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Research Article

Data Analysis in Green Industrial Processes with Modified Chemical Efficiency and Environmental Impact: Smart Urea Production and CO$_2$ Removal

Fatemeh Khandaghi [1,2,3] **and Shahoo Abdollahi**[3]

[1]*School of Chemical Engineering, College of Engineering, University of Tehran, Tehran, Iran*
[2]*School of Chemical Engineering, Iran University of Science and Technology (IUST), Tehran, Iran*
[3]*Research & Development, DanubTech Co., Tehran, Iran*

Correspondence should be addressed to Fatemeh Khandaghi; f_khandaghi@vu.iust.ac.ir

Agriculture can benefit from urea fertilizer because it contains a lot of nitrogen at a reasonable cost. Urea fertilizer can be stored easily and does not pose a fire hazard over time. Due to its acidifying properties, urea fertilizer is an ideal fertilizer for many plants. Input and feed of the urea unit are taken from the output of the ammonia unit (CO$_2$ and NH$_3$). Hence, in this study, two methods of CO$_2$ recovery from combustion gases and CO$_2$ recycling in ammonia units will be used to increase urea production to realize low-carbon and industrial systems (including green agriculture). CO$_2$ recovery also reduces environmental pollution, which is a very important factor in sustainable cities and societies. The results showed that CO reduction increases the overall efficiency compared to the data reported in the world for the same process, which is due to the reduction of CO input to methanize. Collecting information around the globe for constructing the same green system considering various conditions in each environment makes complicated situations in terms of how to design the process and the observed outcomes. However, we could find a new smart design to build the green system in our case study where it is completely acceptable compared to the same systems' outputs. The obtained results indicate that the temperature of the shift reactor can be brought closer to 365°C without reducing the selectivity of the catalysts, which in turn would increase the CO conversion rate, the CO$_2$ output, and the overall efficiency of the unit. Finally, it is shown that the rate of CO escape from shift reactors is decreased.

1. Introduction

Nowadays, green industry concept is becoming a major challenge in modern societies [1]. In this regard, the human population is expected to reach 9,500,000 by 2050. It means food shortages occur as per capita demand almost doubles. Also, the number of acres of agricultural land is declining as a result of economic growth, residential development, and climate change [2].

A large number of fertilizers especially nitrogen fertilizers like urea (NH$_2$CONH$_2$) are needed to have a modern society as well as a green industry [3]. As it is obvious, urea is high in nitrogen and nitrogen is a vital nutrient source for plant product development. In fact, chlorophyll, proteins, and protein-carrying compounds are all made from nitrogen [4]. Compared to other nutrients, nitrogen is a primary and continuous nutrient for plant growth [5, 6]. Nitrogen contributes initially to rapid growth of roots and leaves and chlorophyll production and also increases biomass accumulation and yields [7, 8]. Agricultural products need nitrogen during critical growth stages, which is usually provided by conventional urea fertilizers [9, 10].

In order to achieve sustainable agriculture, it is essential to use pesticides and chemical fertilizers in the minimum amount and optimally [9]. Globally, about 200 million tons of urea fertilizer are produced each year [11, 12]. Since urea can be combined with other solid fertilizers, this fertilizer has become the most widely used fertilizer in the world

[13–15]. In this way, we can further the development of the green industry through its optimal production of fertilizer [5].

Among all solid nitrogen fertilizers, urea contains the most nitrogen (46.7%) [16] and it is hydrolyzed to ammonia and carbon dioxide in the soil. Bacteria in the soil oxidize ammonia produced in this process into nitrate, which can then be absorbed by plants. Often, urea is used in multicomponent formulations of solid fertilizers. Since urea is highly soluble in water, it is also very suitable for use in fertilizer solutions [17].

Urea is produced industrially by the reaction of ammonia and carbon dioxide at high pressures (13 to 30 MPa) and high temperatures (170 to 200°C) [18]. Interestingly, urea was first chemically produced by Farben in 1920 [19]. Urea fertilizer has been produced since the early 1950s. The advantages of urea have led many researchers to investigate how to improve conversion rates and the quality of products [20].

Nowadays, several technologies including chemical solvent adsorption, biological stabilization, membrane separation, and hydrate-based separation are used to recover CO_2.

With the growing population of the world and a need to meet its food needs, agriculture has become increasingly challenging despite the limited amount of land available to grow food. The total ammonia emissions from agriculture can be attributed to mineral nitrogen fertilization (especially urea). To reduce ammonia emissions and increase fertilizer efficiency, it is crucial to minimize ammonia evaporation. As such, it is necessary to reduce the negative impact of human pressure on the environment, including air pollution, soil erosion, and water pollution [21, 22]. The proportion of ammonia to carbon dioxide produced in ammonia production units is higher than the ratio required in its downstream unit (urea unit) due to the lightness of the feed gas. Accordingly, in this study, the CO_2 shortage will be compensated by the CO_2 recovery from flue gas or CO_2 absorption tower was increased in order to increase the urea production capacity. Green systems can be categorized into different parts. We will have green energy, for example, when we find ways to reduce energy consumption. By requiring fewer materials to produce something, we have green materials as well. Air pollution reduction is also one of the most important uses of green systems. Hence, the proposed framework has the potential to achieve green industrial policy. In fact, the proposed framework benefits from economic, social, and environmental aspects.

