

Article

CO₂ Utilization via Integration of an Industrial Post-Combustion Capture Process with a Urea Plant: Process Modelling and Sensitivity Analysis

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Abstract: Carbon capture and utilization (CCU) may offer a response to climate change mitigation from major industrial emitters. CCU can turn waste CO₂ emissions into valuable products such as chemicals and fuels. Consequently, attention has been paid to petrochemical industries as one of the best options for CCU. The largest industrial CO₂ removal monoethanol amine-based plant in Iran has been simulated with the aid of a chemical process simulator, i.e., Aspen HYSYS[®] v.10. The thermodynamic properties are calculated with the acid gas property package models, which are available in Aspen HYSYS[®]. The results of simulation are validated by the actual data provided by Kermanshah Petrochemical Industries Co. Results show that there is a good agreement between simulated results and real performance of the plant under different operational conditions. The main parameters such as capture efficiency in percent, the heat consumption in MJ/kg CO₂ removed, and the working capacity of the plant are calculated as a function of inlet pressure and temperature of absorber column. The best case occurred at the approximate temperature of 40 to 42 °C and atmospheric pressure with CO₂ removal of 80.8 to 81.2%; working capacity of 0.232 to 0.233; and heat consumption of 4.78 MJ/kg CO₂.

Keywords: post-combustion; CO₂ capture; CO₂ utilization; heat consumption; capture efficiency; monoethanol amine

1. Introduction

Inasmuch as the effects of global warming have become more evident, efforts are being executed to mitigate greenhouse gases emissions, mainly CO₂ [1]. The concept of capturing carbon dioxide from power station, refinery, and factory exhaust has long been hailed as crucial in mitigating the climate disaster [2]. Carbon capture and storage (CCS) is a set of technologies which can meaningfully contribute to the reduction in CO₂ emissions; and post-combustion CO₂ capture (PCC) is the more developed technology to be integrated with thermal power plants and the other energy-intensive industries [3]. The technology has some drawbacks such as huge regeneration energy consumption, amine solvent degradation, and equipment corrosion [4]. However, regeneration energy consumption is considered as the main challenge, as it requires more than 70% of the total operating costs of a CO₂ capture plant. Therefore, the reduction in heat duty of regeneration has to be the main priority for more progress in amine-based CO₂ capture technology [5]. There are two commercial-scale power plants in which amine-based PCC using amines are being operated today, namely at Boundary Dam

in Canada [6] and at the Petra Nova project at the Parish Power Station in the US [7]. Such plants demonstrate the technical viability of CCS technology at the commercial scale.

The possibility of eliminating CO₂ from industrial and power generation plants has increased because of environmental issues [8]. However, the former concept of CCS is no longer satisfactory, and it has already been modified to carbon capture and utilization (CCU), including utilization of carbon dioxide [9]. Utilization of carbon dioxide can be divided into two main categories. In the former part, CO₂ has confident utilization with no conversion, particularly in the supercritical state such as enhanced oil recovery with CO₂ flooding or physical solvent applications [10]. Indeed, injection of CO₂ into an oil reservoir increases the production because of high mutual dissolving capability of supercritical CO₂ and hydrophobicity of oil [11]. In the latter part, the CO₂ moiety is rehabilitated via carboxylation or reduction avenues to chemicals and fuels. In fact, utilizing CO₂ as a feasible feedstock for the chemical industry has been the forethought of visionary researchers, and pioneering studies have already made significant achievements towards converting CO₂ to C1 building block chemicals [12]. Annually, about 130 Mt of CO₂ are used to manufacture urea, salicylic acid, cyclic carbonates, and polycarbonates, among which the urea process consumes most of the CO₂ industrially [13]. Universally, great amounts of ammonia are altered to urea, which is in turn mostly utilized as a fertilizer used to increase crop yields and food production. A number of CCU plants employed for urea production are presented in Table 1. Except in the case of Iran, all of the plants were constructed by Mitsubishi Heavy Industries Engineering.

Table 1. Commercialized carbon capture and utilization (CCU) plant for urea production.

