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Optimization of hydrogen sulfide separation from oil-well-produced water in fiber membrane reactors with amine solvents

Abstract

The presence of acidic gases in produced water, a byproduct of oil and gas extraction, is problematic in terms ofthe corrosion of equipment and pollution of the environment. In this study, after examiningthe relevant literature, the process of treatment of produced water in fiber membrane reactors was modeled, and after validation with the existing experimental data, the model was used for prediction and process optimization purposes. The solvents considered for the process were amine as the chemical solvent and nitrogen gas as the physical solvent. After examining the variety of membranes and their performance, the best type for use in the membrane reactor was chosen. The finite element software COMSOL was used for modeling and to achieve convergence in the results. For modeling, the module's geometry was divided into three sections: membrane, inner chamber (feed flow path), and inter-membrane space, the conservation equations for mass transfer and momentum transfer were written for each section, Navier-Stokes velocity distribution was used to insert the velocity term in the momentum equation, and the resulting equations were solved simultaneously according to the boundary conditions. In this process, the walls were given the no-slip condition, and the fluidflow was assumed to remain laminar. The potential reactions between amine and hydrogen sulfide were determined from different sources, and the reaction constants, even for the reaction of hydrogen sulfide ions, were carefully considered. After choosingsuitable solvents, computational fluid dynamics modeling was conducted.

Keywords: *produced water, COMSOL, finite element software, amine solvent, hydrogen sulfide*

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Introduction

Produced water is the greatest byproduct of the oil and gas extraction process. This watercontainsa variety of organic compounds, including many toxic compounds that are hazardous for agriculture, hydrogen sulfide, and heavy hydrocarbons in low concentrations. The gases contained in produced water including carbon dioxide and hydrogen sulfide make this water unusable and cause corrosion inthe wellhead equipment. Oil and gas production facilities are typically allowed to release produced water into the environment [1]. The environmental impact of discharging produced water into sea and land is determined by the amount of organic substances and toxic pollutants contained in this water [2]. One of the majorcontaminantscontained in produced water is itsdissolved salt, which has visible effects wherever oil and gas are produced and is considered to be a major cause of pollution in these areas [3]. Given the potentially dire implications of releasing this salt into theenvironment, it is better to treat produced water before disposal. With appropriate treatment, produced water can be safer than wastewater [4]. This treatment can potentially be done with membranes.

Gas absorption membranes first appeared in 1976 in the medical industry as a means of oxygenating blood. Qi and Cussler were the first to use a membrane to remove carbon dioxide from gas streams [5]. Later,Koonaphapdeelerte et al. introduced a ceramic hollow membrane contactor for separatingcarbon dioxide from themonoethanolamine (MEA) solution at high temperatures[6].In a study by Mavroudiet al., where they investigated the change in mass transfer resistance over time in the absorption of carbon dioxide from water with apolypropylene hollow fiber membrane module, the results showed that the absorption flux significantly decreased over time;an effect that they attributed to the partial filling of the membrane pores by liquid [7].

Zhang et al. investigatedwhether it is possible to capture carbon dioxide in a hollow fiber membrane contactor with distilled water and aqueous diethanolamine (DEA)solution, under the assumption ofthe non-wetting condition for the mixture of carbon dioxide and nitrogen. In another study, Wang et al. conducted a series of laboratory tests to further explore the possibility of carbon dioxide capture in a hollow fiber membrane contactor with water and DEA solutions [8]. Yan et al. investigated the effect of gas flow rate on carbon dioxide flux when potassium glycinate and monoethanolamine are used as adsorbents. These researchers reported that increasing the gas flow rateincreased the carbon dioxide mass transfer, whereas when using water asan adsorbentthere was limited carbon dioxide mass transfer [9]. Hugheset al., Tao et al., and Tsang et al. studied whether produced water can be converted into drinking and irrigation water [10].

In a study by Dindore et al.on the absorption of carbon dioxide in propylene carbonate at high pressures using a propylene hollow fibermembrane contactor, the results showed that the use of gas-liquid membrane contactors with alkanolamines such as methyl-di-ethanolamine can be an effective approach for capturing carbon dioxide [11]. In the tests conducted by Doel et al. by using a combination of a modified polymer and activated granular carbon in a fixed-bed adsorption column, this system managed to reduce all petroleum hydrocarbons to undetectable levels and also reduced soluble hydrocarbons such as benzene, ethylbenzene, toluene, and xylene to an extent that they were hardly detectable [4].

