

Modelling and simulation of membrane separation system for purge gas†

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A mathematical model has been developed for membrane separation of binary gas mixture for a counter-current flow having membrane configurations of flat-sheet and hollow-fibre. A binary gas system of hydrogen and nitrogen has been used to simulate the conditions for the recovery of hydrogen from the purge gas in ammonia plant. Simulation studies showed that the use of counter-current hollow-fibre feed-outside-mode permeator, at higher feed flow rate, and at a high feed pressure as permitted by the mechanical strength of the system, is best for good recovery of hydrogen. It is further observed that while separation from polyimide membrane is superior to cellulose acetate membrane, performance of polystyrene membrane is quite inferior.

Gas membrane separations are replacing other separation processes such as adsorption, absorption and cryogenics because of its low capital investment, ease of operation, low energy consumption, cost effectiveness even at low gas volumes, and good weight and space efficiency. However, in order to improve upon the extent of enrichment better separation schemes are being investigated that include many types of membrane configurations. There are three types of membrane configuration: flat-sheet hollow-fibre and spiral wound permeators. Hollow-fibre permeators are becoming more common than the others because of its higher surface area to volume ratio. The configuration of the hollow-fibre is of shell and tube type. The packing density of this permeator is as high as several thousand square meters per cubic metre of permeator. Also, successful application of the membrane for gas separation depends not only upon membrane with high selectivity and permeabilities but also on flow pattern of feed and permeate streams. The flow patterns generally used are co-current, counter-current and cross-flow.

Modelling of membrane gas separation based on several assumptions have been reported by a number of investigators. Weller and Steiner¹ developed the model for single stage and multistage flat-sheet permeator. Naylor and Becker² developed the method for calculating number of stages. However, in their studies the flow pattern of the gas was not considered. Later, Walawender

and Stern³ and Blaisdell and Kammermeyer⁴ modified the model developed by Weller and Steiner, and Naylor and Becker by incorporating the flow pattern. They showed that the counter-current flow gives better permeation than co-current. Antonson *et al.*⁵ developed a model for hollow-fibre membrane configurations and showed that feed inside counter-current mode is superior to other. Pan and Habgood⁶ studied the effect of permeate pressure drop for hollow-fibre. Further, Pan⁷ considered the asymmetric membrane rather than symmetric membrane. He studied the mode of feed flow pattern in an asymmetric membrane and showed that counter-current flow pattern is not necessarily always preferable.

Although, the work reported so far have contributed significantly to the knowledge and understanding of membrane gas separation, it appears that there is scarcity of information towards the performance of different types of permeators for purge gas system. Therefore, the objective of the present simulation studies is to compare the performance of the permeator for different flow patterns and study the effect of pressure ratio, feed flow rate and temperature by considering three different membranes. A binary gas system of hydrogen and nitrogen has been used for this study to simulate the conditions for the recovery of hydrogen from the purge gas in ammonia plant.

Mathematical Model

Following assumptions are made in developing the theoretical model to simulate the performance of different permeator for a binary gas system: (i) Permeator is operated at steady state, (ii) Permeabilities of species are constant irrespective of

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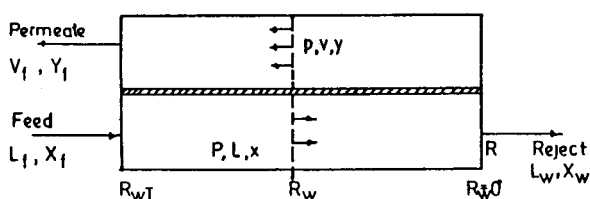


Fig. 1a - Schematic of a flat sheet permeator having counter current flow

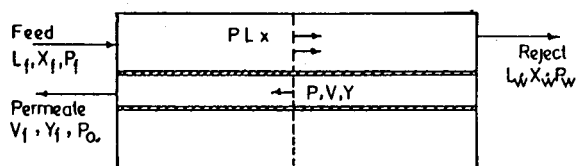


Fig. 1b - Schematic of a hollow fibre feed outside mode gas permeator having counter current flow

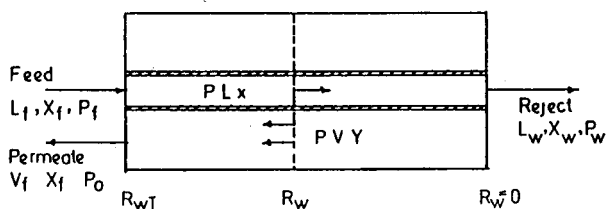


Fig. 1c - Schematic of a hollow fibre feed inside mode gas permeator having counter current flow

pressure and composition, (iii) Membrane is assumed to be homogeneous in structure, (iv) Diffusion along flow pattern is insignificant as compared to bulk flow, (v) There is no radial concentration gradient in the gas phase. (vi) Permeation is isothermal, and (vii) Fibres are not physically deformed on pressurisation.