Considering the importance of this topic, the purpose of this article is as follows:

(I) CO_2 recycling in ammonia unit and combustion gases

(II) Investigating how to reduce air pollution in petroleum units

(III) Increasing the production of urea fertilizer

Gathering the different data for the green systems is a challenging issue when we attend to the outputs and compli-cated processes in each scenario. Fortunately, we could find a smart solution to make the green system in our case study where it is considerable in comparison to other similar systems' outputs.

2. Materials and Methods

In order to facilitate readers, ammonia and urea production procedures are described in Sections 2.1 and 2.2, respectively. Moreover, Section 2.3 depicts the reaction of CO_2 with aqueous ammonia and Section 2.4 describes CO_2 hydration in an aqueous solution, while Section 2.5 demonstrates how to increase the amount of urea. It should be noted that this research was experimental, and data were collected from experiments. A data mining approach is then used to preprocess the collected data. Statistical methods are used to determine the applicability of the proposed framework. Our entire process was carried out on a laptop with the following configuration:

(i) Core i7

(ii) 8 GB ram DDR4

(iii) NVIDIA graphic with 2 GB ram

2.1. Ammonia Production Process. Ammonia was first discovered in the 8th century by Jabir Ibn Hayyan as salt [23]. Valentinus Basilius investigated that ammonia can be obtained by alkaline reactions on ammonia salts in the 15th century [24]. The development of new technology and a need for nitrogen and nitric acid fertilizers as the basis for explosives led scientists to reformulate these compounds from oil and gas [25]. Various industries and agriculture have developed in different countries as a result of ammonia production, including agriculture in Iran, which is now a major market [26, 27]. Nitrogen (ambient air) and hydrogen (hydrocarbons or water electrolysis) are combined to produce ammonia. Equation (1) depicts the general reaction for the production of ammonia [28]. Furthermore, Figure 1 illustrates the schematic of the ammonia production process.

$$N_2 + 3H_2 \longrightarrow 2NH_3 \Delta H700 = -52.5 \, kJ/mol \qquad (1)$$
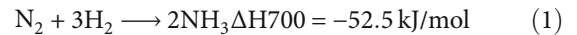
2.2. Urea Production Process. According to Equation (2), urea is formed when the mixture of ammonia liquid and carbon dioxide gas is pressured at 143 kg/cm^2 and heated to about 170-180°C. It should be noted that this process is calorific and rapid, converting ammonia and carbon dioxide into liquid ammonium carbamate (NH_2COONH_4), while urea (NH_2CONH_2) and water are extracted in a slow interaction that is depicted in Equation (3). Consequently, it is essential to provide the conditions in this unit for performing the above two reactions.

$$2NH_3 + CO_2 \leftrightarrow NH_2COONH_4 \qquad (2)$$

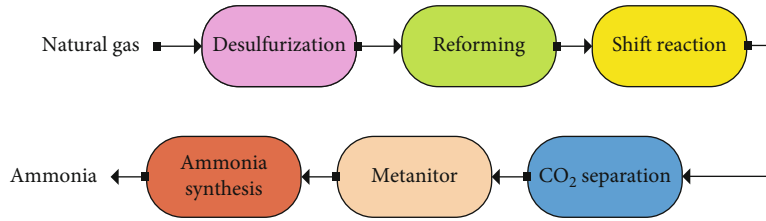$$NH_2COONH_4 \leftrightarrow NH_2CONH_2 + H_2O \qquad (3)$$
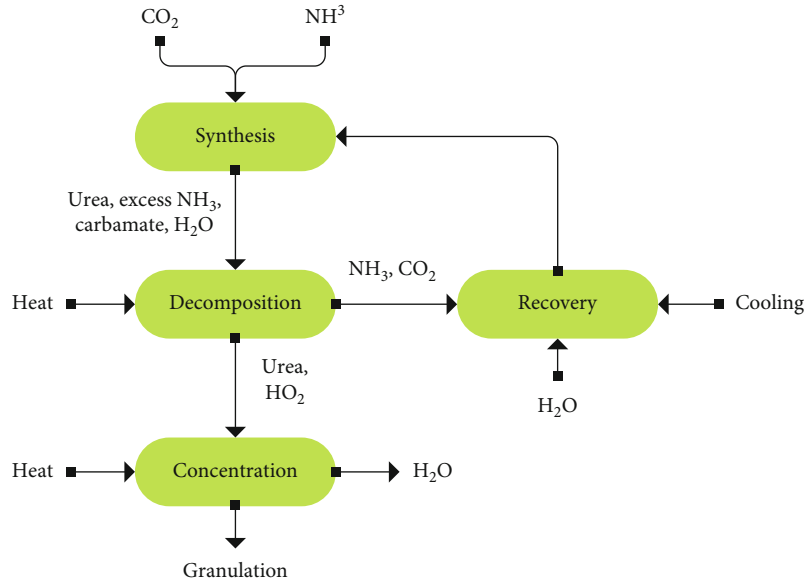
FIGURE 1: Schematic of the ammonia production process.
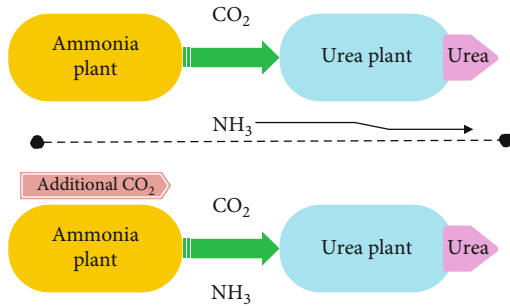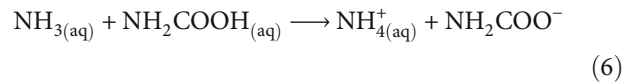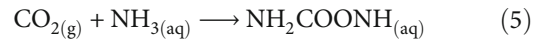

FIGURE 2: Urea production unit flowchart.

