Separation of Bulk Hydrogen Sulfide–Hydrogen Mixtures by Selective Surface Flow Membrane

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The separation performance of binary hydrogen sulfide–hydrogen mixtures by a nanoporous carbon membrane called selective surface flow (SSF) membrane is described. The membrane selectively permeates H_2S from H_2, and a H_2-enriched stream is produced at the feed-gas pressure. A two-stage embodiment of the SSF membrane is described for production of high-purity H_2 with high H_2 recovery from an equimolar H_2S-H_2 feed gas. A novel protocol for operation of the two-stage membrane is needed to achieve that separation goal.

Introduction

A nanoporous carbon membrane called Selective Surface Flow (SSF) membrane has been developed by Air Products and Chemicals, Inc. (Rao et al., 1992; Rao and Sircar, 1993). It consists of a thin layer (2–3 μm) of a nanoporous (6–7 Å pore diameter) carbon matrix supported on a macroporous (<1-μm pore diameter) tubular alumina support. The pore-size distribution of the membrane was very narrow as shown by CH_4 diffusivity measurements (Rao and Sircar, 1996). It is produced by coating the bore size of the tubular support with a polyvinylidene chloride latex film and then carbonizing the polymer by heating in an inert atmosphere. Only one coat of the polymer and a single heating step are used. The details of the carbonizing conditions and pore characteristics of the membrane are given elsewhere (Rao et al., 1992; Rao and Sircar, 1993, 1996; Anand et al., 1997). The pores of the alumina support are much larger than the pores of the carbon layer, and thus the support pores do not influence the separation characteristics of the SSF membrane.

The membrane separates gas mixtures by an adsorption–surface diffusion–desorption mechanism (Rao and Sircar, 1993). The gas mixture to be separated is passed through the bore side of the membrane at a superambient pressure level (P^H) while keeping the permeate side at a near-ambient pressure level (P^L). Certain components of the feed-gas mixture are selectively adsorbed on the pore walls of the carbon membrane, followed by their selective surface diffusion on the pore walls to the low-pressure side where they are desorbed into the permeate gas phase. Consequently, the membrane produces a gas stream enriched in the less selectively adsorbed components of the feed-gas mixture as the high-pressure effluent gas, while a gas stream enriched in the more selectively adsorbed components of the feed gas is produced as the low pressure effluent gas. This is often very desirable because the less selectively adsorbed components form the desired product gas in many practical applications.

Adsorbates having larger molecular size, higher polarizability, and larger permanent polarity are selectively adsorbed on the membrane walls and they selectively permeate through the membrane. This is in contrast with the characteristics of most polymeric membranes where the smaller molecules of a gas mixture preferentially permeate through the membranes by a solution–diffusion mechanism. The separation mechanism of the SSF membrane provides high permeance with high permselectivities of the adsorbed components while requiring relatively low feed-gas pressures (0.24–1.5 MPa). Typical polymeric membranes, on the other hand, exhibit a reciprocal relationship between permeance and permselectivity of the permeated gas, and they require relatively larger feed-gas pressures (>1.5 MPa) for practical operation. These advantages of the SSF membrane have led to many separation concepts of practical interest, such as hydrogen recovery from refinery and chemical-waste gases and fractionation of hydrocarbon mixtures (Rao et al., 1995; Anand et al., 1995).

The purpose of the present work is to report on the separation characteristics of bulk H_2S-H_2 mixtures under various operating conditions using the SSF membrane and to describe a novel mode of operation of the membrane system for enhancing the H_2S-H_2 separation.

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Hydrogen sulfide is selectively adsorbed and permeated through the SSF membrane because (a) it has a large permanent dipole moment (0.97×10^{-18} esu), while the hydrogen has a weak quadrupole moment (0.66×10^{-26} esu·cm^2) (Stogryn and Stogryn, 1966; Weast et al., 1978), and (b) the polarizability of H_2S (37.8×10^{-25} cm^3) is much larger than that of H_2 (7.9×10^{-25} cm^3) (Hirschfelder et al., 1954). The kinetic diameters of H_2S and H_2 are, respectively, 3.60 and 2.92 Å (Breck, 1973).