In an experimentalstudy by Nii et al., the absorption of carbon dioxide and hydrogen sulfate in aqueous carbonate solutions using polyolefin membrane contactors was investigated [12]. In a study by Lee on the fabrication of hollow fiber membranes with a shell-less inner surface for carbon dioxide separation and absorption, the composition of the water coagulation bath hada major impact on the morphology of polyvinylidene fluoride hollow fiber membranes. [13] Ren et al. used a polyvinylidene fluoride hollow fiber membraneforcarbon dioxide removal in the membrane contactor [14]. Al Marzouqiet al. developed a mathematical model for the physical absorption of carbon dioxide in hollow fiber membrane contactors under the non-wetting condition,which showed that axial diffusion becomes important at low Peclet numbers [8]. Say et al. developed a process for the simultaneous absorption of hydrogen sulfide and carbon dioxide by using hindered amines as promotersin hot carbonate solutions. According to their study, using hindered amine in the hot carbonate process has a clear advantage over using conventional promoters such as DEA [15].

Hughes et al., Tao et al., and Tsang et al. studied the conversion of produced water into drinking and irrigation water.

In a study by Li et al., they found that it is much more efficient to remove COD from produced water by a photo-electrocatalytic process than by photocatalytic or chemical oxidation [16]. In a study by Adams et al. on the treatment of produced water where they used a cylindrical reactor with a single-pass continuous flow system in which titanium was used as a substrate and UV lamps were used for the irradiation of reactor tubes,the results showed that the hydrocarbon capacity can be decreased by more than 90% per minute [17].

Highmore and Sandal [18] investigated whether it is possible to separate carbon dioxide and hydrogen sulfide gases simultaneously using the penetration theory with DEA used as the solvent. Say et al. [18] proposed a new process in which hindered amineis used as the promoter in carbonate solutions for the simultaneous removal of carbon dioxide and hydrogen sulfide. Raskol et al. [19] used the film theory to separate the above gases. In a study by Rinker, the simultaneous absorption of carbon dioxide and hydrogen sulfide with a mixture of MDEA and MEA was investigated boththeoretically and experimentally. Al Marzouqiet al. [8] studied the separation of carbon dioxide using MEA and soda. Atchariyawutet al. [9] also used MEA and soda as the solvent for the separation of acidic gases.

Since the methods and equipment currently used for carbon dioxide absorption and removal are very energy-intensive and require large amounts of heat, many researchers have explored whether hollow fiber membranes can be used as analternative for the absorption of organic substances, carbon dioxide, and other acidic gases [7-13]. However, very few studies have been conducted on the removal of carbon dioxide with hollow fiber membranes, and to the best of our knowledge, there is still no industrial or even laboratory process for the removal of hydrogen sulfide from produced water. Considering the importance of having a reliable removal process forrecovering the solvent and in some cases purifying carbon dioxide or hydrogen sulfide, further research is needed to determine whether technologies such as hollow fiber membranes can outperformlegacy methods and equipment. It should also be mentioned that while previous studies in the field havebeen focused onthe separation of certain componentsfrom gas streams, this studyis devoted to the separation of acid gases from liquid streams.

Governing equations of the membrane separation process

In the membrane modules discussed in this study, the total radius of the module (which is the overall radius of the shell) is denoted by r_4, the shell radius is denoted by r_3,the external radius of each membrane is denoted by r_2, and the packing density of the membranes inside the moduleis denoted by$\emptyset$. In all the models previously developedfor such modules, it was assumed that the membrane is engulfed in a hypothetical shell in which the solvent can circulate. The radius of this hypothetical shell is provided by Hoppel’s theory [20] based on the assumption that the velocity on the surface of this hypothetical shell is the highest in the tubes. The highest velocity is in the middle distance between two membranes and is given by Equation (1):

$$V_{Z-Shell} = 2V\left[1-\left(\frac{r_2}{r_3}\right)^2\right]\left[\frac{\left(\frac{r}{r_3}\right)^2-\left(\frac{r_2}{r_3}\right)^2+2Ln\left(\frac{r_2}{r}\right)}{3+\left(\frac{r_4}{r_3}\right)^2-4\left(\frac{r_2}{r_3}\right)^2+4Ln\left(\frac{r_2}{r_3}\right)}\right] \tag{1}$$

$$r_3 = \frac{r_2}{\sqrt{1-\emptyset}} \tag{2}$$

The continuity equation for each component inside a reactive system can be written as follows:

$$\frac{\partial C_i}{\partial t} = -(\nabla . C_i V) - (\nabla . J_i) + R_i \qquad (3)$$