FIGURE 3: NH_3/CO_2 production ratio.

The urea unit receives ammonia and carbon dioxide from the ammonia unit. Pumps dispense liquid ammonia under a pressure of $160 \, kg/cm^2$, and compressors compress carbon dioxide under a pressure of $146 \, kg/cm^2$ [29]. Then, these two streams enter the unit that synthesizes urea. This unit must have the potential to operate at these temperatures and pressures. During the reaction of ammonia and carbon dioxide in a carbamate maker, high-pressure fuels form ammonium carbamate. The reaction is very fast and exothermic, and the generated heat is used to create low-pressure steam ($4.5 \, kg/cm^2$). Steam is used in several parts of the unit that the ammonium carbamate is then converted to urea and water. The urea production process is shown in Figure 2.

2.3. The Reaction of CO_2 with Aqueous Ammonia.

Carbon dioxide and ammonia react mainly in the liquid phase of the gas-liquid interface. Furthermore, Equation (4) illustrates this chemical reaction.

$$CO_{2(g)} + 2NH_{3(aq)} \longrightarrow NH_2COONH_{4(aq)} \tag{4}$$
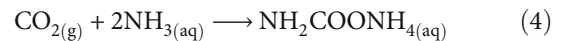
As a matter of fact, Equations (5) and (6) consist of two steps as below.

$$CO_{2(g)} + NH_{3(aq)} \longrightarrow NH_2COONH_{(aq)} \tag{5}$$

$$NH_{3(aq)} + NH_2COOH_{(aq)} \longrightarrow NH_{4(aq)}^+ + NH_2COO^- \tag{6}$$

NH_2COONH_4 is then decomposed in solution to produce free ammonia

$$H_2O + NH_2COO^- \leftrightarrow NH_3 + HCO_3^- \tag{7}$$

$$NH_3 + H_2O \leftrightarrow NH_4 + OH^- \tag{8}$$
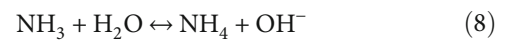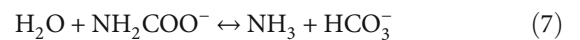
Equation (5) is very swift and irreversible, whereas Equation (6) is momentary and sudden. Also, Equation (7) is relatively slow and does not affect the absorption intensity directly. Therefore, the reaction between aqueous ammonia solution and carbon dioxide is primarily controlled by
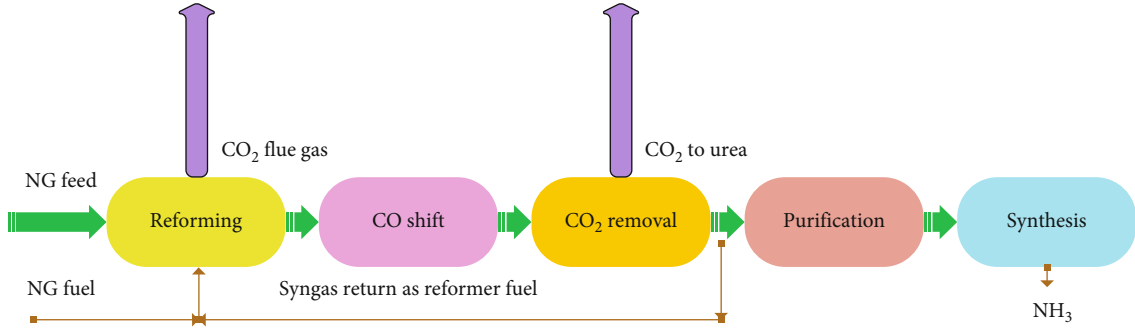
FIGURE 4: Increasing CO_2 production by returning synthesized gas.
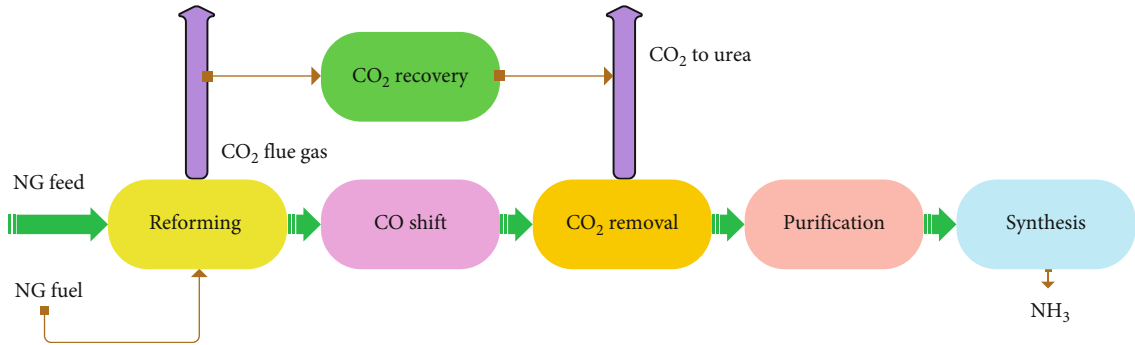
FIGURE 5: CO_2 recovery from combustion gases and flue.