Experimental Methods and Data Analysis

Figure 1 is a diagram of the apparatus used for evaluating the separation performance of binary H_2S-H_2 mixtures by the SSF membrane. A single tube of the SSF membrane (30 cm long, 0.56-cm internal diameter, 0.165-cm-thick support wall) was mounted in a cylindrical membrane holder (2.3-cm ID and 40.0 cm long). The holder was fitted with a high-pressure gas inlet and effluent outlet lines in the bore side of the membrane as well as with a low-pressure effluent outlet line in the permeate side. The feed-gas flow rate into the membrane at pressure P^H and the flow rates of both the high- and low-pressure effluent gases were measured using mass flowmeters (MFM). The low-pressure effluent gas was withdrawn in a direction countercurrent to that of the feed-gas flow. The feed-gas mixtures were premixed. The effluent-gas compositions were measured by a gas chromatograph (GC). A back-pressure regulator was used at the high-pressure effluent line to maintain a constant value of P^H. The low-pressure side of the membrane was maintained at a constant value of ~ 0.11 MPa ($= P^L$) for all tests. The system temperature was ambient (25 ± 3°C). All effluent gases from the membrane were passed through an aqueous–alkali solution in order to remove the H_2S before venting.

The experiments were conducted at a feed-gas pressure of 0.79 MPa using two different feed-gas mole fractions of H_2S ($y^F_{H_2S} = 0.25, 0.50$). The effluent gas flow rates and compositions were measured after steady-state operations were established, which typically took 0.5 h of operation.

The partial pressures of H_2S and H_2 (both high- and low-pressure sides) vary along the length of the membrane under steady-state operation. Consequently, the amounts of H_2S and H_2 adsorbed on the membrane pore walls at both high- and low-pressure sides as well as their surface diffusivities vary along the membrane length. As a result, the H_2S permeance and permselectivity are functions of the membrane length. These functionalities are not known. We therefore chose to describe the separation performance of the SSF membrane in terms of overall H_2 recovery (α_{H_2}) and H_2S rejection (β_{H_2S}). The H_2 recovery is defined by the ratio of the molar flow rates of H_2 in the high-pressure effluent gas stream to that in the feed-gas stream. The H_2S rejection is defined by the ratio of the molar flow rates of H_2S in the low-pressure effluent-gas stream to that in the feed-gas stream.

The separation performance data will be given in terms of β_{H_2S} as functions of α_{H_2} for different feed-gas conditions. High rejections of H_2S at high recoveries of H_2 are the desired properties. This mode of description of the separation performance of the SSF membrane also represents the practical separation characteristics by the membrane.

All gas flow rates and compositions for the membrane system can be expressed in terms of the variables α_{H_2}, β_{H_2S}, the feed-gas flow rate (F), and $y^F_{H_2}$ by carrying out mass balances.

High-pressure-side effluent

$$\frac{P}{F} = \sum y_i^F(1-\beta_i); \quad y_i^P = \frac{y_i^F(1-\beta_i)}{\left[\sum y_i^F(1-\beta_i)\right]} \tag{1}$$

Low-pressure-side effluent

$$\frac{W}{F} = 1-\sum y_i^F(1-\beta_i); \quad y_i^W = \frac{y_i^F\beta_i}{\left[1-\sum y_i^F(1-\beta_i)\right]}, \tag{2}$$

where P and y_i^P, are, respectively the molar flow rate and mole fraction of component i in the high-pressure effluent; W and y_i^W are the corresponding properties of the low-pressure effluent. It should be noted that ($\alpha_i + \beta_i = 1$). The summations in Eqs. 1 and 2 are over both components ($i = 1, 2$).

