Modeling of the CO_2 Separation Process from Acid Gas Feed into the Sulfur Recovery Unit of Asaluyeh Phase 1 Using Membrane

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ABSTRACT: *In the sweetening process, acidic and sour gases, including CO_2, are separated and transferred to the sulfur recycling unit. CO_2 is one of the impurities in natural gas. In addition to its harmful effects on the environment, pipelines, and refinery equipment, it also has many benefits in the field of oil, gas, and petrochemicals. For this reason, the ability to Separation rate CO_2 emissions by high-efficiency tools that are also economically viable is important. In this study, modeling this process before operation can be an important step in reducing the high cost of separation. In this research, the CO_2 separation process using membranes has been modeled by MATLAB software, Then, the effect of CO_2 separation on sulfur recovery rate was performed using sulfur recovery unit simulation by Promax software. As a result, the highest amount of sulfur recovery in the membrane process in Poly Ether Urethane Urea membrane at the level of 100000 m^2 with a selectivity of 1.65 in which the amount of S_2, S_3, S_4, S_5, S_6, S_7, and S_8 are 0.1897, 0.0191, 0.01615, 4.668, 291.3737, 121.5916, and 1821.651 kmol/h, respectively. In poly ether urethane urea membrane with a selectivity of 1.65, the optimal point is obtained at a pressure of 35 kPa and a flow rate of 72.613 mol/s. The optimum point in the dimethyl silicon rubber membrane is achieved at a permeability pressure of 25 kPa and a flow rate of 98.4847 mol/s.*

KEYWORDS: *Sweetening; Sulfur recovery unit; CO_2 Separation; Membrane; MATLAB; Promax.*

INTRODUCTION

In 1980, Perma (Member of the Air Product Union) Separated hydrogen by membrane. This was the first widespread industrial use of membrane gas separation and further the use of membranes in the separation of gases grew significantly, so that the volume of investment in this field increased to $ 150 million per year and more growth is expected in the future [1]. A membrane is defined as a phase through which feed components selectively pass. In other words, the membrane operates in a fuzzy way through which the separating components of the feed pass at different speeds. In this method, there is usually no phase change and Products can also be mixed together [2].

Membranes have significant potential in the separation of gaseous mixtures; unfortunately, the attempt to build economic modules did not make much progress. In the late 1960s and early 1970s High flux membranes and high surface area membrane modules were produced for reverse osmosis applications. Perma used this technology to

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1021-9986/2022/12/4023-4031 9/$/5.09

separate the membrane. The production of polysulfone hollow fiber membrane was the first success of this company, especially for the separation and recycling of hydrogen from the gas stream in the designs used, other companies were encouraged to develop the technology [3].

One of the new methods for separating CO_2 from gas streams is the use of Selector membrane that can separate CO_2 from combustion gases, natural gas and hydrogen. Membranes have been widely used in various industrial separation industries for the last two decades. Industrial applications are mostly done with polymer membranes, but research into the development and application of inorganic membranes has grown exponentially due to their use in new application trends such as fuel cells, membrane reactors, and other high temperature separations. Most gas separation membrane processes require a very thin selector layer to produce an economically acceptable flux. Membranes are usually less than 0.5 micrometers thick or even less than 0.1 micrometers. Membranes are almost permeable and have the ability to separate different components by different mechanisms. Membranes are almost permeable and have the ability to separate different components by different mechanisms (Influence Knudson, Molecular Screening, Separation of dissolution-influence, Surface penetration, and capillary density). Membranes have several valuable benefits, such as no need for energy to regenerate, they have small sizes, and no waste currents compared to adsorption processes, and with the help of several steps by membranes, high purity of separation was achieved [1].

In the sweetening process, acidic and sour gases, including CO_2, are separated and it is transferred to the sulfur recovery unit (SRU). CO_2 is one of the impurities in natural gas which, in addition to its harmful effects on the environment, corrosion of pipelines and refinery equipment, crystallization of gas during liquefaction, and reduction of calorific value of natural gas, has many benefits in the field of oil, gas, and petrochemicals. There are different solutions for CO_2 disposal, and each country uses a solution for disposal and storage according to its resources, industry, and geographical location. The use of this gas in the beverage industry, or increasing the volume of groundwater and the production of dry ice are also among the benefits of this gas. Membrane separation is one of the solutions to absorb this gas. Each membrane has its own characteristics, advantages and disadvantages [4].