The mathematical model defining the membrane separation process requires making a mass balance over the membrane configuration. Usually two types of membrane configurations are used in gas separation, one is flat-sheet and another is hollow-fibre. The hollow-fibre permeator operates in two modes, that is, feed-inside-mode and feed-outside-mode. When high pressure gas is on the shell side and the gas permeates into the hollow-fibre, it is feed-outside-mode and reverse of it is feed-inside-mode. In hollow-fibre the general theory of permeator remains the same except that in hollow-fibre there is a pressure variation inside the fibre. The schematic diagram of these three cases are shown in Fig. 1. Three models, therefore, have been developed for the above three cases in the counter-current flow mode. The developed equations are briefly defined here. Their details are presented elsewhere⁸.

The dimensionless equations developed are similar to that of Bouclif *et al.*^{9,10} and are defined as

follows for different gas permeator working in counter-current flow:

Flat-sheet permeator (Fig. 1a)

$$\frac{dx}{dR_w} = (y-x)/(y-x_w) \{ \alpha(1-x)(x-\tau y) - x[(1-x) - \tau(1-y)] \} \quad \dots (1)$$

$$\frac{dy}{dR_w} = (y-x)/(x-x_w) \{ \alpha(1-y)(x-\tau y) - y[(1-x) - \tau(1-y)] \} \quad \dots (2)$$

with initial conditions:

$$x = x_w \text{ and } y = y_w \text{ at } R_w = 0 \quad \dots (3)$$

Hollow-fibre permeator feed-outside-mode (Fig. 1b)

$$\frac{dx}{dR_w} = \{(y-x)/(y-x_w)\} \{ \alpha(1-x)(x-\tau y) - x[(1-x) - \tau(1-y)] \} \quad \dots (4)$$

$$\frac{dy}{dR_w} = \{(y-x)/(y-x_w)\} \{ \alpha(1-y)(x-\tau y) - y[(1-x) - \tau(1-y)] \} \quad \dots (5)$$

$$\frac{d\tau}{dR_w} = -\beta \{ (x-x_w)(x_f - y_f)^2 \} / \{ \tau(y-x)(x_w - y_f)^2 \} \quad \dots (6)$$

with initial conditions:

$$x = x_w \text{ and } y_w \text{ at } R_w = 0 \quad \dots (7)$$

Hollow-fibre permeator feed-inside-mode (Fig. 1c)

$$\frac{dx}{dR_w} = \{(y-x)/(y-x_w)\} \{ \alpha(1-x)(\phi x - \tau y) - x[\phi(1-x) - \tau(1-y)] \} \quad \dots (8)$$

$$\frac{dy}{dR_w} = \{(y-x)/(x-x_w)\} \{ \alpha(1-y)(\phi x - \tau y) - y[\phi(1-x) - \tau(1-y)] \} \quad \dots (9)$$

$$\frac{d\phi}{dR_w} = -\beta \{ (y-x_w)(x_f - y_f)^2 \} / \{ \phi(y-x)(x_w - y_f)^2 \} \quad \dots (10)$$

with initial conditions:

$$x = x_w \text{ and } y = y_w \text{ at } R_w = 0 \quad \dots (11)$$

The model equations developed above for different permeator are the coupled first order ordinary differential equations and are solved by using fourth order Runge-Kutta method. Computational procedure and its details are already discussed⁸.

Results and Discussion

The developed models were tested with data reported by Bouclif *et al.*¹⁰. The detailed simulation study was carried out taking a membrane configuration and flow scheme of hollow-fibre feed-out-side-mode with counter-current flow pattern as it was found to be superior in performance to others, that is, hollow-fibre feed-inside-mode and flat-sheet permeator⁸.