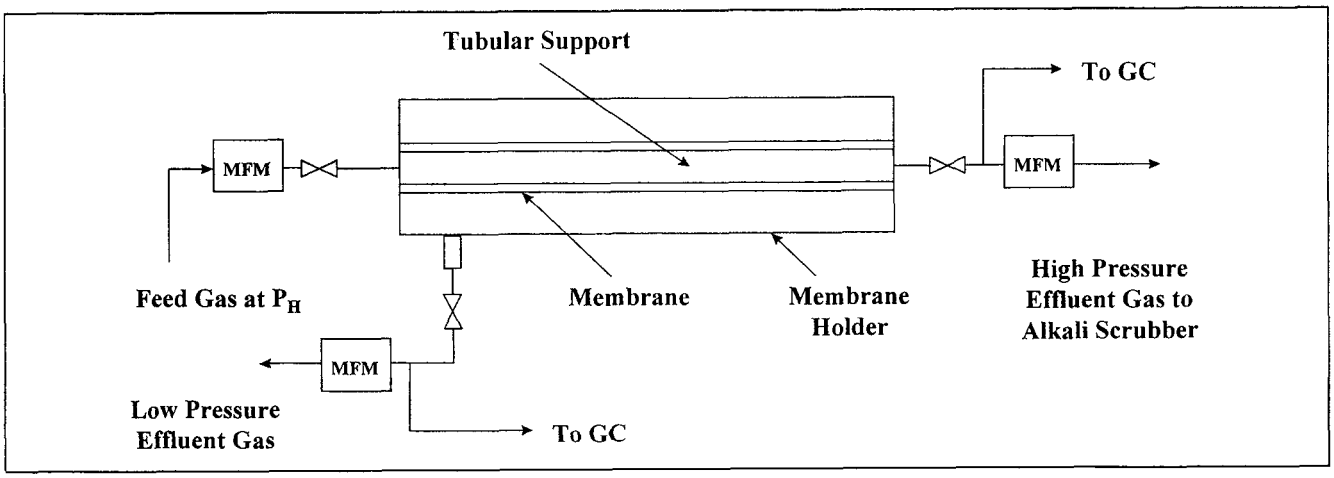

Figure 1. Experimental apparatus.

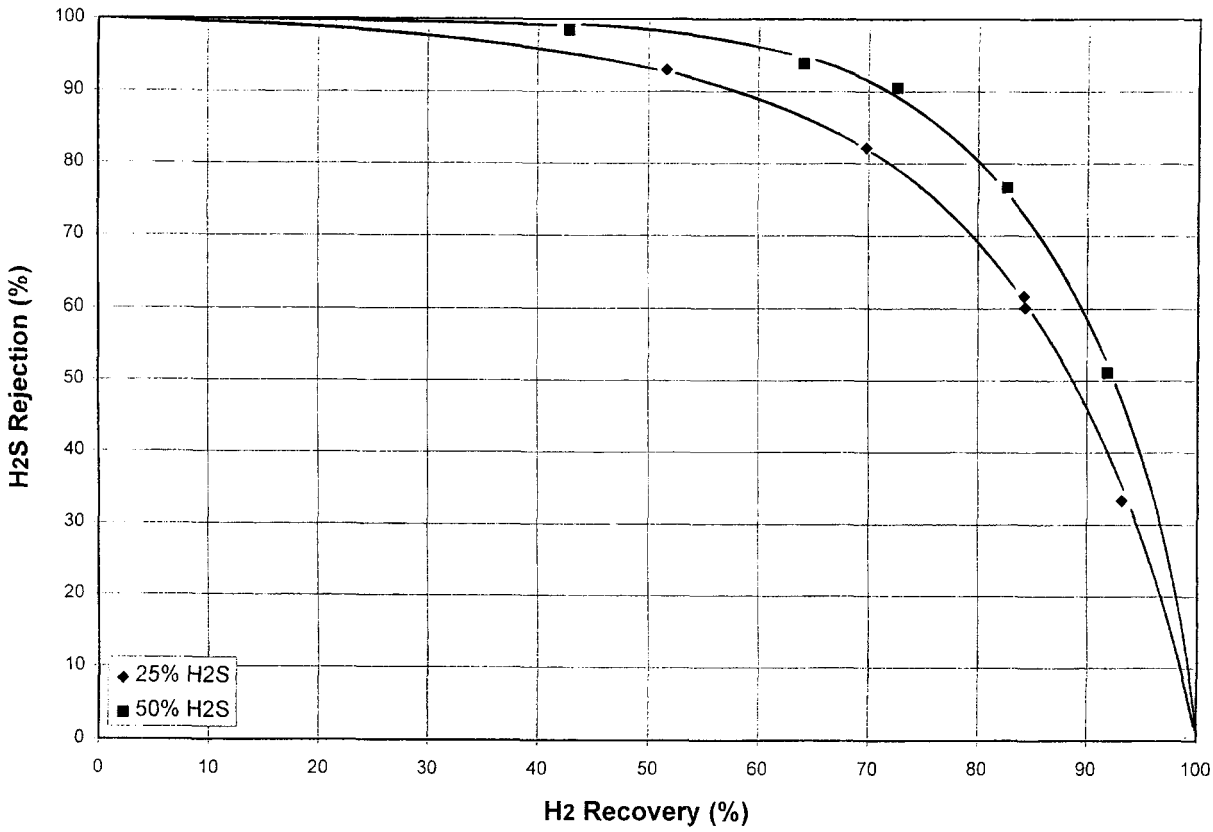

Figure 2. H_2S rejection by the SSF membrane as functions of H_2 recovery.