The parameters V, J_i, R_i, C_i, and tdenote the velocity, diffusion flux, reaction speed, concentration of carbon dioxide or hydrogen sulfide, and time, respectively. Considering the insignificant reaction between the solvent and the dissolved component, here, we assume the reaction speed to be zero. The continuity equation for component i inside the membrane's cylindrical coordinates can be writtenusing the steady-state Fick equation:

$$D_{i-shell}\left[\frac{\partial^2 C_{i-shell}}{\partial r^2} + \frac{1}{r}\frac{\partial C_{i-shell}}{\partial r} + \frac{\partial^2 C_{i-shell}}{\partial z^2}\right] = V_{z-shell}\frac{\partial C_{i-shell}}{\partial z}$$
(4)

Inside the shell, the velocity distribution is governed by the Navier-Stokes equation:

$$\rho\frac{\partial V}{\partial t} - \mu\nabla V + \rho(V.\nabla)V + \nabla p = 0\,;\;\; \nabla . V = 0$$
(5)

Here, ρ, p, μ, and V,denote the density, pressure, viscosity of the fluid inside the shell, and velocity,respectively.

The continuity equation for the transport of substance i across the membrane, which is assumed to occur only through diffusion, based on Fick's lawof diffusion is given by:

$$D_{i-membrane}\begin{bmatrix}\frac{\partial^2 C_{i-membrane}}{\partial r^2} + \frac{1}{r}\frac{\partial C_{i-membrane}}{\partial r} \\ + \frac{\partial^2 C_{i-membrane}}{\partial z^2}\end{bmatrix} = 0$$
(6)

In the steady state, the equation for the transport of substance i from the feed to the solvent can be written as follows:

$$D_{i-tube}\begin{bmatrix}\frac{\partial^2 C_{i-tube}}{\partial r^2} + \frac{1}{r}\frac{\partial C_{i-tube}}{\partial r} \\ + \frac{\partial^2 C_{i-tube}}{\partial z^2}\end{bmatrix} = V_{z-tube}\frac{\partial C_{i-tube}}{\partial z}$$
(7)

The velocity distribution in the tubesection can also be obtained using the Navier-Stokes equation, where u is the average velocity inside the tube. Assuming that changes in the volumetric flow rate of the feed will remain insignificant, the removal ratio for hydrogen sulfide and carbon dioxide can be obtainedfrom the following equation:

$$R\,(\%) = \frac{C_{i_{in}} - C_{i_{out}}}{C_{i_{in}}} \times 100 = \left(1 - \frac{C_{i_{out}}}{C_{i_{in}}}\right) \times 100$$
(8)

In this equation, $c_{i_{in}}$is the concentration of the substance at the inlet and $c_{i_{out}}$ is the concentration of carbon dioxide at the system's outlet.c_{out}at the shell's outlet section (Z=0) can be obtained from the following equation:

$$C_{out} = \frac{\int C(r)dA}{\int dA}$$
(9)

In the above equation, A is the feed outletarea.

Diffusion coefficients inside the membrane can be obtained by using the effective diffusion coefficient. Since it is assumed that the membrane's pores are occupied by gas, the effective diffusion coefficient can be obtained by using a modified version of the diffusion coefficient for carbon dioxide or hydrogen sulfide inside nitrogen:

$$D_{eff,H2S,CO2} = D_{H2S\text{يا}CO2} \times \frac{\epsilon}{\tau}$$
(10)

In this equation, ε is the membrane porosity and$\tau$is a parameter known as membrane curvature, whichisphysically equivalent to the ratio of the length of a pore inside the membrane to the closest distance to the two sides of that pore.

Method

The subject of this study was the separation of carbon dioxide and hydrogen sulfide from produced water using hollow fiber membranes. As shown in Figure (1), hollow fiber membranes are positioned inside a shell.The whole assembly illustrated in Figure 1 is called a hollow fiber membrane module.