Operation Year	Country	Source of Flue Gas	Capacity (TCO ₂ /D)
1999	Malaysia	Natural gas	210
2006	India	Natural gas	450
2006	India	Natural gas	450
2009	India	Natural gas	450
2009	Bahrain	Natural gas	450
2010	UAE	Natural gas	400
2010	Vietnam	Natural gas	240
2011	Pakistan	Natural gas	340
2012	India	Natural gas	450
2013	Iran	Natural gas	132

Aspen HYSYS[®] and Aspen Plus[®] have been employed for simulating a variety of PCC plants. Pouladi et al. [14] used Aspen HYSYS[®] software to simulate a commercial-scale CO₂ capture plant. They employed the amine package equation out of the fluid property packages to calculate the thermodynamic properties of the process. Husebye et al. [15] simulated a capture process by Aspen Plus[®] software using the amines property package in order to obtain better understanding of varying CO₂ concentrations existing in flue gas. Moioli and Pellergrini [16] simulated a capture plant with Aspen Plus[®] and validated the results of simulation by comparing them with experimental data of a pilot plant for the purification of flue gas from a power plant with the aid of monoethanolamine (MEA) washing. Dubois et al. [17] simulated various configuration of PCC, i.e., solvent split flow, rich solvent recycle, and lean/rich vapor compression applied to the exhaust gas coming from the Norcem Brevik cement plant.

Gervasi et al. [18] simulated a PCC plant by Aspen HYSYS[®] using the amine package and Kent–Eisenberg model. The simulated model by MEA 30% obtained a heat consumption of 3.71 GJ/tCO₂ with a capture rate of 85% and was validated with the available literature results considering the St. Marys cement plant in Canada as a case study.

Xue et al. [19] developed a steady-state process model of MEA 30% PCC in Pro/II software and validated the obtained results with the experimental data. They evaluated the system using a variety

of data for different days. In the best case, they obtained heat consumption of 3.71 GJ/tCO₂ with a capture rate of 79.9%.

Øi [20] simulated a natural gas fired power plant integrated with an MEA-based PCC by Aspen HYSYS® with the aid of the Peng–Robinson and amines property package models. The required energy for the regeneration of solvent was obtained at 3.7 MJ/kg CO₂ with respect to CO₂ removal of 85%.

Adeosun et al. [21] simulated a PCC unit for a 600 MWe conventional coal-fired power plant using the equilibrium-based Aspen Plus® for evaluation of amine-blend solvent. They evaluated 30 wt% total mixtures of 2-amino-2-methyl-1-propanol (AMP) and methyl diethanolamine (MDEA) using diethanolamine (DEA) and MEA as activating agents. The mixture of 5 wt% DEA and 25 wt% AMP performed better than the other mixtures by obtaining heat consumption of 3.03 GJ/tCO₂.

Li et al. [22] simulated a CO₂ recovery process using a rate-based model available in Aspen Plus® and validated with the results from the PCC pilot plant trials located at the Tarong power station in Queensland, Australia. After process enhancements, the optimal operating conditions were selected, and heat consumption of 3.6 MJ/kg CO₂ was obtained using 35% MEA.

Morales-Mora et al. [23] simulated a PCC plant with the capacity of 1075 t/day using Aspen HYSYS® version 9. The energy that was required for CO₂ separation from the exhaust gas and compression was 4.360 MJ/kg CO₂, consisting of 0.0024 kg MEA/t CO₂ with a capture efficiency of 95.4% and CO₂ stream purity of 95%.

The majority of CO₂ recovery plants integrated to the urea plants have been established by Mitsubishi Heavy Industries Engineering. However, the case study was constructed by a domestic company, and simulation and performance validation are considered as novelties on the knowledge. As the number of CO₂ capture plants has increased in recent years in Iran, the accurate design of such plants is an important issue. This research aimed to simulate the industrial CCU plant of Kermanshah Petrochemical Industries Co. using Aspen HYSYS®, based on real data, and validate the results by comparing them with the output of the model. The simulation results presented the balance of mass and energy for the whole process, detailed information of units and streams, and energy consumption, as well as the equipment size of main units. A sensitivity analysis of main parameters such as heat consumption, capture efficiency, and working capacity was then employed to evaluate the simulated plant.

2. Case Study and Process Description

Iran is considered among the most CO₂ emitting countries over the past decades. The large amount of fossil resources, and deficiency in planning of integrated energy policies in conjunction with neglect of performance enhancement in energy intensive plants have vastly worsened the climate state. Power plants, refineries, and petrochemicals are the main sources of the emissions. Petrochemicals can be the first priority for implementing and integrating of CCU plants because of the possibility of increasing their product capacity and producing new products. Kermanshah Petrochemical Industries Co., located in the western part of the country, is engaged in manufacturing and selling agricultural fertilizers and chemicals. Its product offerings include urea fertilizer, liquid ammonia, and liquid nitrogen. At the urea production facilities, ammonia is always in excess. This means that by recovering CO₂ from the stack of the ammonia reformer, the stored ammonia can be converted to urea, thus boosting production capacity without the need to invest extensively in major equipment such as reformers and reactors. In this complex, a CO₂ recovery unit for the capture of carbon dioxide from the stack of the primary reformer of the ammonia plant was established. The project at Kermanshah Petrochemical Industries Co. (KPIC) was licensed, designed, and built by Shahrekord Carbon Dioxide (SCD) to recover 132 metric tons per day (MTPD) of CO₂ from the stack of the ammonia reformer. The project was initiated in early 2013, and within 20 months the plant was operating. Utilization of this technology has enabled KPIC to lower its natural gas consumption by 21.1 million normal cubic meters (M Nm³) of natural gas per year, and more than 40,000 tons of CO₂ emissions per annum have been avoided. As a