Experimental Results

Figure 2 shows the H_2S rejection (β_{H_2S}) against hydrogen recovery (α_{H_2}) plots measured in our laboratory. The feed-gas flow rate (F) was varied at constant values of P^H and $y^F_{H_2S}$ in order to generate these performance curves. The overall and component mass balances closed within $\pm 3\%$ for each experimental run and the data were very reproducible. The curves of Figure 2 are bounded by the limits ($\alpha_{H_2} = 0$, $\beta_{H_2S} = 100$) and ($\alpha_{H_2} = 100$, $\beta_{H_2S} = 0$), and a straight line through these limiting points represents the case of no separation.

It may be seen from Figure 2 that H_2S is preferentially permeated through the SSF membrane and the separation of bulk H_2S from mixtures with H_2 improves as the partial pressure of H_2S in the feed gas ($p^F_{H_2S}$) is increased. An identical result was previously reported for separation of bulk CO_2-H_2 mixture by the SSF membrane (Paranjape et al., 1997). Figure 2 also shows that a very high rejection of H_2S at a high H_2 recovery can be achieved when the feed gas H_2S mole fraction is large ($y^F_{H_2S} \sim 0.5$). For example, the H_2S rejections from a feed gas at 0.79 MPa containing 50% H_2S are, respectively, 95+% and 80% at H_2 recoveries of 60% and 80%. The H_2S rejection can be increased to more than 98+% if an H_2 recovery of less than 50% is acceptable. These figures demonstrate that the SSF membrane is very efficient for separation of bulk H_2S-H_2 mixtures even at moderate feed-gas pressures.

Empirical Data Representation

In absence of binary adsorption equilibrium and surface diffusion data for the H_2S-H_2 mixture on the SSF membrane, we empirically describe the rejection-recovery plots of Figure 2 by the following relationship:

$$\beta_{H_2S} = \frac{\psi(1-\alpha_{H_2})}{1+(\psi-1)(1-\alpha_{H_2})}, \tag{3}$$

where ψ is an empirical parameter. It is a function of the partial pressure of H_2S ($p^F_{H_2S}$) in the feed gas. The value of ψ is greater than unity for any separation to occur [$\beta_{H_2S} + \alpha_{H_2} = 1$ for $\psi = 1$]. The higher the value of ψ, the better the separation of H_2S from H_2. Equation 3 exhibits the appropriate boundary conditions for the rejection–recovery plots.

Figure 3 shows the fit of the rejection–recovery data of Figure 2 by Eq. 3. The values of ψ for different feed-gas mole fractions of H_2S are given in the figure. Equation 3 describes the experimental data fairly well. Figure 4 shows that a linear empirical correlation exists between the variables ψ and $p^F_{H_2S}$ over a large range of $p^F_{H_2S}$ values. The line intersects the ordinate ($p^F_{H_2S} = 0$) at a point where ψ is unity (as required). A linear correlation was also observed between the parameter ψ and the feed-gas partial pressures of CO_2. ($p^F_{CO_2}$) for separation of binary CO_2-H_2 mixtures by the SSF membrane (Paranjape et al., 1997). A much larger database was available for that case. The dashed line in Figure 4 shows the $\psi - (p^F_{CO_2})$ plot for comparison. It may be seen that the slope of the $\psi - p^F_{H_2S}$ plot is about three times larger than the slope of the corresponding plot for CO_2-H_2 separation. Thus, the SSF membrane is more efficient for the separation