The first role of the Claus reaction furnace (Claus sulfur recovery unit) is to oxidize H_2S in acidic gas feed to SO_2. This role also gives an important amount of total sulfur produced. The second task of the reaction furnace is to ensure the destruction of harmful compounds in the acidic gas feed vapor. These compounds contain heavy hydrocarbons that cause the decomposition of catalytic beds. Heavy hydrocarbons decompose at high temperatures. Therefore, the reaction furnace temperature must be high enough (above 1100 °C). One of the most important methods to increase the temperature of the reaction furnace is the concentration of H_2S in the acid gas feed, which is possible by separating CO_2 from the acid gas stream by the membrane. For this reason in this study, the CO_2 membrane separation process from acid gas feed into the sulfur recovery unit of Asaluyeh Phase 1 is modeled using Matlab.

Assaluyeh phase one refinery

Phase 1 project is one of the phases of South Pars gas field which is located in the southern part of this region. South Pars Phase 1 onshore facility is located in the Assaluyeh region with an area of about 141 hectares in the Pars Energy Special Economic Zone. The offshore wells and platforms of this project are located at a distance of about 105 km from the azure shores of the Persian Gulf. South Pars Phase 1 development plan was defined with the aim of extracting and transferring one billion cubic feet of gas per day from 12 wells from two satellite well platforms. In which the extracted gas, after being transferred to a process platform for dehumidification is transferred to the onshore section through a 32-in pipeline and is desalinated in onshore facilities and injected into the national pipeline. The separated gas condensate in the refinery is exported via submarine pipeline and SPM.

EXPERIMENTAL SECTION

Sulfur recovery unit

This process is used to recover sulfur compounds from the sour gas streams of gas and crude oil refineries in order to prevent the emission of polluting gases as well as sulfur production. One of the main uses of sulfur in the world is the production of sulfuric acid. Refineries are typically required to reduce the amount of compounds in their products in order to produce products that meet quality and environmental standards, Hydrogen treatment processes are designed and built in refineries for this purpose. In these processes, the sulfur compounds in the crude oil slices are converted to hydrogen sulfide (H_2S) gas. To recover the sulfur in this gas stream, the sulfur recovery unit is used. H_2S is the major source of sulfur in the sulfur recovery unit and its concentration in feed can have a great effect on sulfur recovery. According to the Le chatelier principle, as the concentration of H_2S in the acid gas feed increases, the amount of sulfur recovered by the Claus reaction increases according to the following equations [4].

$$H_2S + 3/2\ O_2 \leftrightarrow SO_2 + H_2O \quad (1)$$

$$2H_2S + SO_2 \leftrightarrow 3/n\ S_n + 2H_2O \quad (2)$$

Modeling

To perform mathematical modeling in this research, the following mathematical relations have been used:

$$n_f = n_p + n_R \quad (3)$$

The degree of penetration of the components into the membrane is as follows:

$$y_P n_p = x_f n_f - x_R n_R = Q(\overline{xP - yp})A_m \quad (4)$$

In this part, x and y are the molar fractions, p penetration pressure of components or permeability pressure, Q membrane permeability, P inlet feed pressure and A_m is the cross section of the membrane. In these relations is $(\overline{xP - yp})$ the average logarithmic partial pressure of the membrane, which is calculated as Equation (5).

$$\overline{(xP - yp)} = \frac{(x_F P - y_p p) - (x_R P - y_i p)}{\ln\left(\frac{x_F P - y_p p}{x_R P - y_i p}\right)} \quad (5)$$

The following y_i shows the permeable components in the membrane residue, which is calculated from the following equation:

$$y_i = \frac{(\alpha^*-1)(rx_R+1)+r}{2(\alpha^*-1)} - \frac{\sqrt{[(\alpha^*-1)(rx_R+1)+r]^2-4(\alpha^*-1)\alpha^* rx_R}}{2(\alpha^*-1)} \quad (6)$$

$\alpha^*$is membrane selectivity and r is the ratio of inlet feed pressure to infiltration pressure.

$$r = \frac{P}{p} \quad (7)$$

Resolving these relationships has been done with the help of MATLAB software and the desired unknowns have been calculated [4].

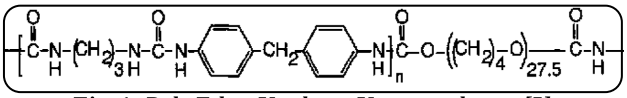

Fig. 1: Poly Ether Urethane Urea membrane [5].

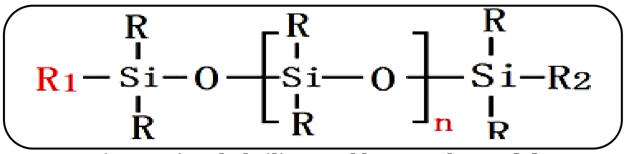

Fig. 2: Dimethyl silicon rubber membrane [5].