result of this project, the production capacity has been boosted by 5%, without any further investment beyond the execution of the plant.

Industrially, urea is primarily produced by the hydration of calcium cyanamide; nonetheless, ammonia accessibility has resulted in the expansion of ammonia/carbon dioxide technology. There are two steps in which ammonia and CO_2 react to form ammonium carbamate, which is then dehydrated to urea. In natural gas-based ammonia and urea plants, CO_2 is recovered from the reformer burner flue gas and used for urea synthesis. Figure 1 shows block flow diagram for the existing plants in Kermanshah Petrochemical Industries Co. The operational data used for simulation of the plant was gained from the CO_2 recovery plant of KPIC. The plant is located in Bisotun and is shown in Figure 2a.

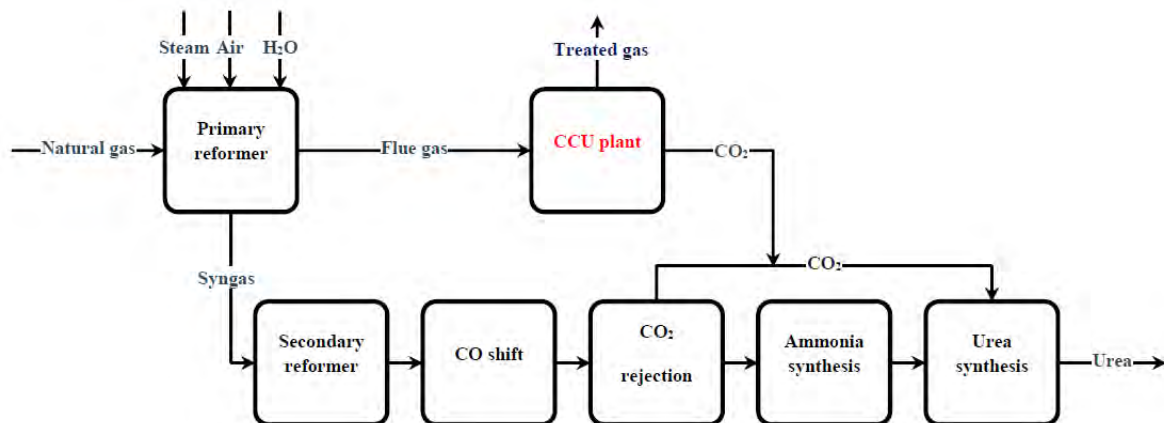


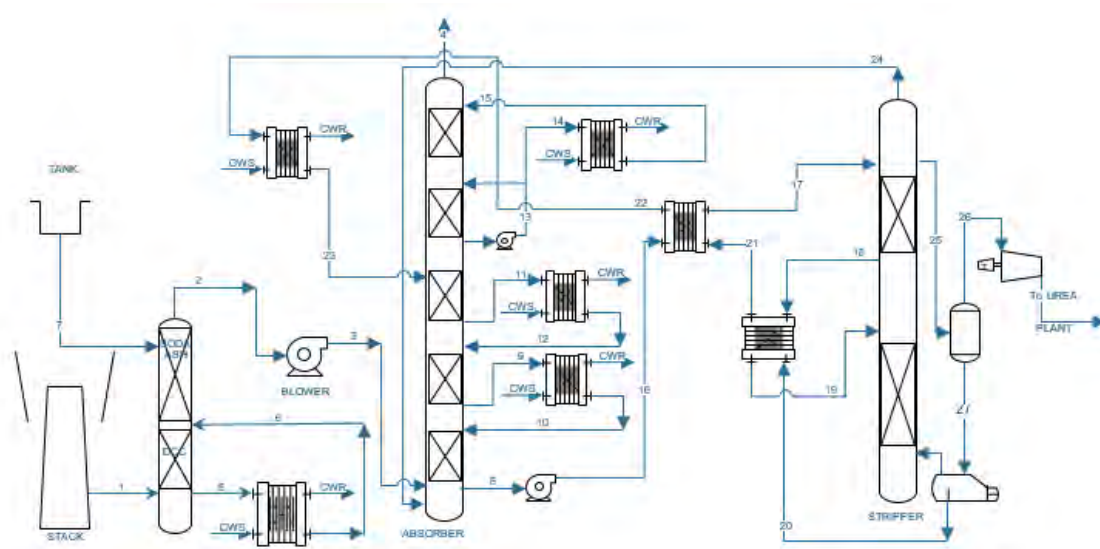
Figure 1. Block flow diagram of the case study.