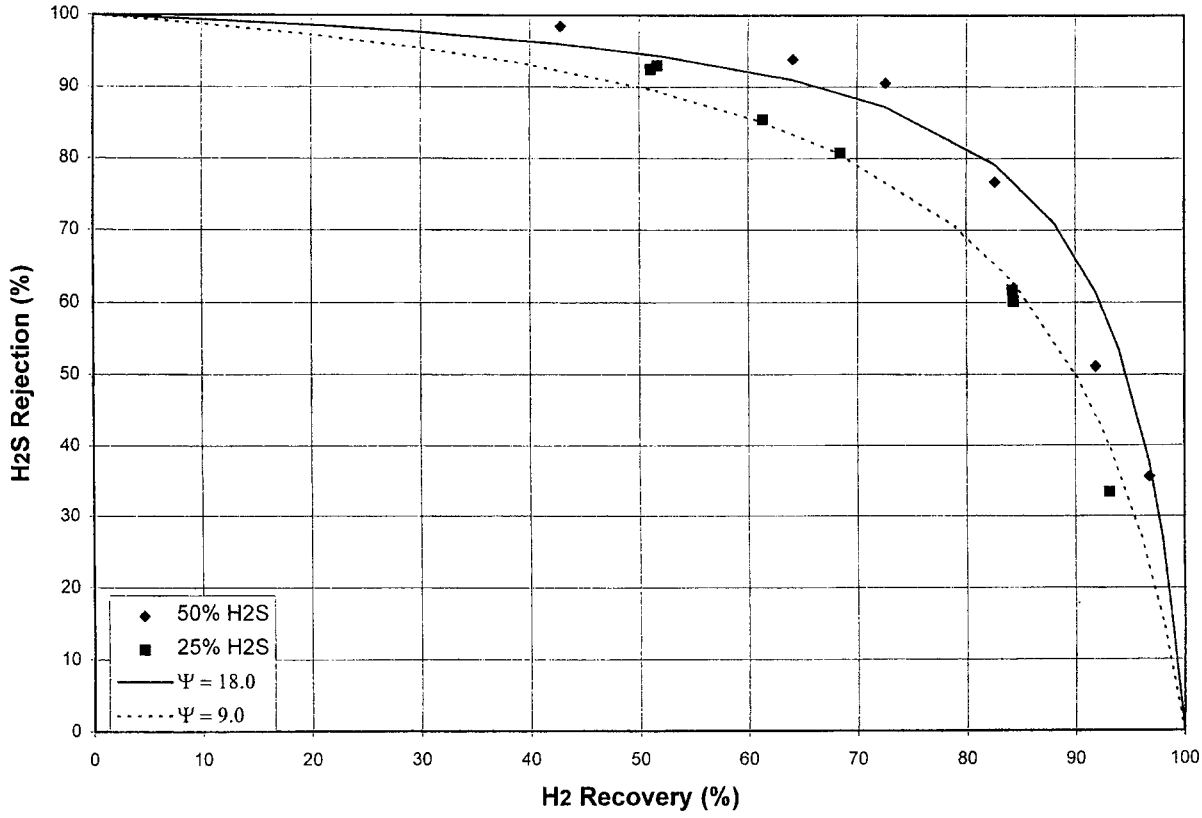

Figure 3. Fit of data of Figure 2 by Eq. 3.