Membranes

In this study, two types of membranes with different selectivity have been used. In the following, membranes are introduced.

Poly Ether Urethane Urea membrane

Poly ether urethane urea is a type of polymeric membrane formed by chemical reactions between terminal isocyanate compounds and polyhydroxyl compounds. Fig. 1 shows the chemical formula of this type of membrane [5].

Dimethyl silicone rubber membrane

Dimethyl silicon rubber membrane is a type of mineral polymer, in the main chain of which O-Si-O bonds are placed instead of carbon atoms, and in its structure, two methyl groups are attached to each silicon atom. Fig. 2 shows its structure [5].

Promax software

In 1974, BR&E began developing simulation software for sulfur recovery units. In 1976, the program was released as Sulfur. Promax simulation software is currently the standard software for designing amine gas desalination units and glycol gas dehumidification.

Needed information

Assaluyeh Phase 1 Sulfur Recovery Unit Data

This section contains information about flow (kmol/h), temperature (℃), pressure (bar_g) the natural gas entering the sulfur recovery unit is phase one of Assaluyeh.

Membrane information

The following is a more detailed description of them in Table 1 [7-10]. That, Information about the membranes used in this study includes the surface area (m^2), selectivity, permeability ($\frac{mol \cdot m}{m^2 \cdot s \cdot pa}$) and penetration pressure (bar_g).

Table 1: Membrane specifications [7-10].

Membrane	H_2S permeability $\left(\frac{mol \cdot m}{m^2 \cdot s \cdot Pa}\right)$	CO_2 permeability $\left(\frac{mol \cdot m}{m^2 \cdot s \cdot Pa}\right)$	$\alpha_{H_2S}/_{CO_2}$ selectivity
Poly Ether Urethane Urea	800.65×10^{-16}	485.75×10^{-16}	1.65
Dimethyl silicon rubber	3350×10^{-16}	1088.75×10^{-16}	3.08

Table 2: Comparison of real data with simulator data [12, 13].

Parameters discussed		Inlet sour gas	Exhaust gas from the simulator	Real exhaust gas	$(1-(X_M/X_R))$
Temperature (℃)		49.7	126.667	130	0.0256
Pressure (bar_g)		0.8	0.1243	0.33	0.6234
Components (in molar percentage)	H_2	0	1.14	1.23	0.073
	N_2	0	45.75	47.69	0.041
	CO	0	1.68	1.11	-0.51
	CO_2	38.10	17.92	16.66	-0.075
	H_2S	55.29	0.5	0.5	0
	COS	0	0.137	0.15	0.086
	SO_2	0	0.24	0.25	0.04
	CS_2	0	0.018	0.02	0.1
	H_2O	6.61	32.77	32.39	-0.0117
	S_X	0	0.0052	0.01	0.48

Simulation

In this research, Promax software has been used to simulate the sulfur recovery unit. In the following, the effect of membrane process on sulfur concentration output from sulfur recovery was investigated.

Initially, the main data of Assaluyeh Phase One Sulfur Recovery Unit was used as software and system input included gas compounds, pressure and temperature [11].

After performing the operation and obtaining the results, Actual data is compared with simulator data (Table 2) [12, 13].

$$R^2 = 1 - \frac{\sum\left(1 - \frac{X_M}{X_R}\right)}{M-1} * 100 = 95.25\% \tag{8}$$

Table 2 compares the actual data with the simulation results of the Assaluyeh Phase One Sulfur Recycling Unit. As can be seen in the table, the data obtained from the simulator are very close to the actual data. To assess the reliability of the obtained data by placing the obtained parameters in Eq. (8), the accuracy of this was confirmed by obtaining 95.25%. This high percentage of confidence indicates that the simulation can be trusted to achieve the desired goal and achieve the desired result.

Table 3 shows the amount of sulfur extracted from natural gas in the Assaluyeh phase 1 sulfur recovery unit.

Table 3: The amount of sulfur extracted in the sulfur recovery unit of phase 1.