TABLE 1: Comparison of two methods of CO_2 production.

	CO_2 recovery from combustion gases	CO_2 obtained from CO_2 recycling in ammonia unit
New equipment	✓	×
Investment	High	Low
Cost	High	High

Equation (5). CO_2 and NH_3 are, respectively, quadratic and first-degree reactions with a temperature of -257.85°C; the constant value of velocity k_2 is about 300 (L/mol·s).

2.4. CO_2 Hydration in Aqueous Solution. Ammonia solution has very weak alkalinity. As a result, immersion in the aqueous solution of CO_2 will occur in the liquid phase

$$CO_2 + H_2O \leftrightarrow HCO_3^- + H^+ \tag{9}$$

$$CO_2 + OH^- \leftrightarrow HCO_3^- \tag{10}$$
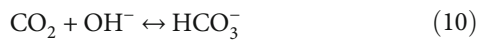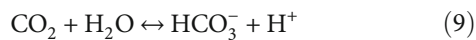
At 24.85°C the contribution of Equations (9) and (10) to the total reaction rate is very small, since $k = 0.026\,s^{-1}$. Therefore, they can usually be ignored. However, at 20°C, the value of k_{OH^-} is 5747.9 m^3/kmol.

2.5. Increasing the Capacity of Urea Production Unit. The output of the ammonia unit (CO_2 and NH_3) constitutes the input and feed to the urea unit. There is often excess ammonia in urea units that cannot be converted into urea due to a lack of CO_2. In fact, the ratio of ammonia and

CO_2 production is not independent of each other. The ideal ratio of CO_2 to NH_3 in the production of ammonia from pure methane (CH_4), air, and water is 1/14 (t/t) [30]. However, depending on the components of the natural gas and the losses during the process, this value can be lower or higher in the actual process. In contrast to the approximate amount, one unit of urea consumes more carbon dioxide and ammonia [31–33]. As mentioned, there is always extra ammonia; hence, the major challenge is insufficient CO_2 production. It means that additional CO_2 to utilize all of the ammonia available for urea production is required (Figure 3).

As follows, this paper suggests two methods for preparing the needed CO_2.

2.5.1. CO_2 Obtained from CO_2 Recycling in Ammonia Unit. Typically, the CO_2 feed for urea production is obtained by separating CO_2 from the synthetic gases in the CO_2 separation section [34]. Hence, by transferring more synthetic gases through this unit, the amount of CO_2 in this area can be increased. Therefore, the low output current from the CO_2 separation unit, which contains additional synthesis gas, is not required for ammonia production and is ignored and sent to the reformer, where it is used as a gaseous fuel. In this process, the gaseous fuel consumption per unit increases, resulting in higher throughput and therefore more work in areas such as desulfurization, reforming, and heat loss recovery. Meanwhile, the natural gas is returned to the reformer as fuel, reducing the consumption of natural gas. Natural gas consumption is still expected to increase as it is illustrated in Figure 4.

TABLE 2: Results of the applied changes.

Temperature	Description	Time (h)					
		9:40	9:50	9:55	11:00	11:20	13:00
AT-10048	Process gas to T-2001	0.413	0.4143	0.415	0.3906	0.3914	0.3955
FQI-2002	T-2002 over head	58046	58112	58279	58264	58380	58318
TI-1085	E-2003 inlet	126.89	126.89	126.94	127.14	127.34	127.17
TI-2011	CO_2 absorber over head	43.43	43.45	43.45	43.74	43.76	43.34
TI-2013	T-2003 over head	86.62	86.57	86.60	87.36	87.46	87.50
TI-2019	T-2002 over head	45.74	45.78	45.17	46.24	46.42	46.34
TI-2003	Co_2 stripper bottom	121.45	121.53	121.55	121.81	121.88	121.83
TI-1084	E-2001 PG outlet	165.16	165.28	165.79	166.78	165.11	165.72
TI-1082	BFW to E-1012	173.53	173.57	173.56	173.60	173.80	173.66
TI-2014	Lean soln pump disch	42.7	42.74	42.79	43.12	43.18	43.23
TI-2017	AMDEA semilean solution	83.08	83.04	83.01	83.96	84.11	84.26
TI-2012	T-2001 liquid outlet	87.62	87.64	87.70	88.53	88.63	88.70
TI-1087	D-2001 inlet	61	61.14	61.24	61.15	61.50	61.38
TI-1088	Reboiler circulation	112.11	112.14	112.06	112.41	112.60	112.60

2.5.2. CO_2 Recovery from Combustion Gases. As shown in Figure 5, CO_2 recovery unit is connected to the exhaust gases from the reformer. The combustion gases from reformers and boilers have a high carbon dioxide content making them another potential source of CO_2 for urea production units. The combustion gases of ammonia unit reformer furnaces contain 12% carbon dioxide. By separating CO_2 from this smoke and providing it to the downstream unit, which is urea, we can prevent its release into the atmosphere and air pollution.