The flue gas stream at $178\text{ }^{\circ}\text{C}$ is delivered to the bottom of soda ash wash and direct contact tower; and its temperature declines up to $42\text{ }^{\circ}\text{C}$, while the water in the flue gas is drained. The flue gas is next connected to the blower, by which the required pressure to pass through absorber tower is provided. The flue gas at the temperature of $47\text{ }^{\circ}\text{C}$ enters into the absorber tower. The absorber is made up of five segments, filled with the random packing. The top sections of the column are the cooling and washing sections with a height of 2 m for each segment, and the rest are absorber packed sections with a height of 3 m for each segment. The overall height of the packed column is equal to 13 m . The contact-cooler in the washing section is mainly to prevent water equilibrium in conjunction with amine loss in the process. There are two intercoolers between the first and second sections as well as second and third sections of the absorption column used for surging in the absorption rate of CO_2 . Going up the column, the exhaust gas contacts the 30% MEA solution, and its CO_2 is absorbed by the solution. Rich MEA solution departs from the bottom of the absorber column and enters into the pump. The rest of the exhaust gas, which mainly comprises nitrogen, steam, and probably some unabsorbed CO_2 , is purged to the atmosphere. Rich MEA pumps into the first stage of the main heat exchanger. Exchanging heat with the other side of the heat exchanger, containing the lean MEA solution from the reboiler, its temperature increases from $52\text{ }^{\circ}\text{C}$ to $87\text{ }^{\circ}\text{C}$. Rich MEA is then sent to the top of the stripper tower and contacts the steam provided in the reboiler. The required energy for regeneration of the solvent is supplied by steam of the reformer in the ammonia plant. The steam latent heat acquires the required energy for CO_2 separation from the rich solution. The CO_2 gas, released from the top of the stripper, contains steam, and a little MEA enters into the condenser. The steam is condensed and the water is recycled to the stripper tower, and CO_2 enters the compressor of the urea plant.

The composition of flue gas extracted from the stack of the ammonia plant is presented in Table 2, and the actual operation conditions of the plant are also presented in Table 3. Figure 2b shows the schematic of the actual plant, which is drawn based on the process flow diagram.



(a)



(b)

Figure 2. (a) The industrial CCU plant. (b) Schematic of the plant based on the process flow diagram.

Table 2. Composition of the flue gas stream.

Substance	Mole Fraction
N ₂	0.724
CO ₂	0.0678
O ₂	0.0326
H ₂ O	0.1756

Table 3. Operation conditions of the plant.

Stream No.	Mass Flow Rate (kg/h)	Temperature	Pressure
		(°C)	(bar)
1	63,297	178	0.8776
2	59,571	42	0.8621
3	59,629	47	0.9007
4	53,968	43	0.8776
5	272,256	56	2.5
6	272,256	40	2
7	115,500	42	2.5
8	121,275	51.7	1.4
9	119,053	51.7	2.4
10	119,053	42	2.2
17	122,483	87	4.4
18	121,372	90	3.8
19	121,372	116	3.6
20	115,612	126	2.35
21	115,612	100	2.15
22	115,612	70	1.95
23	115,612	48	1.75
24	57.47	60	2.2
25	6366	86	2.2
26	5604	43	2
27	762	43	2

3. Simulation and Thermodynamic Framework

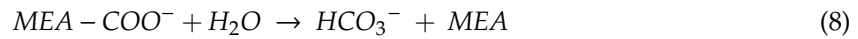
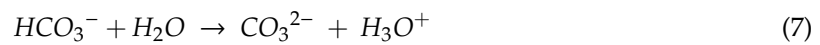
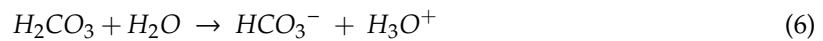
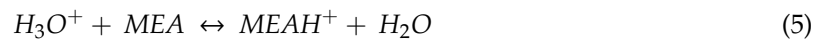
The process model developed by Aspen HYSYS® v.10 software using the acid gas package was used. An important advantage with using a process simulation program is the available models employed for thermodynamic properties. Aspen HYSYS® has an acid gas package in which the Peng–Robinson equation of state for the vapor phase and the electrolyte non-random two-liquid (eNRTL) activity coefficient model for electrolyte thermodynamics in the liquid phase are used [17].