$\sum S_n$	$\sum S_2$	$\sum S_3$	$\sum S_4$	$\sum S_5$	$\sum S_6$	$\sum S_7$	$\sum S_8$
Molar flow (kmol/h)	0.1732	0.01529	0.01467	3.467	266.88	112.948	1664.63

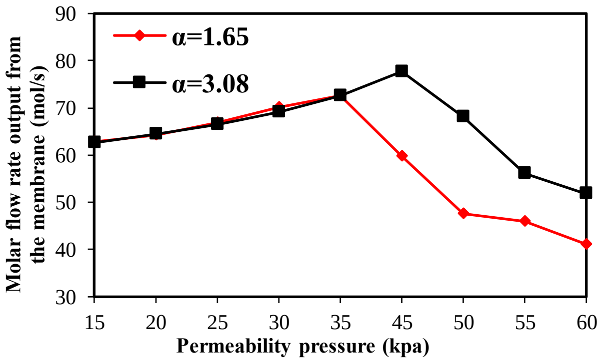

***Fig. 3: Flow rate of the output of the membrane relative to the permeability pressure at the level of 50000 m²*.**

RESULTS AND DISCUSSION

In Fig. 3, the concentration of H_2S after the passage of acid from the two membranes with two different selectivity at the level of 50000 m^2 and at different permeability pressures is investigated.

As shown in Fig. 3, each membrane has a specific optimum point; at that point, it has the highest amount of CO_2 absorption. In poly ether urethane urea membrane with selectivity of 1.65, the optimal point at a pressure of 35 kPa and a flow rate of 72.613 mol/s, where the H_2S concentration is 100%. Also, in dimethyl silicon rubber membrane with selectivity of 3.08, the optimum point at 45 kPa transmittance pressure and 77.1357 mol/s flow rate, where the H_2S concentration at this point is 100%.

In Fig. 4 the concentration of H_2S after passing acidic gas through the two membranes at the level of 100000 m^2 and at different permeability pressures is investigated.

In Fig. 4, the optimal point of poly ether urethane urea membrane at permeability pressure is 15 kPa and flow rate is 99.41 mol/s and the concentration of H_2S at this point is 100%. The optimum point in the dimethyl silicon rubber membrane at a permeability pressure of 25 kPa and a flow rate of 98.4847 mol/s, and the H_2S concentration at this point is 100%.

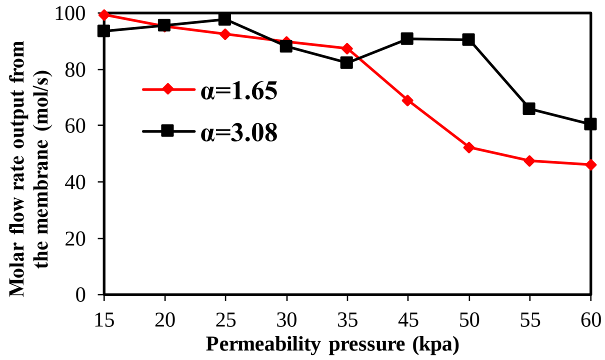

Fig. 4: Flow rate of the output of the membrane relative to the permeability pressure at the level of 100000 m^2.

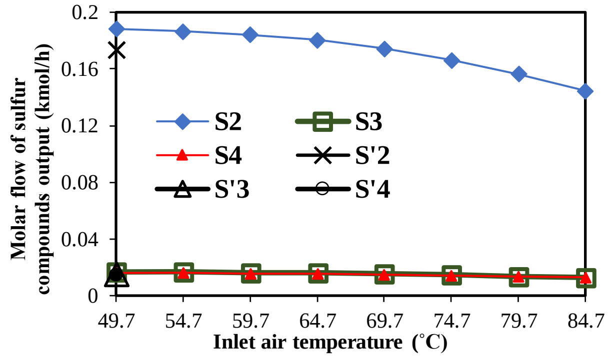

Fig. 6: Changes in inlet air temperature relative to the molar flow of sulfur compounds at the surface of 50000 m^2 membrane.

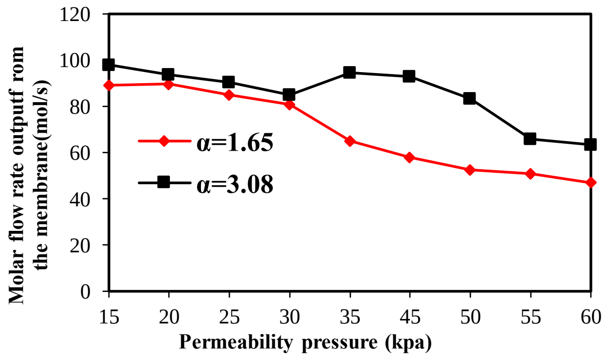

Fig. 5: Flow rate of the output of the membrane relative to the permeability pressure at the level of 150000 m^2.