During this process, the flue gas passes through a large tower, called an absorber tower. As a result of the adsorption process, exhaust gases are exposed to adsorbent fluid (amines dissolved in water), which creates a weak chemical bond between carbon dioxide and the amines. This carbon dioxide is then transferred to a tower known as a stripper, where the solvent is heated and CO_2 is separated from amines. Amines are then reused to reabsorb CO_2. The remaining ammonia in the CO_2-free output stream is removed in the wash tower. Additionally, fans and pumps are used to compensate for the pressure drop in the absorption tower. They are also used to pump the amino solution and to cool the water, as well as to compress or cool the CO_2 before it is transferred [35].

It should be noted that the CO_2 recovered in this way is of high quality and can be mixed with the existing CO_2 input to the CO_2 compressor to participate in the urea synthesis. Additionally, amines are used as solvents to counteract the side effects of combustion gases such as O_2, NOx, and SO_2.

It is worth mentioning that carbon dioxide emissions are heavily influenced by operation costs. Although almost all CO_2 can be separated, separating the last few percent requires a large amount of energy and is expensive. Normally, amines can separate CO_2 from gas in about 59% of cases. Operation cost for this process includes the cost of steam to recover the solvent as well as electricity cost to supply pumps and fans. Moreover, there are additional costs for

other stuff like leveling and replacing the waste solvent with fresh and additional solvent.

Table 1 compares these two methods in detail. According to Table 1, both methods have their advantages and disadvantages. The optimal method is determined by the amount of CO_2 required and the cost of energy. Recovering CO_2 from combustion gases is the best choice and solution for large amounts of CO_2 or a unit with a high gas cost, while, if a small amount of CO_2 is needed or the energy price is not too high, increasing the production of synthetic gas can be considered. It is worth mentioning that air pollution is another important factor to select the optimal solution. By capturing CO_2 from combustion gases, the amount of CO_2 emitted from the unit to the atmosphere will be greatly reduced. The development of various chemicals for CO_2 recovery has been studied in recent years to improve the processes and reduce recovery costs. The proposed framework can be used wherever there is CO_2 available as well as heat for starting the urea fertilizer production process. Therefore, ships seem like a good choice for testing the applicability of the proposed framework on moving objects.

3. Results and Discussion

The study was carried out at Pardis Petrochemical's second ammonia unit. The ratio of molar steam to gas flow in the reformer was set at 3.2. In addition, steam flow is 145 tons per hour and natural gas is 56 thousand normal cubic meters per hour according to the design. The steam input was therefore increased by one ton per hour for the experimental study, which reached 146 tons per hour after the necessary coordination. Temperatures in the carbon dioxide removal section were monitored. Moreover, Table 2 illustrates how the temperature indicator (TI) changes when the steam flow increases.
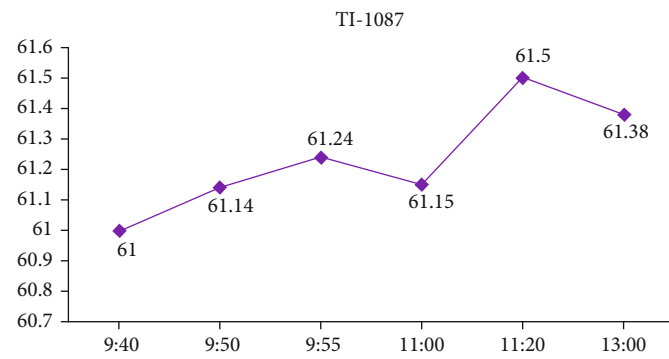
The TI-1085 displays the gas temperature as input to the E-2003 converter. Process gas heat is used to heat the aerated
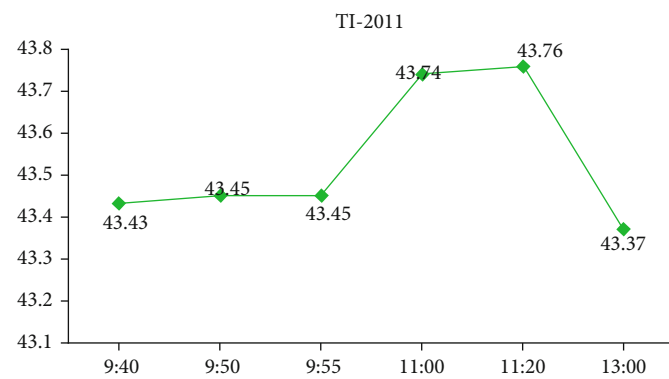
TI-1085

(a)

TI-1088

(b)

