

Simulation of Cellulose Acetate Membrane Module for CO₂ Separation from Natural Gas.

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Abstract: In this work, a mathematical numeric model is suggested to estimate the CO₂ separation from CH₄/CO₂ gas mixture via solution diffusion method using Cellulose Acetate based polymer membrane via CFD approach by means of numerical finite element method in order solve the principal equation of model. Influence of various parameters on removal efficiency of CO₂ from the mixture is investigated. The achieved modelling results then matched with the available industrial data to confirm the validation of the model results. Comparison of simulation extracted data with industrial results shown that the established model can calculate the CO₂ removal performance. Results indicated that increment of gas flow rate and inlet gas temperature reduces the of CO₂ removal efficiency. In contrast, CO₂ removal efficiency can be augmented by increasing the membrane area requirement.

Keywords: Separation; Membrane; Mathematical modeling; Numerical simulation; Mass transfer

1. Introduction

The World population was estimated to be 7 billion in 2011 and the daily global energy consumption was 16 Terawatt. By this proportion the currently daily global energy consumption has to be more than 18 Terawatt and all such energy is consumed in several ways. Like Commercial, Residential, Industrial and transportation... And yes 18 Terawatt is a big number. To give an idea, 1 terawatt can light up 10 billion, 100 watt bulbs at once. Since it is anticipated that by the 2050, world total population would be around 09 billion and the total global energy requirement would exceed 45 terawatts. This will take us to bigger climatic problems and its consequences than the world is facing today [1].

The power requirement is generated from different sources of energy. Namely Oil, Coal, Natural Gas and renewable energies such as Solar, Wind, Hydropower, nuclear energy and there are others as well. It is estimated that 24% of our daily energy consumption is generated from Natural Gas, which is the most abundant, cleanest and safest form of fossil fuel available. Why it is called cleanest because upon combustion natural gas produces 26% lesser CO₂ than Oil and 41% lesser CO₂ than Coal. And the CO₂ emissions are the sole reason for our global warming and climatic problems [2]. In order to mitigate the environmental problems, Natural Gas processing is performed to make on specification product. The requirement for natural gas treatment is described below;

1.1 Natural Gas Processing

Natural gas, that we consume, is far dissimilar to the N.gas which is taken out below the earth crust above to the surface. Even though the natural gas treatment is in several ways easier than the crude oil processing. The natural gas that we consume is mostly composed of CH₄ molecule. However, N.gas taken out at the wellhead assembly is primarily composed of CH₄. Raw N.gas is extracted from

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three kinds of hydrocarbon wells: i.e. oil wells, gas wells, and condensate wells. N.gas which arises from oil wells is called 'associated gas'. N.gas which arises from gas well and condensate wells, where there is a slightly presence of crude oil or not at all, is called 'non associated gas'. Gas wells naturally produces raw N.gas by itself, whereas condensate wells produces N.gas alongside a lighter liquid hydrocarbon called condensate. Whatsoever the basis of the N.gas, once the associated oil is removed (if contains) it generally carries liquid hydrocarbons; mainly C2, C3, C4, and C₅. In addition, raw N.gas holds moisture, CO₂, H₂S, He, N₂, and further compounds. N.gas processing involves removal of the undesired HCs and liquids from the product grade N.gas, to make its quality according to the pipeline specification i.e. dry N.gas. Transmission companies generally enforce limitations with the specifications for the N.gas which can be permitted to flow through their pipelines. Many of the accompanying HCs, generally called as 'natural gas liquids' (NGL) can be very valuable and useful by-products of N.gas treatment. NGL include C₂, C₃, nC₄, iC₄, and naphtha. The NGL products can be marketed and vended alone and have several applications; like enhancement of oil recovery from oil wells, raw materials for petroleum refineries or petrochemical industries, and as means for energy generation. Once all of these liquid HCs are removed from the N.gas, the N.gas is subjected to further gas processing. Mostly importantly Gas dehydration and gas sweetening [3].

After separating liquid HCs from the N.gas, it is necessary to remove the accompanying water content. Almost all of the free water is eliminated by simple separation technique. However, the removal of water content that exists in the vapor form in N.gas requires dehydration which usually involves processes like physical or chemical absorption or adsorption. Absorption is a process in which the absorbed vapor is removed by using a dehydrating agent. While in adsorption process, the vapors are condensed on its surface [3].