The acid gas package advanced by Aspen allows one to simulate the acid gas removal of CO₂ and H₂S. It comprises the physicochemical specifications of the acid gases, water, amines alone, e.g., MEA and PZ, along with several mixtures, e.g., MDEA + PZ. Additionally, it provides a rate-based calculation model as well as a makeup unit operation to make up losses in water and amine in the system. The package was developed on the basis of extensive investigation in the chemical absorption process, simulation of the rate-based process, and models of molecular thermodynamic for amine solutions. It comprises the eNRTL model parameters and other transport property model parameters recognized from regression of wide thermodynamic and physical property data for amine solutions [24–26].

CO₂–amine interactions are part of solvent chemistry. After the amine–CO₂–H₂O systems reach chemical and vapor–liquid equilibrium, the amine solutions contain multiple cations (AmH⁺), free molecules (Am), and anions (carbamate, HCO₃[−], CO₃^{2−}), instantaneously. These “ternary” systems contain 8–9 ionic or neutral species. Their concentrations are limited by mass balance and reaction equilibrium [4].

MEA overall reaction:





All of the equipment is herein simulated using the acid gas package except the soda ash wash-direct contact tower, which is simulated by eNRTL.

Table 4 shows the specification of the all equipment employed in the simulation for the industrial PCC plant. In the first section, the specification of the three columns' internal dimensions, including height, diameter, type, material, and dimension of each packed sections, are presented. In the second part, the specification of the compression equipment, including differential pressure, pressure ratio, adiabatic efficiency, and consumed power, are provided. In the last section, the specifications of the heat exchangers, i.e., intercoolers of absorber, MEA rich-lean heat exchangers (stage 1 and stage 2), reboiler, condenser, and the reset of heat exchangers, are provided.

Table 4. Specification of columns' internal, compression equipment and heat exchangers for the industrial post-combustion CO₂ capture (PCC) plant.

Column	Packed Sections	Packed Diameter (m)	Packed Height (m)	Packing Type, Material, and Dimension
DCC & Soda Ash	Soda Ash	3.8	3	Saddle, Ceramic, 3"
	DCC	3.8	3	Saddle, Ceramic, 3"
Absorber	Cooling	3.5	2	Pall, Plastic, 2"
	Washing	3.5	2	Pall, Plastic, 2"
	Absorber 3rd	3.5	3	Pall, Plastic, 2"
	Absorber 2nd	3.5	3	Pall, Plastic, 2"
	Absorber 1st	3.5	3	Saddle, Ceramic, 3"
Stripper	Stripper 2nd	2.4	3	Pall, Ceramic, 2"
	Stripper 1st	2.4	7	Pall, Ceramic, 2"
Compression equipment	Pressure ratio	Adiabatic Efficiency	consumed power (kW)	ΔP (kPa)
Blower	1.045	75	89.6	3.86
Pump	3.36	75	13.76	330
Heat exchangers	Type	Hot inlet-outlet temperature (°C)	Cold inlet-outlet temperature (°C)	Base duty (kW)
E-1	Intercooler	52.6–40	20–25	4020
E-2	Cooler	92.8–48	20–25	4279
E-3	Intercooler	50.3–39	20–25	1100
E-4	Intercooler	52–41.1	20–25	1200
E-5	Intercooler	60.3–51.3	20–25	1000
E-6	Condenser	94.1–43.3	20–25	780
E-7	Process Exchanger	123.5–92.8	55–87	3680
E-8	Process Exchanger	127.2–123.5	114.2–115.5	440
E-9	Heater (reboiler)	153–133	45–127	7420

4. Results and Discussion

4.1. Model Validation

In order to validate the simulation results with the actual data, important parameters of the CCU plant were compared and are provided within Table 5. The simulation relative error was achieved using the following relationship:

$$\text{Relative error} = \left| \frac{i_{\text{actual}} - i_{\text{simulation}}}{i_{\text{actual}}} \right| \quad (9)$$

Table 5. Validation of simulated results with important parameters of the CCU plants.