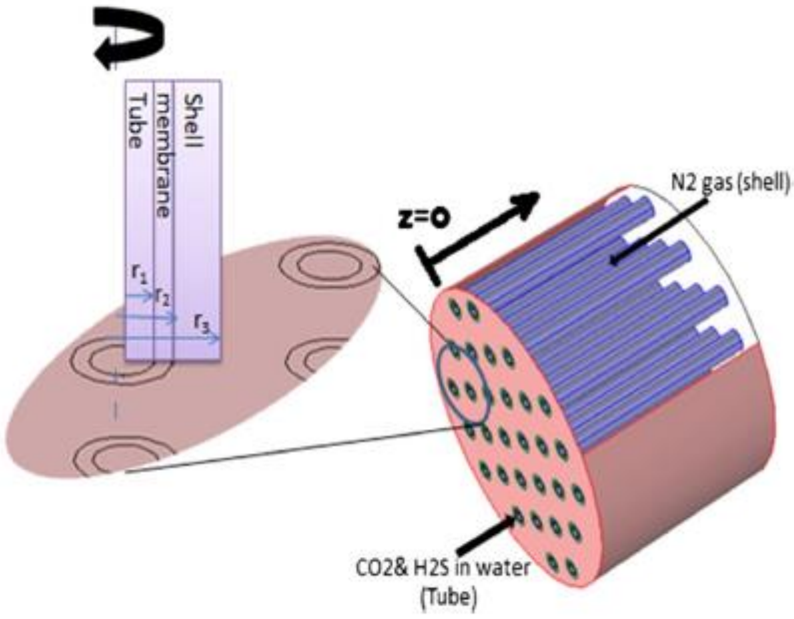


Figure 1: A view of thehollow fiber membrane module considered in the modeling process and its two-dimensional section

The module chosenfor this study is comprised of 30 hollow membranes inside a shell as shown in Figure (1). The feed given to this module isproduced water (from oil wells) containing carbon dioxide and hydrogen sulfide, which enters the membranes while the solventis passing through the space between them.

In this study, separation is done once physically by nitrogen gas and a second timechemicallyby the amine solvent. Using a physical solvent, there will be no reaction between the solvent and the dissolved components (here, carbon dioxide and hydrogen sulfide). On the contrary,using a chemical solvent like amine triggers reactions with carbon dioxide and hydrogen sulfide, which must be considered in the model. It is generally better to do theseparation withphysical solvents -also called sweeper gases- as they do not react with the substances to be separated.

The solvent gas enters the shell section (the space between the tubes) while the feed consisting of hydrogen sulfide and carbon dioxide in water circulates in the space inside the membranes.

In this study, the module wasmodeled not only as a single assemblybut also using an oldermodeling method, which involvesmodeling a membrane outside the module and generalizing the results to the whole module. In other words, modeling wasperformedtwice: one timeby modeling all 30 membranes together, and a second timeby modeling oneofthese 30 membranes and attributing the results toallmembranes (the older modeling method). This was done to compare the accuracy of old modeling methodsversus new ones.

The mass transfer equations for all three sections along the length of the contactor were solved by applying the boundary conditions with thesoftware COMSOL version 4.2, which uses the finite element method to achieve convergence. The parameter valuesused in the solution process are given in Table (1).

Table 1: Specifications of the hollow fiber membrane module in the models

Value	**Variable**
0.95	The outer diameter of the membrane (mm)
0.6	The inner diameter of the membrane (mm)
14	The inner diameter of the module (mm)

582	The density of membranes inside the module (m^2/m^3)
270	Length of the module
30	Number of membranes in the module
0.77	Porosity
298	Temperature (°K)

Results

This section presents and discusses the results obtained from the three-dimensional model of the hollow fiber membrane for the simultaneous removal of hydrogen sulfide and carbon dioxide with pure nitrogen as solvent. Sinceit was found that using higher solventvelocitiesand lower feed velocities increases the system's efficiency, in practice, it is better to use highersolvent-to-feedvelocity ratios. Sincethis efficiency improvement is more a function of the reduced feed velocity than the increased gas velocity, it is better to increase the said ratio by reducing the feed velocity.

