difference, the proposed flowsheet will increase to 90.82%.

#### Dynamic and control results

The controllers were tuned based on the process reaction curve. Controller specifications have been presented in Table 4.

Robustness of the process control structure was tested against changes in flowrate, composition, and temperature of the input gas. Fig. 5 demonstrates the process response to  $\pm 5\%$  step change in feed flowrate. The proposed flowsheet is quite robust against disturbances in feed flowrate in a short time. The results of minor test shows that the system is strongly nonlinear, and operating in low flowrates for a long time would require the controllers to be retuned.

Fig. 6 shows the process response to  $\pm 5\%$  change in  $\text{CO}_2$  composition of the feed. According to the figure, the proposed flowsheet is robust against changes in  $\text{CO}_2$  composition. Comparing figures 5 and 6 reveals that the process is more sensitive to disturbances in flowrate

than flow composition. In other words, due to the nonlinearity of the process, changes in either feed flowrate or feed composition would result in alteration of process parameters, with this alteration being more significant for flowrate disturbance. Therefore, it is recommended that controllers be retuned if the change in flowrate is going to be permanent.

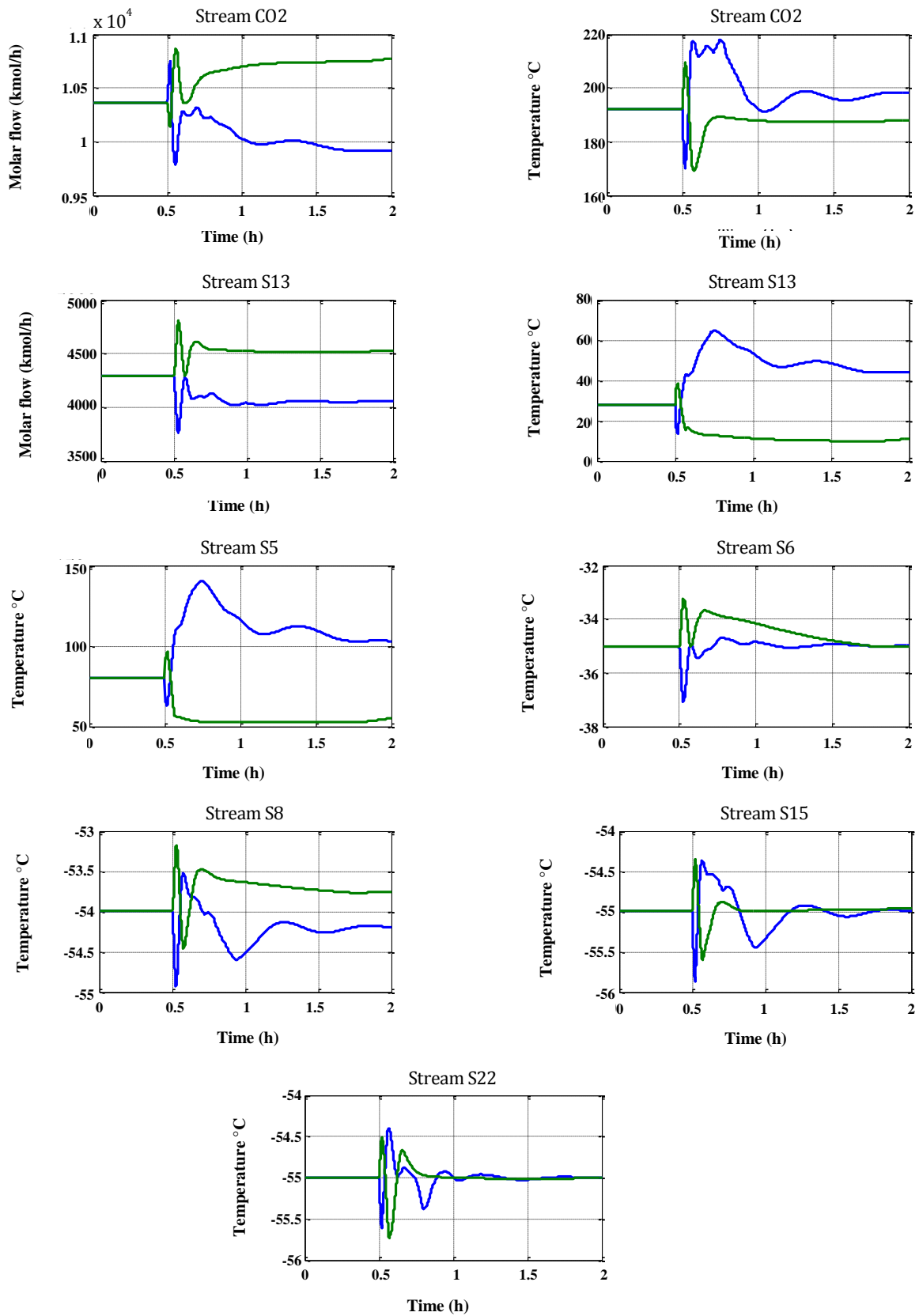
Fig. 7 shows the process response to  $\pm 5\%$  change in feed temperature. The proposed structure provides a quick response and offers a desirable performance against temperature changes.