TI-1087

(c)
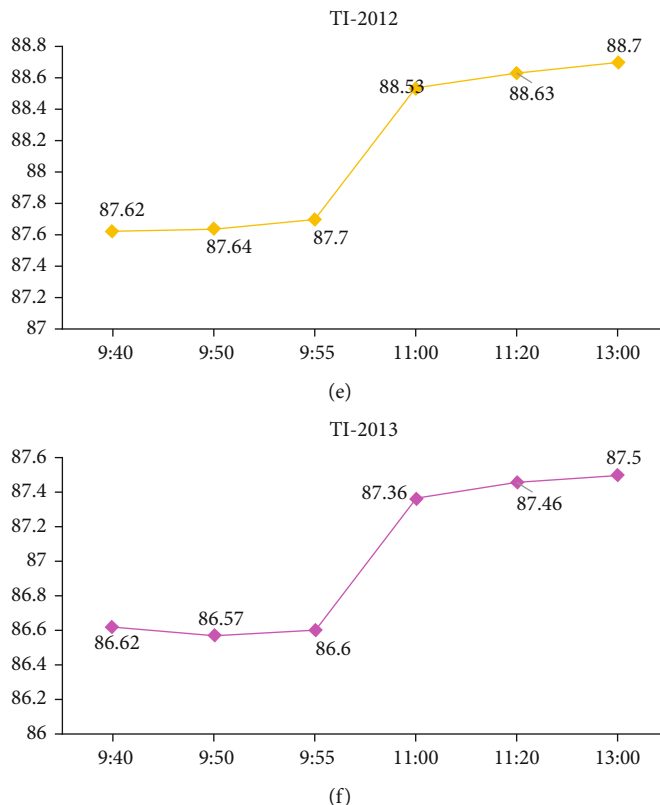
TI-2011
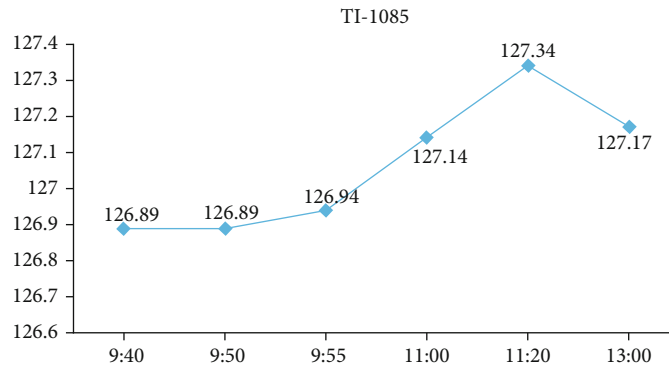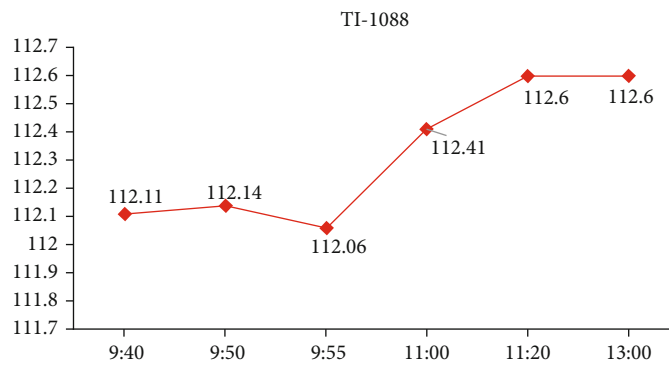
(d)

FIGURE 6: Continued.

(e)

(f)

FIGURE 6: Temperature changes of (a) input process gas to the E-2003 converter over time, (b) outlet water from E-2003, (c) process gas after E-2003, (d) gas imaged from adsorption tower, (e) MDEA liquid at the outlet of the adsorption tower, and (f) rivet gas output from the flash drum.

water. According to Figure 6, when excess steam is present, it transfers more heat to the water, which makes it warmer.

Figure 6 provides an evaluation of the outlet water temperature of the E-2003 converter (TI-1088). TI_1087 represents the process temperature after the E-2003 converter. The process gas transfers its heat to water up to the condensate temperature. When this temperature is reached, water vapor condenses and separates from the gas flow in the D-2001 drum. Excess heat is then released from the converter. The TI-2011 also indicates the temperature of the purified gas from the adsorption tower (Figure 6). As the steam rises, the temperature increases. TI-2012 MDEA liquid temperature is the output of the absorption tower, while TI-2013 is the gas temperature of the flash drum output tower. This gas is an unabsorbed gas in the MDEA system. In fact, some of the process gas that comes out of the absorption tower with MDEA is flashed in this drum and removed from the system.

CO_2 temperature is the output of the discharge tower in TI-2019, while TI-2003 is the disposal tower's low temperature that is the two-phase temperature of the MDEA system and provides the necessary heat to the disposal tower. Moreover, the temperature of the semilean amine moving towards the adsorption tower to reabsorb CO_2 is TI–2017. The lean amine temperature for adsorption of CO_2 and the exhaust gas temperature of the E-2001 converter are TI–2014 and TI-1084, respectively. It heats the water from the air conditioner to make steam. Furthermore, TI-1082 is the outlet water temperature of the E-2001 converter (Figure 7).

Results indicated that it is possible to increase the heat of a gas stream by adding steam to it, which increases its heating capacity. However, there are limitations to how much steam can be used. Figures 8 and 9 illustrate the effect of two other factors, including FQI-2002 and AT-1004B.

FQI-2002 represents the flow rate of CO_2 extracted in the CO_2 removal section. This amount has increased with the addition of steam, which is related to the rise in discharge tower temperatures. In order to conduct further investigation, the high temperature of the tower needs to be maintained constant, which is not possible given the unit conditions (Figure 9).

The second aspect that must be examined is the amount of CO entering the adsorption tower (process according to T-2001). Clearly, this value has decreased, meaning that the unit is operating in better conditions, which is why FQI-2002 has increased. With a lower amount of CO escaping in this section compared to the previous sections, the amount of CO_2 being converted has increased, which has provided very favorable conditions (Figure 9).