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Like water, other impurities that are present in N.gas and requires processing includes H₂S and CO₂. N.gas holds significant amount of H₂S and CO₂ as an impurity. This N.gas is termed as 'sour gas'. Sour gas is detrimental as the sulfur compounds present in it are enormously dangerous, even deadly to our respiratory system. Sour gas is also corrosive in nature. In addition, the sulfur compounds present in the N.gas can be removed and sold as a byproduct. CO₂ also lessens the calorific value of N.gas In transmission pipelines. For LNG, the CO2 removal is a crucial as it can achieve its freezing point during liquefaction process in chillers. There are numerous technologies available for acid gas treatment, which includes chemical absorption technique, such as the Benfield TM process (hot potassium carbonate as solvent) and Amine-Guard process (enhanced solvent); cryogenic distillation; adsorption technique, such as pressure swing adsorption, thermal swing adsorption, iron sponge; and polymeric membranes. Every method has their own pros and cons, but polymeric membrane separation technique is progressively being adapted for new projects, particularly for higher flowrates, higher CO₂ concentrations, and at remote locations [3].

2 Literature Review

The process in which Cellulose Acetate membranes are used for the separation of CO_2 is involved, the system uses a non-porous polymeric membrane for separating gas molecules of two or more gas components mixture. The membrane separation works on the principles of solution diffusion method or in other words it works on the partial pressure difference of gaseous mixture. So when a high pressure gas enter the membrane the molecules are initially sorped by the membrane at the higher pressure side, the molecules then penetrates along the membrane and finally desorped at the lower pressure side. All this happen because of the fact that certain gas molecules permeates more rapidly than others [4].

In Membranes, there are two significant factors that defines the separation efficiency of a membrane.

- 1. Permeability
- 2. Selectivity

A gas mixture is separated on the basis of their relative permeability as a gas mixture of two or more gases of varying permeability can be separated into two streams. One stream rich in more permeable component (i.e. CO_2 in my case) and the other stream rich in less permeable component (i.e. CH_4). Permeability is a combination of two factors, i.e. diffusion and solubility. The permeability of the membrane is mainly controlled by the solubility of the gas, and solubility of a gas molecule can be defined as the condensing capability to pass through the membrane compared to CH_4 because of the greater condensability, which can be shown by their critical temp. Tc, $CO_2 = 304$ K (30 C) and Tc, $CH_4 = 191$ K (-81 C) [4].

While the membrane selectivity is largely control by the membrane polymeric structure which allows the diffusion of molecules with respect to their molecular sizes, which is also called as kinetic diameter. So here the kinetic diameter for CO_2 is 0.33 nm and for CH_4 is 0.38 nm [4].



Fig. 2.1 Spiral Wound Membrane Element

So as you can see the figure 2.1, when a gas mixture is subjected to enter as a feed inlet stream in the membrane first at the higher pressure side, the polymeric membrane structure is designed to favor the fast moving molecules to move through the membrane and penetrates to the core. CO_2 molecules having high permeance rate then the rest of the gases and it will make it through the core and that stream becomes rich in CO_2 and it is called as permeate stream The slow gases which are unable to penetrate through the membrane will pass through along the membrane and leave the membrane as a residue gas which is rich in non-penetrant gas such as HCs [5].

2.1 Polymer Membrane

Cellulose Acetate is composed of a very thin non-porous polymer membrane layer attached to a denser and very porous coating of the same polymer. The structure of this membrane is stated to as a-symmetric, opposite of homogenous arrangement, in which membrane porosity is likely to be uniform all over. Figure 2.2 demonstrate the structure.



Fig. 2.2 Structure of a-symmetric polymeric membrane

The non-porous polymer part meet the needs of the perfect membrane, as, it is thin and extremely selective in nature. While the porous polymer part delivers reinforcement and permits the fast moving molecules to penetrate through the non-porous part. Since a-symmetric polymeric membrane is a huge development on homogenously arranged membrane, but it brings one disadvantage. Since it is made up of a single material, it is expensive, highly modified polymeric material, which practically is yielded in small quantities.