Parameter	Actual Value	Values of Simulated Results	Error (%)
Heat consumption (kW)	7800	7420	4.80
Blower power (kW)	94.2	89.6	4.80
Pump (kW)	14.43	13.76	4.60
CO ₂ mass flow rate (kg/h)	5604	5657	0.90
CO ₂ composition (mol %)	95.5	95.5	0
Temperature of lean stream 20	126	127.2	0.95
Pressure of lean stream 20	2.35	2.35	0
MEA rate in lean amine (kmole/h)	547.9	549.1	0.22
CO ₂ rate in lean amine (kmole/h)	93.5	93.7	0.21
MEA content in lean amine (% mass)	28.95	29	0.17
CO ₂ content in lean amine (% mass)	3.56	3.57	0.28
Temperature of rich stream 8	52.5	54	2.86
Temperature of rich stream 17	87	87	0
Temperature of semi lean stream 19	116	115	0.86
MEA content in Rich amine (% mass)	27.6115	27.6	0.04
CO ₂ content in Rich amine (% mass)	7.9654	7.96	0.06
MEA rate in rich amine (kmole/h)	548.234	547.86	0.06
CO ₂ rate in rich amine (kmole/h)	219.5	219.27	0.10

It is necessary to say that there was no way to measure some parameters such as compositions and flow rates of elements in some streams. Consequently, the results of simulation for some parameters were validated with the design parameters provided in data sheet of components and the process flow diagram. Figure 3 shows a schematic of the simulated plant by Aspen HYSYS®. The number of errors shows that there was good agreement between simulated results and real performance of the plant under different operational conditions. The amount of heat consumption in MJ/kg CO₂ removed in the simulated case was 380 kW less than in the actual one with a percent error of 4.8%. Additionally, the amounts of required power for the blower and pump in the simulated case were 4.6 kW and 0.67 kW less, respectively, than the actual data with percent error of 4.8% and 4.6%, respectively. These three parameters had the highest percent errors; however, the magnitude of errors was less than 5% between actual and simulated data, through which the accuracy of the simulation was proven. The rich stream temperature in the outlet of absorber column in the simulated case was 1.5 more than the actual data with percent error of 2.86%. The amount of CO₂ mass flow rate in the simulated case was equal to 5657 (kg/h), while the actual data was equal to 5604 (kg/h). The amount of the percent error for CO₂ mass flow rate was equal to 0.94%, which is a rational value for this important parameter. The percent errors for the other parameters provided within the Table 5 were less than one percent. For clarification, the specification of inlet and outlet streams are provided within the flowsheet of simulation in Figure 3.

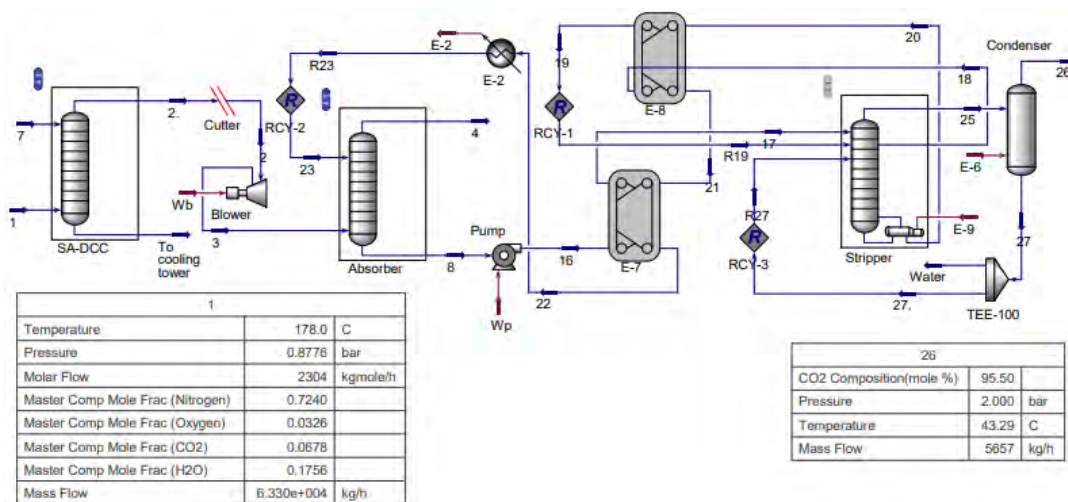


Figure 3. Schematic of the simulated plant by Aspen HYSYS®.

4.2. Sensitivity Analysis

Sensitivity calculations were employed using variation in the main operational parameters, i.e., inlet pressure and temperature of absorber column, to evaluate the main parameters such as capture efficiency in percent, the heat consumption in MJ/kg CO₂ removed, and the working capacity of the plant.