The concentration of ammonia and the pH of the solution were measured through titration. The data in Table 3 illustrate CO_2 uptake in aqueous ammonia at 20°C. Based on the results, $k_{OH-}[OH^-]$ is much smaller than $k_2[NH_3]$.
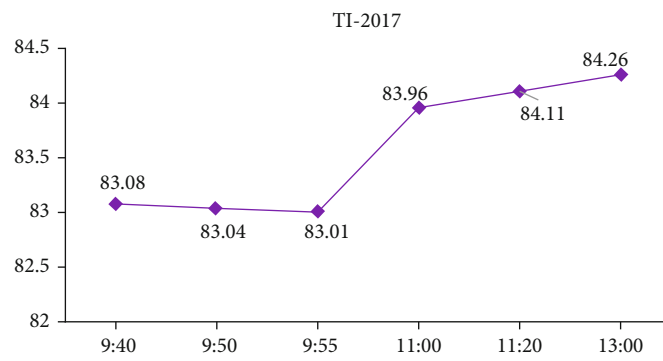
The ratio of $k_{OH-}[OH^-]$ to the reaction rate constant ($[k_2[NH_3] + k_{OH-}[OH^-]]$) is approximately 6.92% if 2% of the solution is aqueous ammonia. Furthermore, the contribution of $k_{OH-}[OH^-]$ decreases as the concentration of

TI-2019

(a)

TI-2003

(b)

TI-2017

(c)

TI-2014
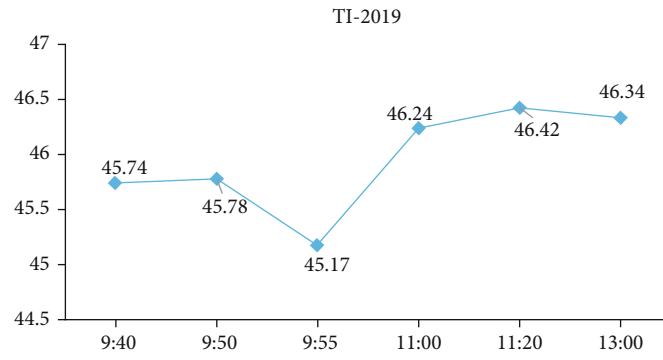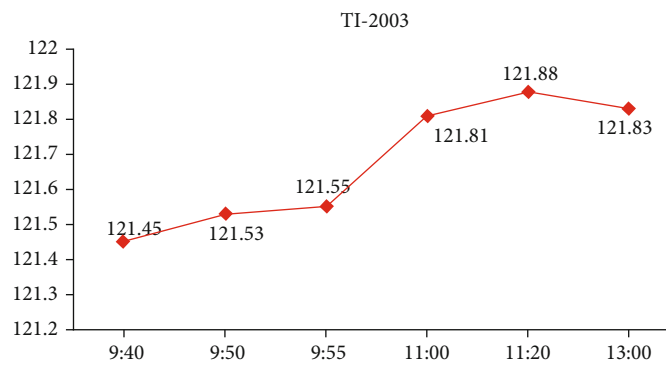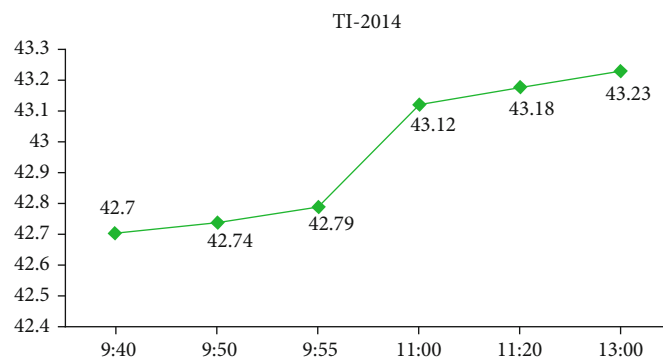
(d)

FIGURE 7: Continued.

TI-1084
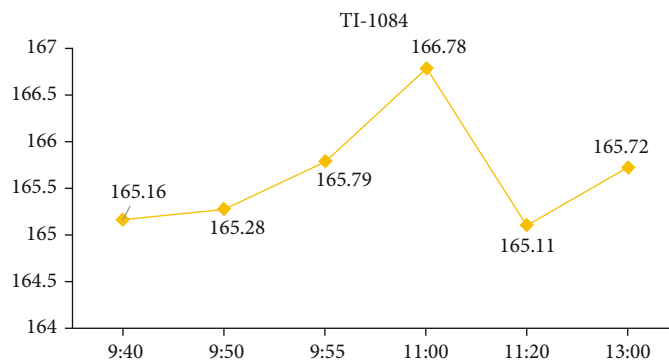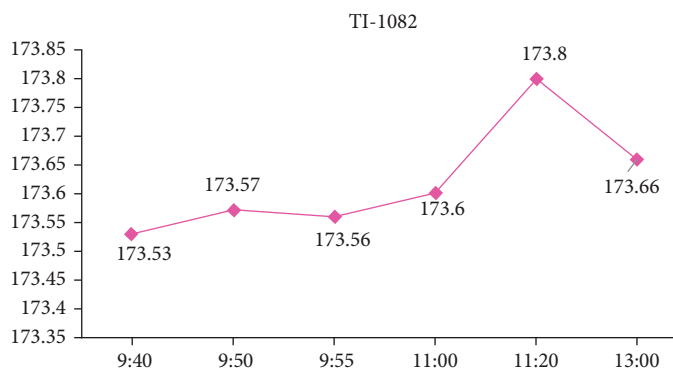

(e)

TI-1082

(f)