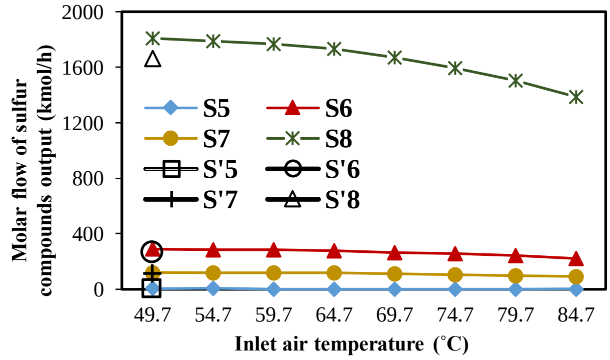

Fig. 7: Changes in inlet air temperature relative to the molar flow of sulfur compounds at the surface of 50000 m^2 membrane.

In Fig. 5 the amount of H_2S concentration after passing acidic gas through the two membranes at the level of 15000000 m^2 and at different permeability pressures is investigated.

In Fig. 5, the optimal point of the first membrane at a permeability pressure of 20 kPa and a flow rate of 89.7427 mol/s and the concentration of H_2S at this point is 100%. The optimum point in the second membrane is 15 kPa permeability pressure and 105.164 mol/s, and the H_2S concentration at this point is 93.18%.

In this part of the research, the data obtained from modeling are used as the input of the simulator unit. These data include the molar fraction of H_2S and the molar fraction of CO_2 and the amount of flow rate from the membrane at the cross-section of 50000 m^2. In this section, the changes in inlet air temperature to the sulfur recovery unit were discussed and its effects as a result of the work were investigated below.

Initially, the optimal point of poly ether urethane urea membrane at the initial level of 50000 m^2 studied is the input of the sulfur recovery unit. Figs. 6 and 7 show the amount of sulfur extracted (S_2-S_8) in this unit relative to the inlet air temperature changes.

As can be seen in Fig.s 6 and 7, changes in S_2 to S_8 molar flow rate have decreased with increasing temperature. In justifying the reasons for the changes in the downward trend of the changes in the flow rate due to the increase in temperature, we can also refer to the Loshatelia’s principle [14]. Loshatelia's principle states that chemical equilibrium reacts in the opposite direction to any change to reverse its effect. According to Eq. (1), an increase in temperature shifts the reaction in a direction that is accompanied by heat consumption to counteract the increase in temperature. In other words, increasing the temperature leads the reaction to the heater and decreasing the temperature leads the reaction to the exothermic [15, 16].

In the next part, the optimal point of dimethyl silicon rubber membrane at the level of 50000 m^2 is the input of the simulation unit. Fig.s 8 and 9 show the amount of sulfur extracted in this unit relative to changes in inlet air temperature.

Table 4: The effect of membrane process on the amount of sulfur extracted in the sulfur recovery unit of phase one using poly ether urethane urea membrane with a cross section of 50000 m^2.

$\sum S_n$	$\sum S_2$	$\sum S_3$	$\sum S_4$	$\sum S_5$	$\sum S_6$	$\sum S_7$	$\sum S_8$
Molar flow (kmol/h)	0.1882	0.016714	0.016	4.6365	289.38	120.752	1808.689

Table 5: The effect of membrane process on the amount of sulfur extracted in the sulfur recovery unit of phase one using dimethyl silicon rubber membrane with a cross section of 50000 m^2.

$\sum S_n$	$\sum S_2$	$\sum S_3$	$\sum S_4$	$\sum S_5$	$\sum S_6$	$\sum S_7$	$\sum S_8$
Molar flow (kmol/h)	0.1883	0.01678	0.016	4.63	289.24	120.69	1808.22

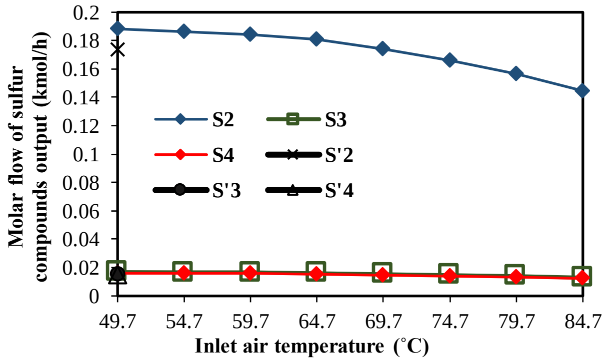

Fig. 8: Changes in inlet air temperature relative to the molar flow of sulfur compounds at the surface of 50000 m^2 membrane.

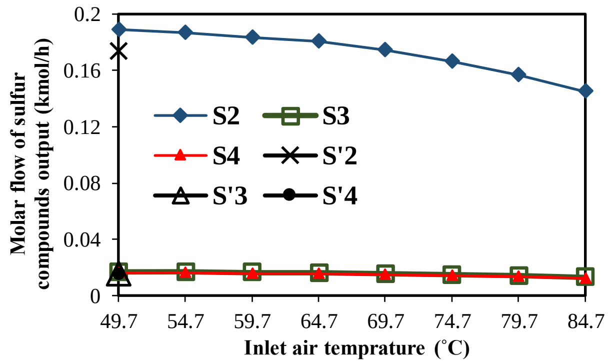

Fig. 10: Changes in inlet air temperature relative to the molar flow of sulfur compounds at the surface of 100000 m^2 membrane.