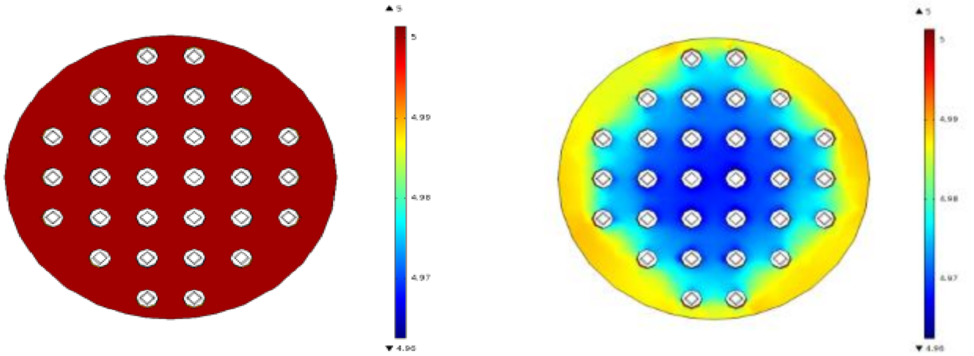
Although reducing the feed velocity will reduce the feed flow rate, the effectcan be offset by increasing the number of membranes. Considering the negligible efficiency impactof the amount of carbon dioxide in the inlet, this system can be used to treat streams that contain large amounts of hydrogen sulfide and carbon dioxide.

In the models, amine concentration was considered to be 4 mol/m^3, and the concentration of carbon dioxide and hydrogen sulfide was also assumed to be4 mol/m^3. Table (2) shows the values of constants in the solution process, which werederived from relevant references. It should also bementioned that the dimensions of the membrane and the type and dimensions of the module for this separationwere the same asfor nitrogen separation:

Table 2: Constants and parameters needed for the numerical solution of the equations

Source reference	Value	Variable/Constant
45	1.8×10^{-5}	$D_{CO2\text{-shell}}$ (m^2s^{-1})
45	1.51×10^{-9}	$D_{CO2\text{-tube}}$ (m^2s^{-1})
45	$D_{CO2\text{-tube}}(\frac{\epsilon}{\tau})$	$D_{CO2\text{-membrane}}$ (m^2s^{-1})
45	2.01×10^{-5}	$D_{H2S\text{-shell}}$ (m^2s^{-1})
45	1.52×10^{-9}	$D_{H2S\text{-tube}}$ (m^2s^{-1})
45	$D_{H2S\text{-tube}}(\frac{\epsilon}{\tau})$	$D_{H2S\text{-membrane}}$ (m^2s^{-1})
45	0.8	m_{CO2} (mol mol^{-1})
45	2.3	m_{H2S} (mol mol^{-1})

Inside the membrane, solvent and feed circulate in the opposite direction with the pressure on the sides of the membrane kept constant,resulting in anisothermsteady process. As shown in Figure (2), amine enters the shell sectionwith a concentration of 4 mol/$m^3$and,as the colored bands indicate, has the highest concentration at the inlet part of the shell.As Figure (3) shows, the concentration of amine decreases as it moves toward the shell's outlet whilebeing consumed in the reaction. The amine concentration distribution obtained at the outlet is indicative ofhow amine has reacted differentlywith dissolved components in different parts of the shell. This is because of the higher concentration of the dissolved reactant in the areas near the membranes, which results in higher reaction speeds in these areas. Ascan be seen, higher reaction speed near the membranes has led toincreasedamine consumption and decreased amine concentration at the outlet. It should be noted that this analysis is for asolvent velocity of 0.1 m/s and a feed velocity of 0.5 m/s. The impacts of solvent andfeed velocities are discussed later in the paper.



a b

Figure 2: a)Amine concentration distribution at the shell'sinletfor an initial concentration of 5 mol/m^3
b)mine concentration distribution at the shell's outletfor an initial concentration of 5 mol/m^3

In Figure (2a), the red color means that the amine concentration is the highest possible across the shell's inlet. The disappearance of this red color in Figure (2b) indicatesanamine concentration reduction at the outlet,which is strong in areas near the membranes and weak in other areas.