Simulation results, shown in Figure 4, were employed to depict the effect of inlet temperature of flue gas entered to the absorber versus capture efficiency and heat consumption. Capture efficiency increased with temperature from 36 to 42 °C; it then had a constant trend. The best condition occurred at approximately 42 °C for both heat consumption at 4.78 MJ/kg CO₂ and capture efficiency at 80.8%.

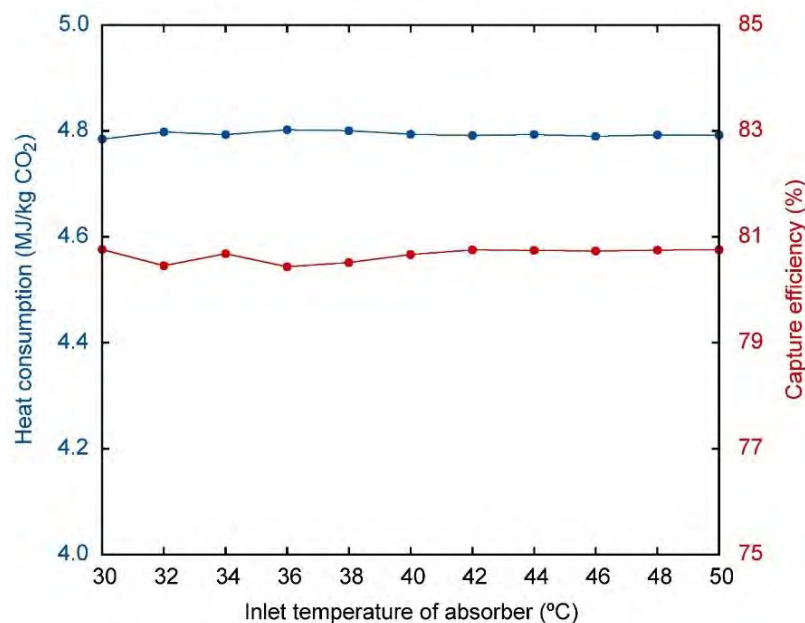


Figure 4. Variation of inlet temperature versus heat consumption and capture efficiency.

Figure 5 shows the variation of the inlet pressure of absorber column versus the heat consumption and the capture efficiency. The best condition approximately occurred at atmospheric pressure, i.e., 1 bar, for both the heat consumption and the capture efficiency. The trend of heat consumption was constant, and there were smooth fluctuations in the capture efficiency after the pressure of 1.02 bar. The highest

amount of capture efficiency was equal to 81.2%, and the lowest amount of heat consumption was equal to 4.78 MJ/kg CO₂, which both occurred at atmospheric pressure.

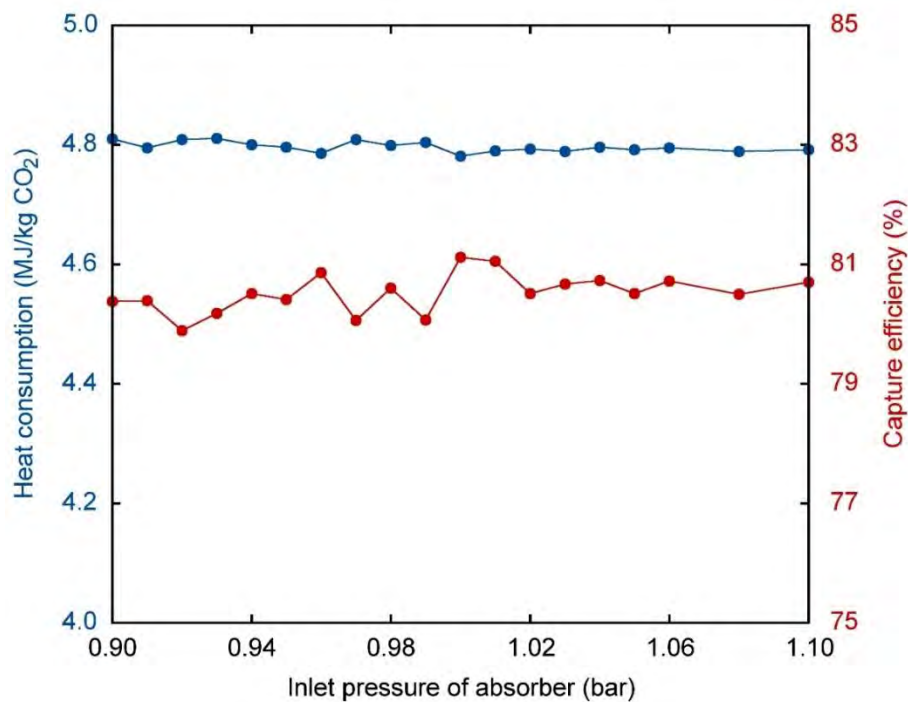


Figure 5. Variation of inlet pressure versus heat consumption and capture efficiency.