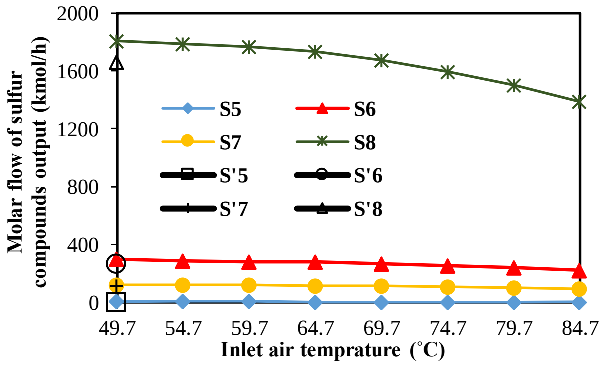

Fig. 9: Changes in inlet air temperature relative to the molar flow of sulfur compounds at the surface of 50000 m^2 membrane.

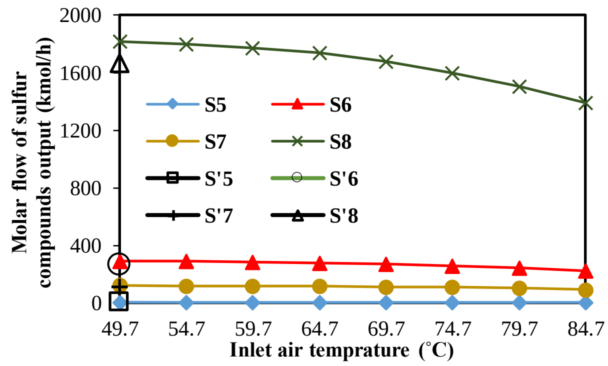

Fig. 11: Changes in inlet air temperature relative to the molar flow of sulfur compounds at the surface of 100000 m^2 membrane.

As can be seen in Fig.s 8 and 9, changes in the S_2 to S_8 molar flow rate have decreased with increasing inlet air temperature to the sulfur recovery unit. In addition, the reason for the decrease in the amount of molar flow of recycled sulfur can be referred to Loshatelia's principle, like the previous membrane.

In this section, as in the previous section, changing the inlet air temperature and its effect on the rate of sulfur extraction was studied. In this section, the optimal point of poly ether urethane urea membrane at the level of 100000 m^2 is the input of the simulated sulfur recovery unit. Figs 10 and 11 show the amount of sulfur extracted (S_2-S_8) in this unit relative to the inlet air temperature changes.

Observing Fig.s 10 and 11, it can be seen that the changes in (S_2-S_8) molar flow rate have decreased with increasing the inlet air temperature to the sulfur recovery unit. Reducing the molar flow rate means reducing the amount of sulfur recycling. In this section, the reason for this

decrease can be referred to the Loshatelia's principle.

In this section, as in the previous section, the optimum point of the dimethyl silicon rubber membrane at the level of 100,000 m^2 is the input of the simulated sulfur recovery unit. Fig.s 12 and 13 show the amount of sulfur extracted (S_2-S_8) in this unit relative to the inlet air temperature changes.

As can be seen in Fig.s 12 and 13, changes in the (S_2-S_8) molar flow rate have decreased with increasing inlet air

Table 6: The effect of membrane process on the amount of sulfur extracted in the sulfur recovery unit of phase one using poly ether urethane urea membrane with a cross section of 100000 m²

ΣS_n	ΣS_2	ΣS_3	ΣS_4	ΣS_5	ΣS_6	ΣS_7	ΣS_8
Molar flow (kmol/h)	0.1897	0.0169	0.01615	4.668	291.374	121.592	1821.66

Table 7: The effect of membrane process on the amount of sulfur extracted in the sulfur recovery unit of phase one using dimethyl silicon rubber membrane with a cross section of 100000 m²

ΣS_n	ΣS_2	ΣS_3	ΣS_4	ΣS_5	ΣS_6	ΣS_7	ΣS_8
Molar flow (kmol/h)	0.18906	0.01683	0.01608	4.651	290.303	121.142	1814.9