This struggle is countered by creating a composite structure, which involves a thin selective part prepared from single polymer material attached on an a-symmetric membrane structure, which is made up of a different polymer. The composite polymeric membrane structure allow producers to use easily existing materials for the asymmetric layer and personally designed and equally optimized polymeric substance required for the separation, for the selective layer. Figure 2.3 shoes such a membrane structure.



Fig. 2.3 Composite Polymeric Membrane Structure

These composite polymeric membranes are being utilized as the new CO_2 separation technique using polymeric membranes since the characteristics of membrane selective part is easily altered without causing membrane price to hike.

2.2 Molecule transport in polymer membrane

Solution diffusion philosophy is actually the phenomenon used by cellulose acetate polymer membrane for separating CO_2 molecule from CH_4/CO_2 mixture [6]. The permeation theory in the solution-diffusion method is comprised of following mechanisms, the same is described in figure 2.4.

- Sorption (at membrane's entrance)
- Diffusion (along membrane structure)
- Desorption (at membrane's permeate)



Fig. 2.4 Solution-Diffusion mechanism

The phenomenon of sorption and desorption are reverse in nature and can be written as function of CO₂/membrane solubility by applying Henry's Law;

$$J_S = S_i (C_F - C_P)$$
 2.1

 J_S is described as the molar flux due to desorption; S_i is the solubility factor of the membrane structure and the entering feed gas; C_F is the CO₂ ratio in the feed gas; C_P the CO₂ ratio in the permeate gas.

Likewise, Fick's law defines the diffusion process and can be written as;

$$J_D = \frac{D_i}{\gamma} (C_F - C_P) \qquad 2.2$$

 J_D is the molar flux due to diffusion; D_i is coefficient of diffusion of the membrane which is a function of various operating parameters and is greatly influenced by inlet feed gas temperature; γ is the membrane thickness; C_F is the CO₂ ratio of the entering feed gas; C_P is the CO₂ ratio of the permeate gas. Taking each phenomenon independently, we can write the total molar flux equation as;

$$J_T = J_S + J_D 2.3$$

$$J_T = S_i.(C_F - C_P) + \frac{D_i}{\gamma}.(C_F - C_P)$$
 2.4

$$J_T = \frac{S_i \cdot D_i}{\gamma} \cdot (C_F - C_P)$$
 2.5

Multiplying both sides with membranes required area, the molar flow through the membrane is obtained

$$J_T \cdot A = F_T = \frac{A \cdot S_i \cdot D_i}{\gamma} \cdot (C_F - C_P) \qquad 2.6$$

Eqn. 2.5 and 2.6 shows the dependency among the mass flowrate and de-sorption and diffusion coefficients, consequently the total mass flux will depend on these two phenomenon. While working with polymeric membranes, knowing the subjective gas component permeation rate is important. To calculate the polymeric membrane's capability to separate out a binary gas mixture into two components i.e. *i* and *j*. having considered the ratio of their permeability (α_{ij}); the product we get is called membrane selectivity, which is written as:

$$\alpha_{i/j} = \frac{P_i}{P_j} = \frac{D_i S_i}{D_j S_j}$$
 2.7

 D_i/D_j is the ratio between the coefficients of diffusion of two gases components and is considered as the mobility selectivity which is proportional to the ratio of the kinetic diameter of the penetrating molecules. S_i/S_j is the ratio of the coefficients of sorption, which specifies the concentration of *i* and *j* components in the polymeric membrane. The membrane selectivity is proportionate to the relative condensability of *i* and *j* components.

Using polymeric membranes technique for CO_2 separation the mobility selectivity D_i/D_j always support the penetration of smaller molecules like CO_2 (having kinetic diameter 0.33 nanometer) over bigger ones such as CH_4 (having kinetic diameter 0.38 nanometer).

To estimate the membrane separation efficiency of a gas mixture, two characteristics must be evaluated; the permeability and selectivity. Unluckily, higher CO_2 permeation does not relate to higher selectivity in polymeric membranes. Infact the association between these two factors happens to be inversely proportional to each other. Reaching a better breakeven of both parameters is a continuous objective for membrane's researcher.

To describe the scientific correlation on the trade-off curve between selectivity & permeability, Robeson defined an Upper Bound function which is shown in figure 2.5 as Robeson's trade-off curve for $CO_2/natural gas pair [7]$. It is significant to follow that this boundary progresses with technological advancement, and therefore is continuously under development.