FIGURE 7: Temperature changes of (a) CO_2 at the exhaust tower, (b) discharge tower, (c) semilean liquid, (d) lean liquid, (e) exhaust gas from the exchanger (E-2001), and (f) outlet water from the E-2001 converter.
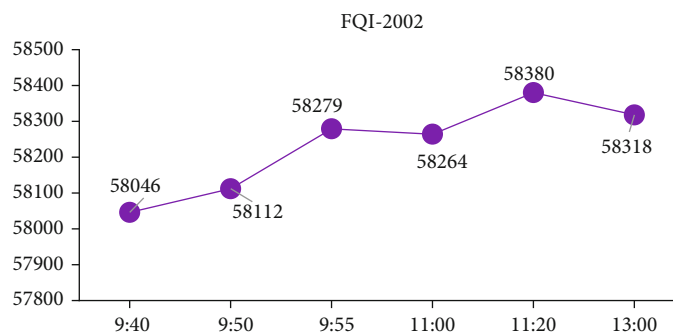
FQI-2002

FIGURE 8: EQI rate of carbon dioxide extracted (Nm^3/h).
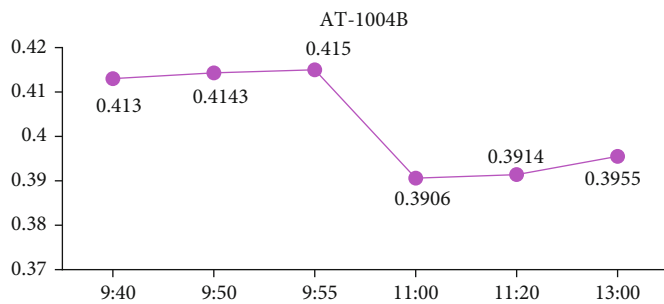
AT-1004B

FIGURE 9: The amount of CO entering the adsorption tower.

TABLE 3: Synthetic data for CO_2 uptake in aqueous ammonia solution.

Temperature (˚C)	C (NH_3) (%)	[NH_3] (kmol m^{-3})	[OH^-] (kmol m^{-3})	$k_2[NH_3]$ (A) (S^{-1})	$k_{OH-}[OH^-]$ (B) (S^{-1})	$B/(A+B)$ (%)
20˚C	2	1.16	0.0045	348	25.87	6.92
20˚C	4	2.3	0.0063	690	36.21	4.99
20˚C	6	3.43	0.0077	1032	44.26	4.11
20˚C	8	4.54	0.0088	1362	50.58	3.58
20˚C	12	6.71	0.0107	2013	61.5	2.96
20˚C	16	8.81	0.0123	2543	70.7	2.51

aqueous ammonia increases. In this case, the share of reaction explained by Equation (9) in the total reaction rate is less than 7% when the mass fraction of aqueous ammonia solution exceeds 2%. Since the rate of reaction between CO_2 and ammonia solution is mainly determined by Equation (10), it can be ignored that CO_2 reacts with negative ions. The reaction between NH_3 and CO_2 is extremely fast, and the absorption of CO_2 in ammonia occurs in a fast first-order reaction. In reality, cars are a major source of CO_2 emissions. We will have less air pollution if we reduce CO_2 emissions from cars. Therefore, future work will focus on how to store CO_2 and convert it into a less hazardous material in hybrid vehicles to reduce air pollution. As a result of the obtained results in this paper, removing CO_2 not only reduces environmental impact but also reduces extra ammonia, resulting in a more environmentally friendly industry [36–38].

4. Conclusion

In this paper, laboratory experiments were conducted to measure the carbon dioxide adsorption rate in an aqueous ammonia solution by using a filled tower. The temperature has a significant effect on CO_2 uptake into aqueous ammonia solutions, as demonstrated by the obtained results. An optimal temperature for this experiment was found to be around 40˚C; temperatures above which had detrimental effects on absorption rates. In a filled tower, CO_2 adsorption rates are determined by the main operating parameters. Furthermore, it increases with liquid flow intensity, gas flow intensity, ammonia concentration, and input CO_2 concentration. It was also observed that ϕ increases with the partial pressure of the gas mass (PCO$_2$). On the other hand, the total mass transfer coefficient (KGav) is determined by calculating the slope of straight lines on the diagram of the adsorption rate of the gas mass versus its partial pressure at various concentrations of ammonia. Consequently, it seems that an aqueous ammonia solution is an ideal adsorbent for absorbing CO_2 from combustion gases. This solution can be used as an adsorbent with low operating costs that absorbs CO_2 from the exhaust gases of the ammonia unit in order to boost urea production.

In the CO_2 adsorption unit, the biggest loss was attributed to adsorption and disposal towers. It is possible to improve the thermodynamic conditions of the tower by altering the feed conditions and using reboilers and side condensers. An analysis of tower diagrams shows that a uniform distribution of the driving force leads to higher thermodynamic efficiency. Changes to the feed of the ammonia unit and an increased amount of vapor have resulted in a rise in temperature in the reboiler of the disposal tower.

As found in this paper, converting CO to CO_2 increases the overall efficiency of the unit. In fact, less CO is needed to methanize. It was also found that the temperature of the shift reactor could be approached up to 365˚C. In this case, the rate of conversion of CO to CO_2 and the overall efficiency of the unit increase, while the rate of CO escape from shift reactors decreases.

Data Availability

Data are available on request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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