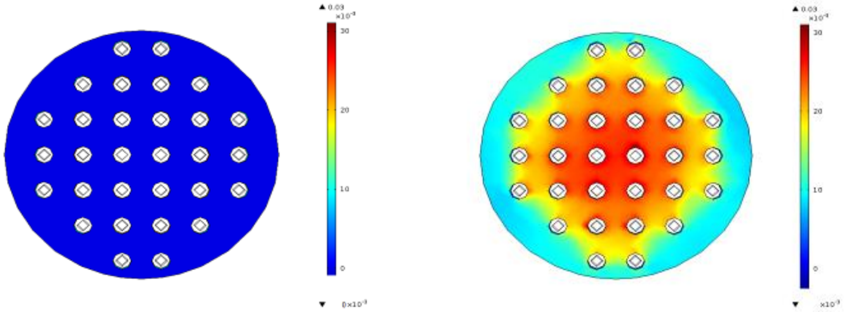


Figure 3: a)Amine cation concentration distribution at the shell's inlet,b)Amine cation concentration distribution at the shell's outlet (due to reactions)

In Figure (3a), whichshows the concentration of amine cation at the inlet,the entire section is blue because amine is given to the system in a pure form and thus the amine cation concentration is zero across the section. Figure (3b) shows how amine cations have appeared at the outlet,which is because of how amine has reactedwith dissolved components afterarrival in pure formas indicated in the previous figure. A similar trend can be observed for dissolved components,in thesense that there has been an increase in theconcentration of reaction products and a decrease in the concentration of dissolved components with a distribution that is completely dependent on proximity to the membranes inside the shell.

Figures (4) show the distribution of the concentration of reactive components at the inlet and outlet of the hollow fiber membranemodule.

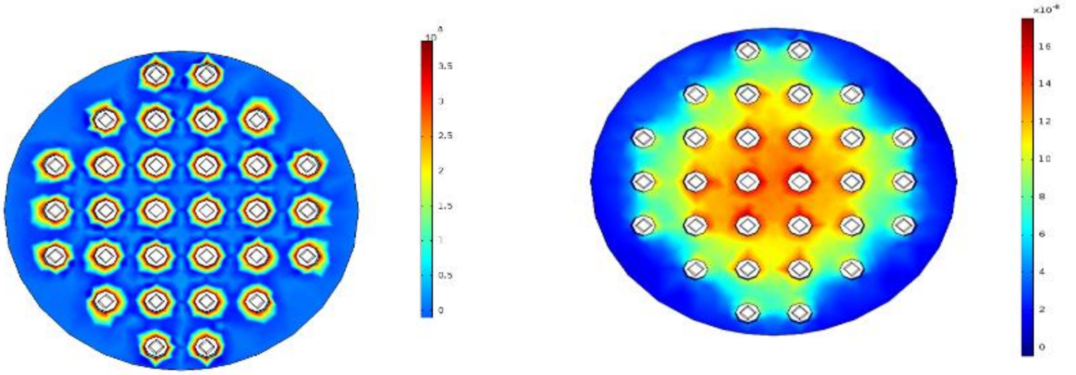


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Figure 4: a)Carbon dioxide concentration distribution at the shell's outlet for an initial feed concentration of 4 mol/m^3,b)ydrogen sulfide concentration distribution at the shell's outlet for an initial feed concentration of 4 mol/m^3

Figures 4 show that while the concentration of carbon dioxide inside the shell is limited to the areas in the immediate vicinity of the membranes, for hydrogen sulfide, the concentration is spread over the shell and hasa broader gradient. This can be attributed to the higher solubility of hydrogen sulfide inside amine and also its higher diffusion coefficient inside amine, which allows it to spread more easily in the medium. This is while carbon dioxide has lower solubility and diffusion, thus quickly reactingwithand being swept away by the solvent. (Figure 5)