Figure 2.5 Robeson's trade-off curve for CO₂/natural gas pair

3 Methodology

For calculations, a non-dimensional 1-D mathematical numeric model for multi-component steady state penetration in no sweep mode and co-current arrangement is used. In our case of binary compounds ($CO_2 \& CH_4$), the model equation is comprises of two ordinary differentials qwuations (for the retentate side) and two algebraic equations (for the permeate side) as shown below (2.8)– (2.11).

Feed/retentate side

$$\frac{\mathrm{d}\varphi_{\mathrm{CO}_2}^{\text{Retentate}}}{\mathrm{d}\zeta} = -\frac{1}{\phi}\Theta_{\mathrm{CO}_2}\left(\phi x_{\mathrm{CO}_2}^{\text{Retentate}} - x_{\mathrm{CO}_2}^{\text{Permeate}}\right) \qquad 2.8$$

$$\frac{\mathrm{d}\varphi_{\mathrm{CH}_{*}}^{Retentate}}{\mathrm{d}\zeta} = -\frac{x_{\mathrm{CO}_{2}}^{Feed}}{x_{\mathrm{CH}_{*}}^{Feed}\alpha_{\mathrm{CO}_{2}/\mathrm{CH}_{*}}} \Theta_{\mathrm{CO}_{2}}\left(\phi x_{\mathrm{CH}_{*}}^{Retentate} - x_{\mathrm{CH}_{*}}^{Permeate}\right) \quad 2.9$$

Permeate side

$$\varphi_{\rm CO_2}^{Permeate}(\zeta) = \varphi_{\rm CO_2}^{Feed} - \varphi_{\rm CO_2}^{Retentate}(\zeta)$$
 2.10

$$\phi_{\rm CH_*}^{Permeate}(\zeta) = \varphi_{\rm CH_*}^{Feed} - \varphi_{\rm CH_*}^{Retentate}(\zeta)$$
 2.11

In above equations, ϕ_{CO_2} and ϕ_{CH_4} are the non-dimensional molar flowrates for components CH₄ & CO₂, and ζ is the non-dimensional element length. [8]

$$\varphi_i = \frac{Q_i}{Q_i^{Feed}} \qquad 2.12$$

$$\zeta = \frac{z}{L}$$
 2.13

 Θ_i and φ are the permeation number and feed inlet gas to permeate gas pressures ratio. These operating parameters affecting the CO₂ removal performance of membrane.[8]

$$\Theta_{\rm CO_2} = \frac{\text{Permeance}_{\rm CO_2} A^{\text{Membrane}} P^{\text{Feed}}}{x_{\rm CO_2}^{\text{Feed}} Q^{\text{Feed}}} \qquad 2.14$$

$$\phi = \frac{P^{Feed}}{P^{Permeate}} \quad . \tag{2.15}$$

The results attained from these models are defined as plots of CO_2 concentration in permeate gas versus CO_2 recovery. It's beneficial for investigating what values are to be used for various operating parameters most commonly mass flowrate, feed temperature, pressure and pressure ratio and for evaluating the factors that can affect the separation performance of the membrane. Validation of the model by comparing the results with industrial data is also the scope of the research. [8]

The equations for non-dimensional entities such as Θ_i and ϕ which can be defined as the permeation number and the inlet feed gas to permeate line pressure ratio, respectively.

$$\Theta_{\rm CO_2} = \frac{\prod_{\rm CO_2} A^{Membrane} P^{Feed}}{x_{\rm CO_2}^{Feed} Q^{Feed}}$$
 2.16

$$\Pi_{\rm CO_2} = \frac{\rm CO_2 \ permeating \ flux}{\Delta P_{\rm CO_2}} \qquad 2.17$$

$$\Delta P_{\rm CO_2} = P_{\rm CO_2}^{Feed/Retentate} - P_{\rm CO_2}^{Permeate}$$
 2.18