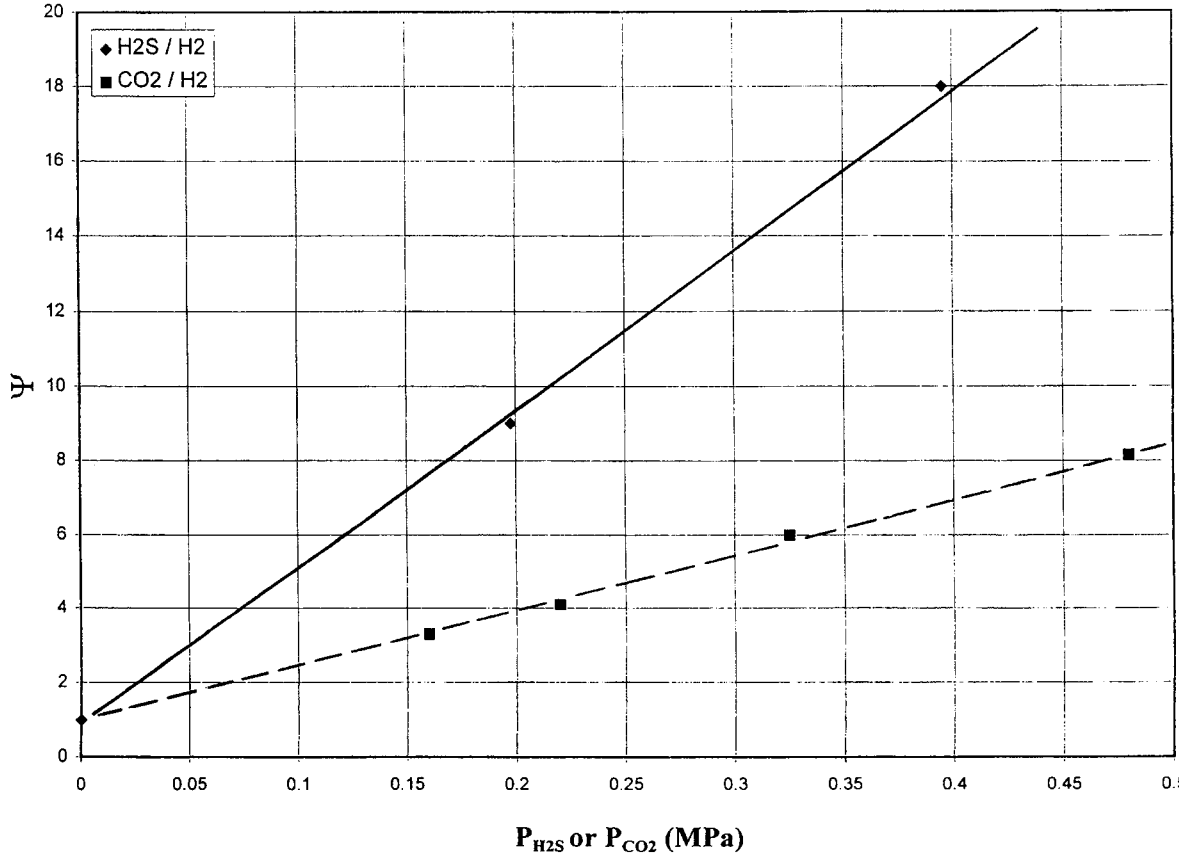

Figure 4. Dependence of empirical parameter (ψ) on feed-gas partial pressures of H_2S ($p^F_{H_2S}$) or CO_2 ($p^F_{CO_2}$).

of bulk H_2S-H_2 mixtures than that for CO_2-H_2 mixtures. This is caused by the larger affinity of adsorption of H_2S on the nanoporous carbon walls than CO_2.

Novel Mode of Operation of SSF Membrane

The separation goal is to produce a highly enriched stream of H_2 at feed-gas pressure with high H_2 recovery from a bulk H_2S-H_2 mixture by selectively rejecting H_2S through the SSF membrane. We found that a two-stage embodiment of the SSF membrane described by Figure 5 can achieve that goal when it is operated using a specific protocol (Sircar and Parrillo, 1997).

The feed gas is introduced into the high-pressure side of the stage 1 of the membrane assembly at pressure P^{H1} and a H_2-enriched product gas is withdrawn from that membrane at essentially the feed-gas pressure. The H_2S-enriched permeated gas is withdrawn countercurrently from the low-pressure side of the stage 1 membrane at pressure P^L. It is then compressed to a pressure level of P^{H2} ($\leq P^{H1}$) and introduced into the feed side of stage 2 of the membrane assembly. The high-pressure effluent gas from the stage 2 membrane is recompressed to P^{H1} and reintroduced as feed gas to the stage 1 membrane by mixing it with the fresh feed gas. The low-pressure effluent from stage 2 membrane at pressure p^L is countercurrently withdrawn as the H_2S enriched product gas. Neither stage uses any purge gas at the low-pressure side. This configuration of a two-stage membrane is well known (Spillman, 1989). However, it will be demonstrated later that a specific mode of operation of the two-stage membrane can simultaneously provide high recovery and purity of the H_2 product.

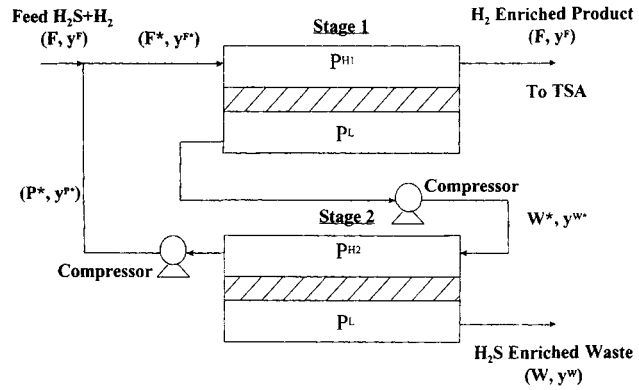

Figure 5. Flowsheet for two-stage operation of the SSF membrane.

The variables F, F^*, P, W^*, P^*, and W represent the molar flow rates of various streams entering and leaving the two stages of the membrane assembly as depicted by Figure 5. The variables y^F, y^{F^*}, y^P, y^{W^*}, y^{P^*}, and y^W represent the mole fractions of H_2S in the corresponding streams. We designate the H_2 recoveries of membrane stages 1 and 2 by the variables α_1 and α_2, respectively, and the H_2S rejections of the membrane stages 1 and 2 by the variables β_1 and β_2, respectively. Equations 1 and 2 can then be combined with appropriate mass-balance equations for the membrane stages to obtain analytical expressions describing the flow rate and composition of each stream of Figure 5 in terms of feed-gas flow rate (F), the feed-gas composition (y^F) and the variables α_1, α_2, β_1, and β_2. They are summarized below.