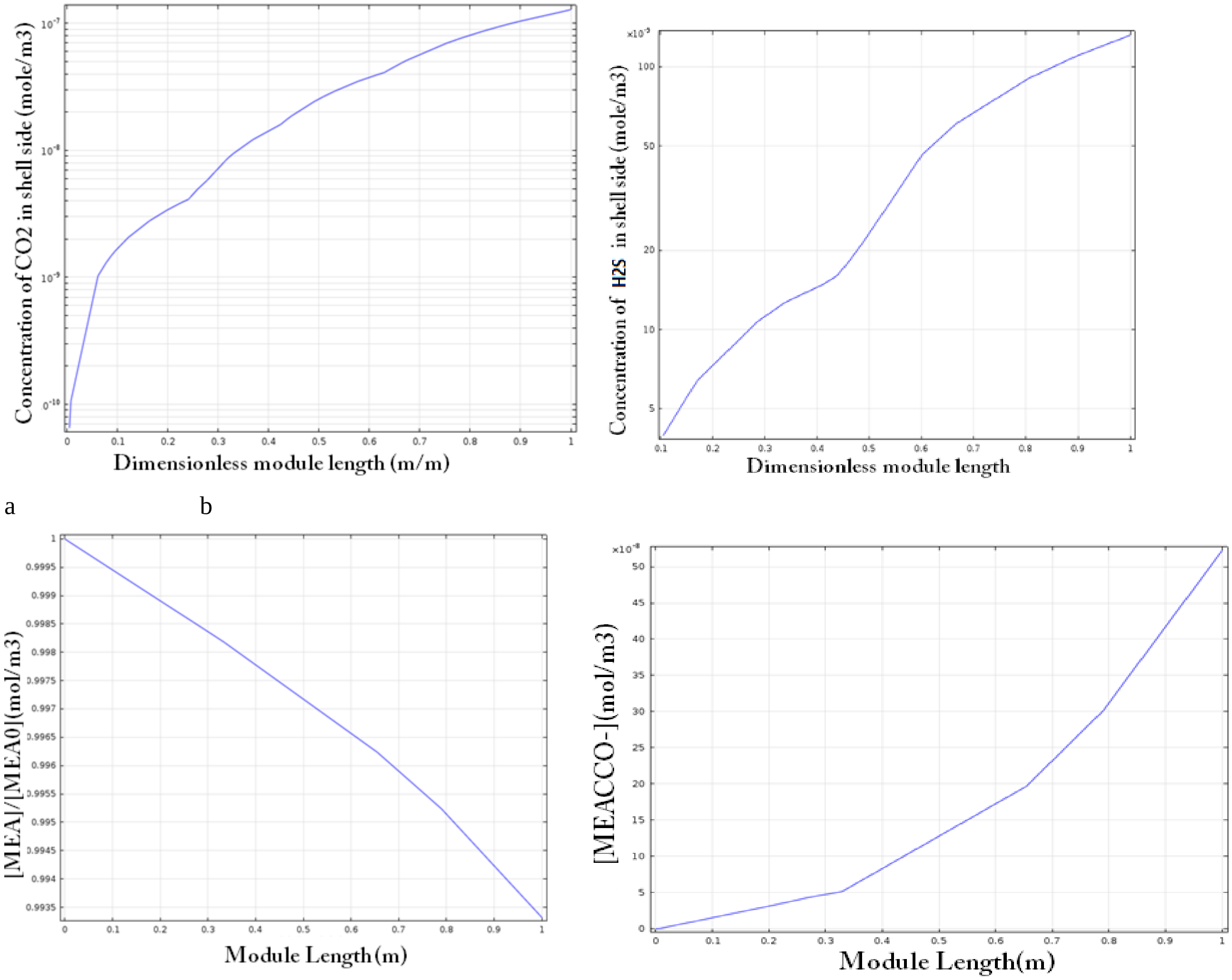

Figure 5: a)Change in carbon dioxide concentration along the module, b)Change in hydrogen sulfide concentration along the module, c) Change in Amine concentration along the module,d)Change in amine cation concentration along the module,e)Change in amine anion concentration along the module

Figures5 show the variations in the concentration of reactants and some products along the length of the module. In these diagrams, the horizontal axis represents the dimensionless length, obtained by dividing the coordinates of each point by the total module length, andthe vertical axis represents the dimensionless concentration of the substance, obtained by dividing the concentration of the substance at the point by its input concentration.

To determine the impact of solvent and feed velocities on the removal efficiency, different feed velocities and different amine velocities were defined for the model and then asurface integral was taken from the module's outlet cross-section.