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The permeation number (2.12) indicates an association between the two key transport tendencies. Higher the permeation number higher the membrane requirement are or permeance and to a higher permeation through the membrane module per total flux. The pressure ratio (2.15) denotes one of the most significant factor that can enormously affect the removal efficiency of the membrane and is also the motive force for the separation. For a predefined feed inlet composition, membrane characteristics (such as permeability and selectivity), membrane element geometry (overall membrane flux area and element length), and process operating parameters (inlet gas flowrate, pressure and temperature), this mathematical model predicts species non-dimension flowrate and composition profiles along the membrane element for both the retentate and permeate sides. Membrane element separation efficiency in terms of, for example, retentate stream's composition and overall CO₂ recovery in the permeate line can be formulated using equation 2.19. [8]

$$CO_2 \text{ recovery} = \frac{CO_2 \text{ permeate flow rate}}{CO_2 \text{ feed flow rate}} \times 100$$
 2.19

The model equations were used to calculate for nine cases, each having a different set of operating parameters and membrane characteristics. The data for each case is selected from industries where the membrane separation techniques is already in use. For each case membrane separation efficiency is taken as an account for the model accuracy.

Table 3.1 shows the process parameters, membrane element permeability and selectivity, and operating parameters used in the case studies in the above models.

Property	Simulation Cases									
	1	2	3	4	5	6	7	8	9	
Mass flowrate (kg/sec)	0.1	0.1	0.1	0.2	0.2	0.2	0.3	0.3	0.3	
pressure	500	500	500	500	500	500	500	500	500	
Inlet Temp. (°C)	35	40	45	35	40	45	35	40	45	
Pressure Ratio	50	50	50	50	50	50	50	50	50	
CO ₂ Conc. (Feed) %	7.0	7.0	7.0	7.0	7.0	7.0	7.0	7.0	7.0	
CO ₂ Conc. (Retentate) %	1.9	2.2	2.4	2.1	2.5	2.9	2.8	3.3	3.9	
CO ₂ Conc. (Permeate) %	78	76	74	72	68	64	67	62	56	
Membrane selectivity	50	50	50	50	50	50	50	50	50	
Permeation number	10	10	10	10	10	10	10	10	10	

Table 3.1 Process parameters used in case studies.

For selectivity any value can be used to study the predictive feature of the given model. Hence, a constant value for selectivity is considered similar to the Cellulose Acetate membrane element to restrict its effects on the separation efficiency membrane calculated by the model.

4 **Results and Discussion**

As discussed above, the CO_2 removal efficiency of a membrane unit is rigorously associated to 03 major factors: the CO_2 conc. in the feed inlet gas, the design and operating parameters, and the membrane characteristics used to achieve desired results. The study discusses the importance of these 03 factors and their reasonable effects on the separation performance of the membrane element. There were 09 simulated cases in total each having a set of different parameters and keeping the CO_2 composition in the retentate gas as a point of separation efficiency of the membrane model. The model results are mentioned in table 4.1.

Property	Simulation Cases									
	1	2	3	4	5	6	7	8	9	
Mass flowrate (kg/sec)	0.1	0.1	0.1	0.2	0.2	0.2	0.3	0.3	0.3	
Inlet pressure (Psig)	500	500	500	500	500	500	500	500	500	
(°C)	35	40	45	35	40	45	35	40	45	
Pressure Ratio	50	50	50	50	50	50	50	50	50	
CO ₂ Conc. (Feed) %	7.0	7.0	7.0	7.0	7.0	7.0	7.0	7.0	7.0	
CO ₂ Conc. (Retentate) %	1.9	2.2	2.4	2.1	2.5	2.9	2.8	3.3	3.9	
CO ₂ Conc. (Permeate) %	78	76	74	72	68	64	67	62	56	
Membrane selectivity	50	50	50	50	50	50	50	50	50	
Permeation number	10	10	10	10	10	10	10	10	10	
CO2 removal %age	82	79	76	62	59	55	46	40	35	

Table 4.1. Process parameters with calculated removal percentage

4.1 Effect of mass flowrate

For a given membrane area for gas separation, the mass flowrate of the inlet gas stream is directly proportional to the membrane area requirement. Hence a substantial increase in the overall mass flowrate of the gas can leads towards an increases in the overall membrane area requirement for that portion of the gas to process. The inlet gas flowrates also increases the overall hydrocarbon loss of the system in order to achieve the desired results. According to table 4.1 the mass flowrate changes after every 03 cases keep the other factors constant, the flowrate increases gradually hence the removal efficiency as per the model suggests also drops with the increase in flowrates as the CO_2 in the retentate also increases. Figure 4.1 shows the graph between flowrate and CO_2 removal percentage in the retentate gas.