Finally, although the new flowsheet was designed based on optimizations in our previous work [15], re-optimization and use of other control structures are still recommended.

#### CONCLUSIONS

A new process flowsheet was proposed and developed for  $\text{CO}_2$ -CPU unit using one cold-box instead of two or three ones suggested in previous researches. This new flowsheet utilizes less equipment compared with other suggested one, which would result in decreased investment costs and its degree of freedom. Therefore, it is feasible and has a simpler control structure. The process control can be easily implemented using typical temperature, pressure, and level controllers. Accurate and narrow control of the temperature of the first separator is vital





*Fig. 5. Process response to  $\pm 5\%$  change in feed flowrate (green: increase; blue: decrease).*

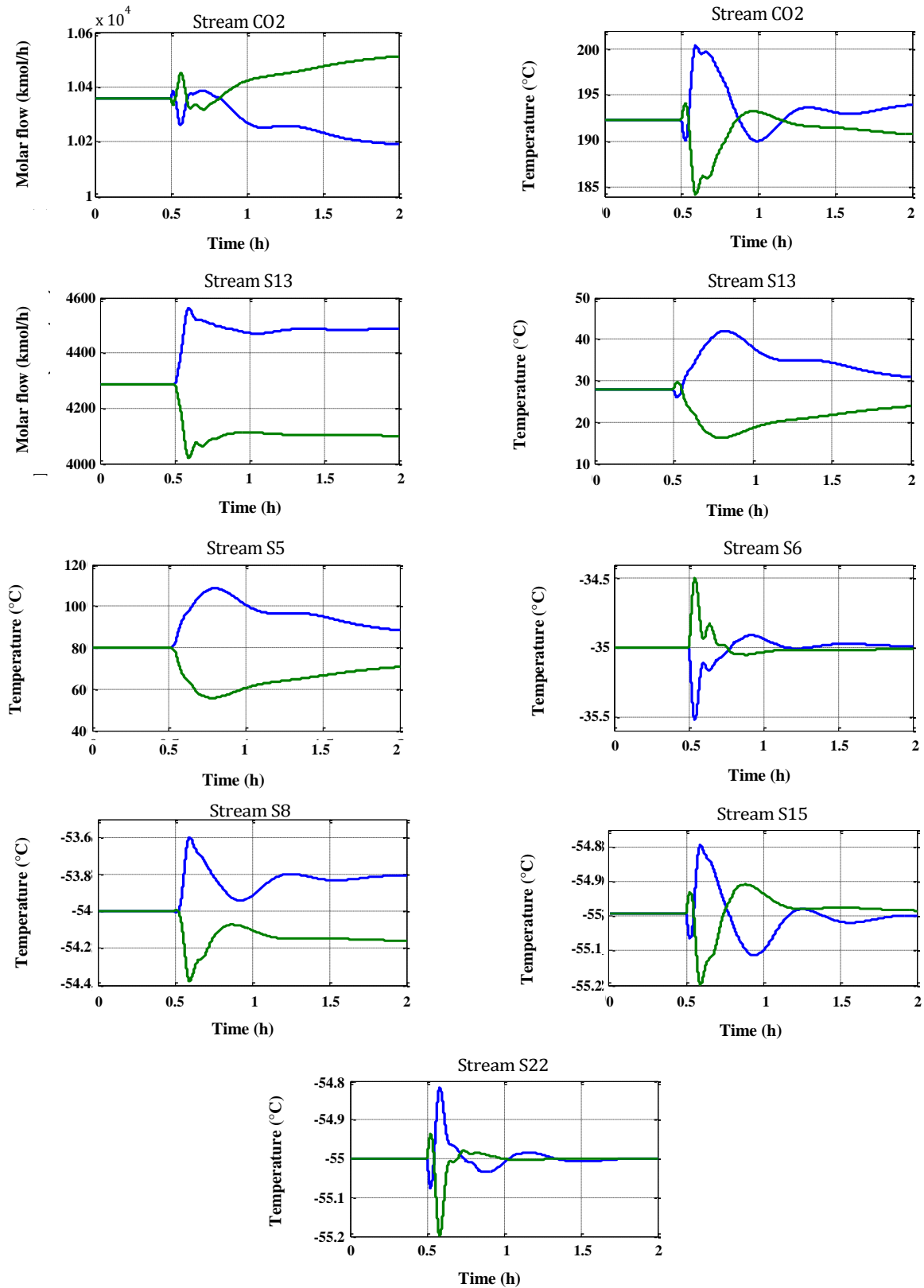


Fig. 6. Process response to  $\pm 5\%$  change in feed  $\text{CO}_2$  composition (green: increase; blue: decrease).

to process stability and prevention of controller saturation. Optimization of operating parameters will increase LMTD, significantly decreasing the required surface area compared with previous flowsheets. The reduction in the number of cold-boxes, and the improved energy integration, all contribute to cost reduction. Also, the proposed flowsheet yields a product of higher purity (96.7%) compared with previous flowsheets and provides a desirable recovery rate of 90%. The process responses to disturbances in feed flow rate and composition indicates that the system is nonlinear, thus, it is suggested that the controllers are required to be returned if the change in flowrate is high enough and permanent.

Received : Sep. 3, 2019 ; Accepted : Dec. 23, 2019

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