Flow rates

$$F^* = \frac{F[1-(1-\alpha_1)\alpha_2 y^F - \beta_1(1-\beta_2)(1-y^F)]}{[1-\beta_1(1-\beta_2)][1-\alpha_2(1-\alpha_1)]}$$

$$P^* = \frac{F[\beta_1(1-\beta_2)\{1-\alpha_2(1-\alpha_1)\}y^F + \alpha_2(1-\alpha_1)\{1-\beta_1(1-\beta_2)\}(1-y^F)]}{[1-\beta_1(1-\beta_2)][1-\alpha_2(1-\alpha_1)]}$$

$$W^* = \frac{F[1-\{1-\beta_1[1-\alpha_2(1-\alpha_3)]\}y^F - \{\alpha_1 + \beta_1(1-\beta_2)(1-\alpha_1)\}(1-y^F)]}{[1-\beta_1(1-\beta_2)][1-\alpha_2(1-\alpha_1)]}$$

$$P = \frac{F[(1-\beta_1)\{1-\alpha_2(1-\alpha_1)\}y^F + \alpha_1\{1-\beta_1(1-\beta_2)\}(1-y^F)]}{[1-\beta_1(1-\beta_2)][1-\alpha_2(1-\alpha_1)]}$$

$$W = \frac{F[1-\{1-\beta_1\beta_2[1-\alpha_2(1-\alpha_1)]\}y^F - \{\beta_1(1-\beta_2)+[1-\beta_1(1-\beta_2)][\alpha_1+\alpha_2-\alpha_1\alpha_2]\}(1-y^F)]}{[1-\beta_1(1-\beta_2)][1-\alpha_2(1-\alpha_1)]}.$$

Compositions

$$y^{F^*} = \frac{y^F[1-(1-\alpha_1)\alpha_2]}{[1-(1-\alpha_1)\alpha_2 y^F - \beta_1(1-\beta_2)(1-y^F)]}$$

$$y^{P^*} = \frac{y^F[1-\alpha_2(1-\alpha_1)][\beta_1(1-\beta_2)]}{[\beta_1(1-\beta_2)\{1-\alpha_2(1-\alpha_1)\}y^F + \alpha_2(1-\alpha_1)\{1-\beta_1(1-\beta_2)\}(1-y^F)]}$$

$$y^{W*} = \frac{y^F(\beta_1)[1-\alpha_2(1-\alpha_1)]}{[1-\{1-\beta_1[1-\alpha_2(1-\alpha_1)]\}y^F - \{\alpha_1+\beta_1(1-\beta_2)(1-\alpha_1)\}(1-y^F)]}$$

$$y^P = \frac{y^F(1-\beta_1)[1-\alpha_2(1-\alpha_1)]}{[(1-\beta_1)\{1+\alpha_2(1-\alpha_1)\}y^F + \alpha_1\{1-\beta_1(1-\beta_2)\}(1-y^F)]}$$

$$y^W = \frac{y^F\beta_1\beta_2[1-\alpha_2(1-\alpha_1)]}{[1-\{1-\beta_1\beta_2[1-\alpha_2(1-\alpha_1)]\}y^F - \{\beta_1(1-\beta_2)+[1-\beta_1(1-\beta_2)][\alpha_1+\alpha_2-\alpha_1\alpha_2]\}(1-y^F)]}.$$

It can also be shown that the overall H_2 recovery [$\alpha = P(1-y^P)/F(1-y^F)$] and H_2S rejection [$\beta = Wy^W/Fy^F$)] by the membrane assembly of Figure 5 are given by:

$$\alpha = \frac{\alpha_1}{1-(1-\alpha_1)\alpha_2} \tag{4}$$

$$\beta = \frac{\beta_1\beta_2}{1-\beta_1(1-\beta_2)}. \tag{5}$$

By choosing a set of values for the variables F, y^F, α_1, α_2, P^{H1}, and P^{H2}, and by guessing the values of parameters of y^{F*} ($< y^F$) and y^{W*} ($> y^F$), one can estimate the values of parameters ψ_1, ψ_2, β_1, and β_2 by using Eq. 3 and Figure 4. The analytical equations just listed can then be used to calculate y^{F*} and y^{W*}, and the process repeated until convergence between the chosen and calculated values of these two variables is obtained. The converged values of ψ_1, ψ_2, β_1, and β_2 can then be used to calculate the flow rates and compositions of all streams as well as the overall H_2 recovery (Eq. 4) and the overall H_2S rejection (Eq. 5) by the membrane system. Alternatively, the two-stage membrane can be operated using constant partial pressures ($p^F_{H_2S}$) of H_2S in the feed gas for both stages (F^* and W^*). Thus, ($\psi_1 = \psi$), and a single rejection–recovery curve (Figure 2) determines β_{H_2S} as functions of α_{H_2} for both stages. One can then choose values of the parameters, F, y^F, α_1, and α_2 and calculate the flow rates and compositions of all streams of Figure 5 as well as α and β, using the equations just listed and Eqs. 4 and 5. The operating pressures (P^{H1} and P^{H2}) of the two stages of the membrane can then be easily calculated from $p^F_{H_2S}$, y^{F*}, and y^{W*} values.