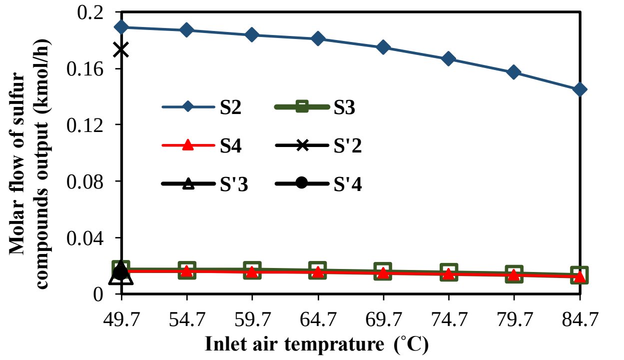

Fig. 12: Changes in inlet air temperature relative to the molar flow of sulfur compounds at the surface of 100000 m² membrane.

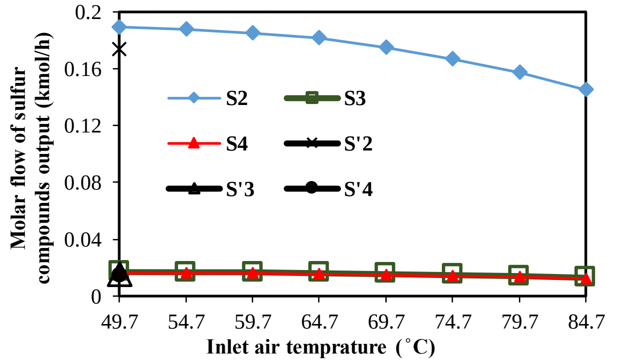

Fig. 14: Changes in inlet air temperature relative to the molar flow of sulfur compounds at the surface of 150000 m² membrane.

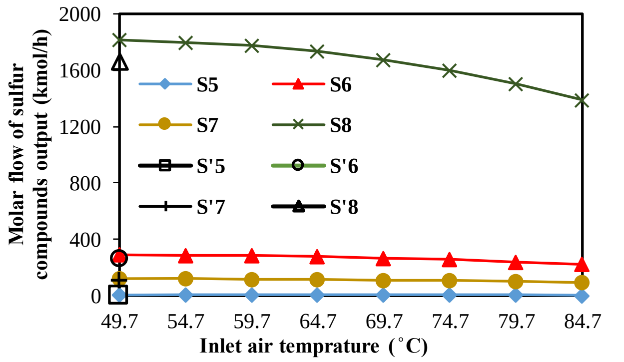

Fig. 13: Changes in inlet air temperature relative to the molar flow of sulfur compounds at the surface of 100000 m² membrane.

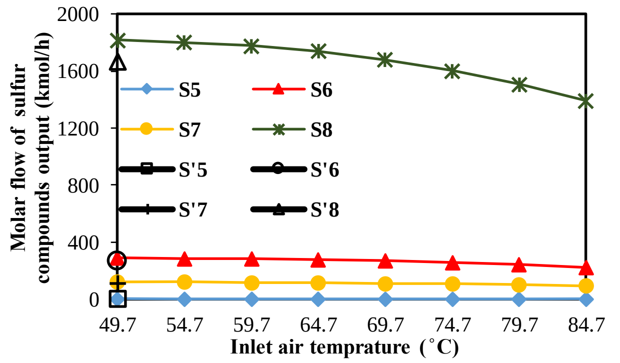

Fig. 15: Changes in inlet air temperature relative to the molar flow of sulfur compounds at the surface of 150000 m² membrane.

temperature to the sulfur recovery unit. Reducing the molar flow rate means reducing the amount of sulfur recovery. In this section, the reason for this decrease can be referred to the Loshatelia's principle. In the obtained information, as can be seen, the amount of sulfur recovery (S_2-S_8) decreases with increasing inlet air temperature per unit. As a result, the best temperature for obtaining the highest amount of sulfur is the lowest temperature (49.7℃).

In this section, as in the previous section, by changing the inlet air temperature, its effect on sulfur extraction was investigated. In this section, the optimal point of poly ether urethane urea membrane at the level of 150000 m² is the input of the simulated sulfur recycling unit. Figs. 14 and 15 show the amount of sulfur extracted (S_2-S_8) in this unit relative to the inlet air temperature changes.

Observing Fig.s 14 and 15, it can be seen that the changes in (S_2-S_8) molar flow rate have decreased with increasing the inlet air temperature to the sulfur recovery unit. Reducing the molar flow rate means reducing the amount of sulfur recycling. In this section, the reason for this decrease can be referred to the Loshatelia's principle.

In the next part, the optimal point of the dimethyl silicon rubber membrane with a cross section of 150,000 m² is the input of the simulated unit. In the following, as in previous, the effect of increasing the inlet air temperature on the amount of extracted sulfur is investigated in the Figs. 16 and 17.