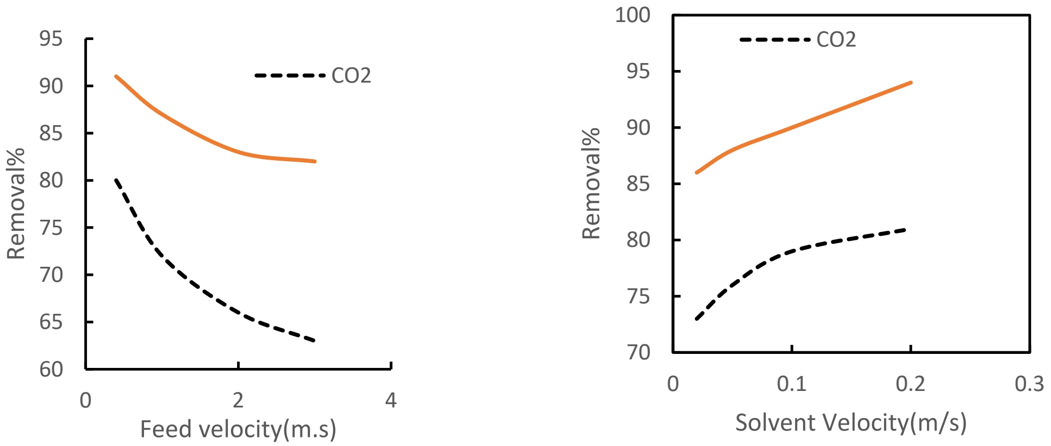

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Figure 6: a)Dissolved component removal ratefordifferent feedvelocities at asolvent velocity of 0.1 m/s, b)Dissolved component removal rate for different solvent velocities at a feed velocity of 0.5 m/s

As shown in Figure6 , the removal rate increased with the increase in solvent velocity and decreasedwith the increase in feed velocity.This is similar to what can be expected from a physical solvent,but this removal rate is much higher than whatcan be achieved with a physical solvent. Clearly, this is becausethe chemical reaction between the dissolved component that appears in the shell and the solvent in the shell has reduced the concentration of the dissolved component in the shell.

The decline in the removal efficiency with the increase in feed velocity in Figure (6a) is due to reducedfeed-membrane proximity and the shorter time the membrane has to remove dissolved components from the feed.This is because in the inner parts of the membranes where the feed flows, the concentration of dissolved components is always high and there is no concentration boundary layer in the sense that it is in the shell section. In other words, the feed is considered a solution acting as the source of dissolved components, the required amount of which is the same at all flow points inside the membrane.

This reduced concentration in the shell causes a greater mass transfer gradient and a higher removal ratio. Given the faster reaction of hydrogen sulfide with amine, this gradient is higher for hydrogen sulfide than for carbon dioxide,resulting in a higher removal ratio for this substance. The increase in the removal ratio with the increase in solvent velocity in Figure (6b) is due to the decreasing thickness of the concentration and fluid boundary layer created in the outer body of the membranes.

Conclusion

In this study, a three-dimensional model of the simultaneous removal of hydrogen sulfide and carbon dioxide using a hollow fiber membrane module with physical and chemical solvents was developed and used toexamine this process. When using nitrogen as the physical solvent, it was found thatusing high solvent velocities and low feed velocities increases the system's efficiency. Thus, it is better to use higher solvent-to-feedvelocity ratios. Since themodule's efficiency withnitrogen as solventdepends more on feed velocity than on gas velocity, it is preferable to increase the solvent-to-feedvelocity ratio by reducing the feed velocity.While decreasing the feed velocity will reduce the feed flow rate, the effect can be offset byincreasing the density of the membranes (using more membranes in one shell) or linking multiple modules in parallel. Considering the negligible efficiency impact of increasing the carbon dioxide load, this system can be used for streams that contain large amounts of hydrogen sulfide and carbon dioxide.

For the physical solvent, the results showed that the feed temperature change at different solvent and feed velocitiesare quasi-linear functions. The removal ratio increasedas the temperature increased from 25°C to 45°C, whichcan be attributed to the change in solubility and diffusion coefficients. This function is ascending, meaning that a higher removal ratio can be achieved at higher temperatures.

For amine as the chemical solvent, the results showed that it can yield a higher removal ratiothan the physical solvent.With this solvent, theremoval efficiency for hydrogen sulfide at normal temperature was as high as95% under optimal conditions. Given the higher solubility and reaction speed of hydrogen sulfide in amine compared to carbon dioxide, the removal ratio was higher for hydrogen sulfide than forcarbon dioxide. With the chemical solvent, the concentration at the module's outlet was more distributed across the outlet cross-section. This is expectable since chemical removal is typically moreefficient than physical removal.

Considering the positive effect of temperatures on the removal process, it is recommended to conduct the process at high temperatures as much as possible to improvesolubility as well as diffusion. Since reducing the feed velocity increases the efficiency, it might be better to use modules inparallel or use modules with a large number of membranes to avoid a declinein the feed flow rate throughout the process. In future studies, the economic viabilityofseparating such components from produced water with physical and chemical solvents needs to be carefully analyzed.

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