We used the second approach to estimate the performance of the two-stage membrane embodiment of Figure 5 for separation of an equimolar H_2S-H_2 feed-gas mixture ($y^F = 0.5$). Different combinations of α_1 and α_2 values were chosen. A feed gas H_2S partial pressure of 0.395 MPa (curve of Figure 2) was chosen for both stages of the membrane. The results are shown in Table 1.

It may be clearly seen from Table 1 that the desired separation goal can be achieved by operating the two-stage membrane embodiment of Figure 5 using a low recovery of H_2 for stage 1 membrane and a high recovery of H_2 for stage 2 membrane ($\alpha_1 < \alpha_2$). For example, using the values of 0.3 and 0.9 for the variables α_1 and α_2, respectively, this novel protocol for operation of the membrane system can yield an ~98% pure H_2 product stream at 81% H_2 recovery. The product is obtained at the feed-gas pressure (~1.01 MPa). About 98% of feed H_2S is rejected by the membrane system and the H_2S-enriched effluent gas contains 83.8% H_2S. It is produced at near-ambient pressure. Increasing the ratio of (α_1/α_2) significantly lowers the H_2 product purity or its recovery.

One major advantage of directly producting a high-purity H_2 product ($y^P_{H_2} > 97\%$) by the membrane system is that it can be further purified to an essentially pure H_2 ($y_{H_2} = 99.99+\%$) product stream at feed gas pressure by removing the dilute H_2S impurity in a conventional thermal-swing adsorption (TSA) process. The typical H_2 loss in a TSA purification system is only between 5 and 10%. On the other hand, if the H_2 enriched product gas from the membrane system contains more than 3% H_2S, a pressure-swing adsorption (PSA) process will be needed to produce a pure H_2 product. The typical H_2 loss in a PSA purification system is typically between 20 and 30%. Thus, the novel mode of operation of the two-stage membrane described earlier allows higher overall recovery of pure H_2 from a bulk H_2S-H_2 mixture by the use of a hybrid membrane–TSA separation device.

Summary

The SSF membrane can be efficiently used to separate bulk H_2S-H_2 mixtures. H_2S is selectively permeated through the membrane. The H_2S rejection–H_2 recovery characteristics of

Table 1. Performance of Two-stage SSF Membrane for Bulk H_2S-H_2 Separation (Equimolar Feed Gas)

Stage H_2 Recoveries			H_2-Enriched Product Gas		H_2S-Enriched Reject Gas		Operating Pressures	
			H_2 Purity	Recovery	H_2S Purity	Rejection	pH1	pH2
α_1	α_2	(α_1/α_2)	(mol %)	(α %)	(mol %)	(β%)	(MPa)	(MPa)
0.30	0.90	0.33	97.9	81.0	83.8	98.3	1.00	0.83
0.30	0.60	0.50	98.0	51.7	67.2	98.9	1.06	0.86
0.30	0.30	1.00	97.4	38.0	61.5	99.0	0.89	0.74
0.60	0.30	2.00	95.7	68.1	75.2	97.0	0.84	0.58
0.75	0.30	2.50	87.1	81.0	82.2	87.8	0.82	0.52
0.90	0.30	3.00	68.2	92.7	88.6	56.7	0.80	0.47

the membrane can be described by a single-parameter empirical model.

A two-stage embodiment of the SSF membrane can be operated to produce a high-purity H_2 product (mole fraction $\sim 98.0\%$) with high H_2 recovery ($\sim 80\%$) at the feed-gas pressure from an equimolar H_2S-H_2 feed gas. This can be achieved by a novel protocol of operation of the membrane where the H_2 recoveries of the two membrane stages are controlled in a specific manner.

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Notation

F^* = total feed-gas flow rate to stage 1 of membrane system of Figure 5

P^* = flow rate of high-pressure recycle gas in membrane system of Figure 5

W^* = flow rate of feed gas to stage 2 of membrane system of Figure 5

Subscripts and superscripts

1,2 = stages 1 and 2 of membrane of Figure 5

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