Table 8: The effect of the membrane process on the amount of sulfur extracted in the sulfur recycling unit of phase one using poly ether urethane urea membrane with a cross-section of 150000 m^2.

ΣS_n	ΣS_2	ΣS_3	ΣS_4	ΣS_5	ΣS_6	ΣS_7	ΣS_8
Molar flow (kmol/h)	0.1894	0.0169	0.0161	4.6596	290.841	121.3676	1818.28

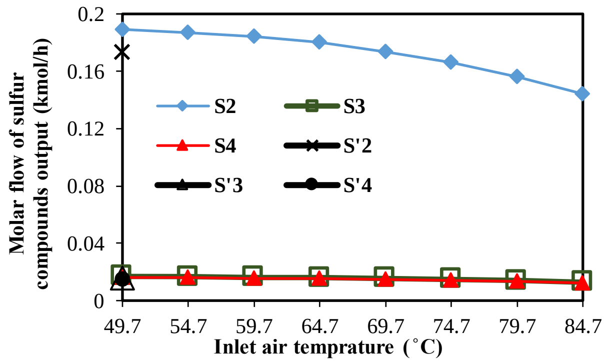

Fig. 16: Changes in inlet air temperature relative to the molar flow of sulfur compounds at the surface of 150000 m^2 membrane.

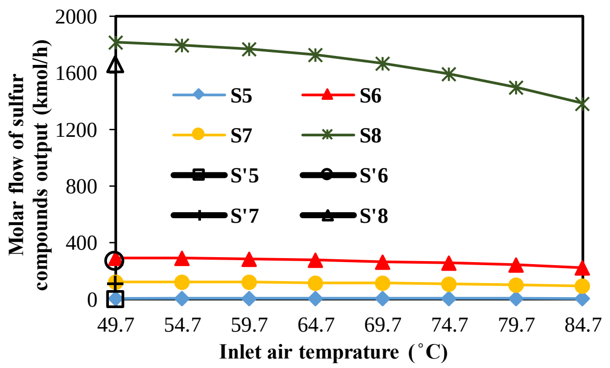

Fig. 17: Changes in inlet air temperature relative to the molar flow of sulfur compounds at the surface of 150000 m^2 membrane.

As can be seen in Fig.s 16 and 17, changes in (S_2-S_8) molar flow rate have decreased with increasing inlet air temperature to the sulfur recovery unit. Reducing the molar flow rate means reducing the amount of sulfur recovery. In explaining the reasons for the changes in the downward trend of the changes in the flow rate with respect to the increase in temperature, we can refer to Loshatelia's principle, which was described in the previous step.

CONCLUSIONS

In a recent study, the rate of separation of CO_2 gas from natural gas compounds entering the sulfur recovery unit of phase one of Assaluyeh using a membrane process

Table 9: The effect of membrane process on the amount of sulfur extracted in the sulfur recovery unit of phase one using dimethyl silicon rubber membrane with a cross section of 150000 m^2.

ΣS_n	ΣS_2	ΣS_3	ΣS_4	ΣS_5	ΣS_6	ΣS_7	ΣS_8
Molar flow (kmol/h)	0.1892	0.0168	0.0161	4.6556	290.596	121.27	1816.86

in MATLAB software has been investigated and in the following the effect of this separation on the amount of sulfur recycling was investigated using Promax software.

According to research conducted using the membrane process, the amount of CO_2 in the gas stream to the sulfur recycling unit can be minimized (0%). Separation CO_2 affects the amount of recycled sulfur compounds from the sulfur recovery unit and increases the recycling rate. By examining the diagrams drawn based on the simulation results in three different cross sections and for two types of membranes with different selectivity, it is observed that the best type of membrane for CO_2 separation is poly ether urethane urea membrane at the level of 100000 m^2 with the optimal points of that membrane. The highest amount of sulfur recovery in the membrane process was in Poly Ether Urethane Urea membrane at the level of 100000 m^2 with a selectivity of 1.65 in which the amount of S_2, S_3, S_4, S_5, S_6, S_7 and S_8 were 0.1897, 0.0191, 0.01615, 4.668, 291.3737, 121.5916 and 1821.651 kmol/h, respectively. In poly ether urethane urea membrane with selectivity of 1.65, the optimal point was at a pressure of 35 kPa and a flow rate of 72.613 mol/s. The optimum point in the dimethyl silicon rubber membrane achieved at a permeability pressure of 25 kPa and a flow rate of 98.4847 mol/s

Received: Dec. 25, 2021 ; Accepted: May. 16, 2022

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