Figure 4.1. Effect of Mass Flowrate on removal efficiency of CO₂.

4.2 Effect of inlet gas temperature

The temperature as a property of the gas acts crucial in process operations. Temperature can alter the efficiency of various operations because it can change the behavior of certain gas molecules of a gas mixture. Upon rise in temperature the gas molecules start moving more rapidly than before and this changes the volume of the gas. An addition to this, the partial pressure of the gas molecules also increases with the increase in gas temperature and the partial pressure of gas molecules are major driving force in the solution diffusion separation techniques. Due to increase in volume of the gas with the increase in temperature, the membrane area requirement also increases. As we can see from the figure 4.2 with increase in inlet gas temperature the separation efficiency decreases as an effect of it.



Figure 4.2. Effect of Inlet Gas Temperature on removal efficiency of CO₂.

4.3 Effect of Membrane Selectivity

This membrane property is important for the CO_2 composition in the final permeate gas. Its effect is incomplete when the permeation tendency is insufficient;

consequently, with lower CO_2 composition in the inlet feed gas stream, low pressure ratio may not attain higher CO_2 compositions in a single stage system, even using polymeric membranes with higher selectivity.

4.4 Effect of Pressure Ratio

Pressure ratio has a dynamic role in the separation efficiency of the membrane element. The effect of pressure ratio property relies on the various factors that can affect the separation performance, such as the inlet gas CO₂ composition, membrane selectivity, and permeation number. Technically, a membrane element with optimized membrane characteristics such (selectivity as & permeability) operating at higher pressure ratios may not certainly shows higher CO₂ composition in permeate stream but may possess a larger product recovery at high inlet gas CO₂ concentration due to the effect on the overall driving force promoting the CO₂ permeation.

5. Conclusion

The prospect of a polymeric membrane technique used for the separation of a mixture of two or more gases is heavily depended upon three major factors: the inlet gas composition, Process operating parameters, and the membrane characteristics used for the respective application. Thus, along with membrane metallurgy, the membrane operation performance for separating CO_2 from natural gas (in our case) is relied on membrane engineering, it includes the process design and optimization of the operating parameters. Accordingly in my work 09 cases with in interrelated operating parameters each having a different set of conditions were examined to study their effect on the removal efficiency on the membrane using the model equations.

Starting from case-1 with mass flowrate of 0.1 kg/sec and inlet gas temperature of 35°C the membrane efficiency is found to be maximum with the CO₂ removal % age of 82% and in case-2 and case-3 where the mass flowrate remain the same but the inlet gas temperature changes to 40°C and 45°C respectively and the separation efficiency drop to 79% and 76% respectively due to the effect of increased inlet gas temperature as discussed in above chapter. Similarly in case-4 to case-6, the mass flowrate changes to 0.2 kg/sec and remains same for all three cases but the temperature changes from 35°C in case-4 to 40°C in case-5 and to 45°C in case-06. Their separation efficiency is calculated to be 62%, 59% and 55% respectively. Likewise in cases from case-7 to case-9, the mass flowrate changes to 0.3 kg/sec now and remains same for all three cases but the temperature changes from 35°C in case-7 to 40°C in case-8 and to 45°C in case-09. Their separation efficiency is calculated to be 62%, 59% and 55% respectively.

Along with the feed gas composition and operating conditions, one factor that considerably altering the efficiency of the membrane element is the feed/permeate pressure ratio. The permeation number is an influential parameter for the membrane element efficiency. For a defined inlet gas flowrate, membrane nature, and pressure ratio, a lower permeation number shows lower product recovery and higher permeate composition, and reciprocal to it. The low CO_2 composition in the inlet gas does not

provide higher permeate concentration, even with higher pressure ratios, and hence require more phases to achieve the desired results. The effect of the membrane selectivity on the removal efficiency of the membrane element is insignificant at lower pressure ratios but becomes significant as it gets high. For a higher value of membrane selectivity, making the pressure ratio twice increases the

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chances of up to 2 to 3 fold higher recovery and increases

the CO_2 in permeate concentration stream.

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