Figure 6 show variation of working capacity versus inlet pressure and temperature of the absorber. The best condition for the working capacity approximately occurred at atmospheric pressure and a temperature between 40 and 42 °C. The highest amount of working capacity with respect to inlet pressure of absorber occurred at atmospheric pressure with the amount of 0.233, while the value with respect to the inlet temperature of absorber occurred at 40 °C with the amount of 0.232, although this value was approximately the same at 41 and 42 °C. The working capacity decreased after these pressures and temperatures and had a constant trend with smooth fluctuations.

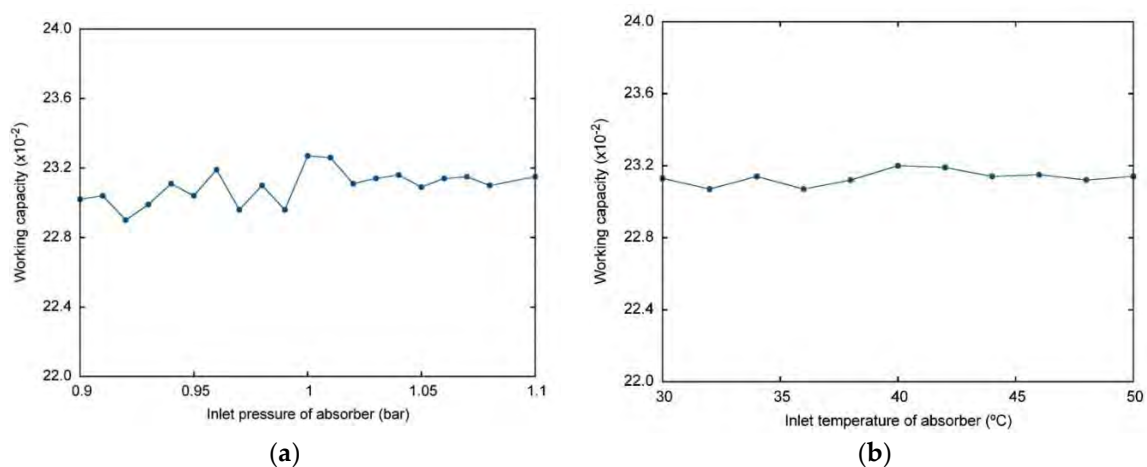


Figure 6. (a) Variation of working capacity versus inlet pressure of absorber. (b) Variation of working capacity versus inlet temperature of absorber.

5. Conclusions

A chemical process flowsheet simulator, i.e., Aspen HYSYS[®] v.10, was herein employed to simulate the largest industrial CCU plant in Iran. Acid gas property package models, which are available in Aspen HYSYS[®], were employed to calculate the thermodynamic properties. The simulation results were validated by the actual data provided by Kermanshah Petrochemical Industries Co. Good agreement was obtained between simulated results and real performance of the plant under diverse operational conditions. Sensitivity analysis of the main parameters, i.e., heat consumption, capture efficiency, and working capacity, was then conducted using various operational conditions to evaluate the simulated plant. Considering various temperatures in the inlet of the absorber column, the best condition occurred at approximately 42 °C, in which heat consumption and capture efficiency were equal to 4.78 MJ/kg CO₂ and 80.8%, respectively. Considering various pressures in the inlet of the absorber column, the best condition approximately occurred at atmospheric pressure in which heat consumption and capture efficiency were equal to 4.78 MJ/kg CO₂ and 81.2%, respectively. The highest amount of working capacity with respect to the aforementioned operational parameters was equal to 0.233. The high amount of heat consumption compared to the reported value in the literature is due to the oversized design of each pack of the column, the column diameter, and the heat transfer areas, as well as low-energy price of fossil fuel in Iran. This issue can also be justified when the plant increases its capacity to capture larger amounts of CO₂ for feeding the urea plant.

Author Contributions: R.S.: Conceptualization, Methodology, Software, Formal analysis, Validation, Visualization, Resources, Writing—Original Draft, Review & Editing. A.A.: Project administration, Funding acquisition, Supervision, Resources, Writing—review & Editing. R.G.: Project administration, Funding acquisition, Supervision, Resources, Writing—review & Editing. L.M.R.: Supervision, Conceptualization, Methodology, Writing—review & Editing. All authors have read and agreed to the published version of the manuscript.

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