The simulation study for purge gas was done to obtain the effect of operating parameters, such as,

Table 1 – Range of variables studied for purge gas system

S. No.	Parameter	Range
1	Feed flow rate	0.5–4.0 cm ³ /s
2	Feed pressure	25 × 10 ⁵ Pa
3	Pressure ratio	0.1–0.6
4	Temperature	298–773 K
5	Viscosity	2.4 × 10 ⁻⁵ Pa.s
6	Feed composition (mole fraction)	
	(i) Hydrogen	0.65
	(ii) Nitrogen and others	0.35
7	Separation factor	
	(i) Polystyrene	21.91
	(ii) Cellulose acetate	96.15
	(iii) Polyimide	255.68
8	Specific permeability of second component for	
	(i) Polystyrene	3.365 × 10 ⁻¹¹ mol/s.m ² .Pa
	(ii) Cellulose acetate	2.953 × 10 ⁻¹⁰ mol/s.m ² .Pa
	(iii) Polyimide	6.078 × 10 ⁻¹¹ mol/s.m ² .Pa
9	Inside diameter of hollow fibre	1.5 × 10 ⁻⁴ m
10	Outside diameter of hollow fibre	3.0 × 10 ⁻⁴ m

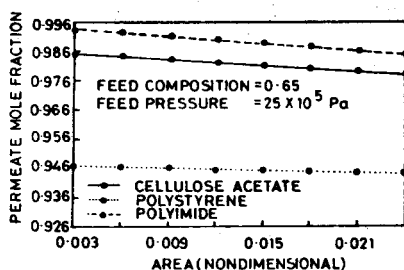


Fig. 2 – Effect of membrane on permeate composition for purge gas system

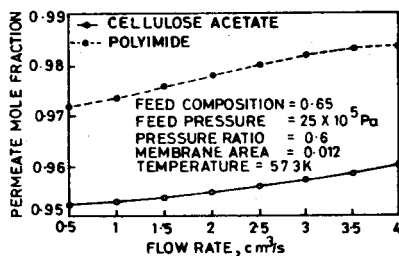


Fig. 3 – Effect of flow rate on permeate composition for purge gas system

flow rate, temperature and pressure ratio on permeate composition. Three different membrane materials, such as, cellulose acetate, polystyrene and polyimide were taken for the study. The simulation study was carried out for hollow-fibre feed-outside-mode. Table 1 defines the range of variables studied for purge gas system. For this study a fixed non-dimensional membrane area (R_w) of 0.024 was used.

The permeate composition obtained for the three different types of membranes are given in Fig. 2. It is evident that the polyimide membrane gives the higher permeate composition compared to cellulose acetate and polystyrene membranes. Polystyrene membrane was found to give the poorest performance. Therefore, for further study, only cellulose acetate and polyimide membranes were considered.

Effect of feed flow rate—The performance of cellulose acetate and polyimide membranes were studied by varying the feed flow rate from 0.5 to 4.0 cm³/s, keeping the other parameters constant. Fig. 3 shows the effect of feed flow rate on the permeate composition. It is evident from figure that there is an increase in permeate composition with increase in flow rate, but the pattern of increase depends upon the membrane. For cellulose acetate there is a marginal increase upto 2.0 cm³/s and after that there is a gradual increase in permeate composition upto 4.0 cm³/s, whereas for polyimide membranes there is gradual increase in permeate composition upto 2.5 cm³/s and after that there is a marginal increase upto

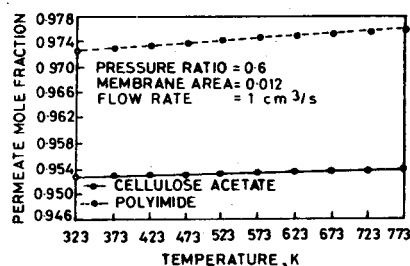


Fig. 4 – Effect of temperature on permeate composition for purge gas system

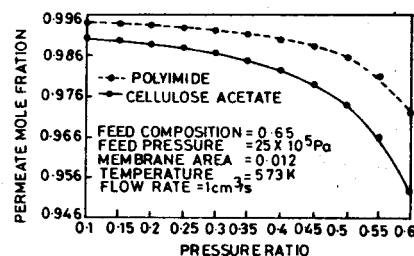


Fig. 5 – Effect of pressure ratio on permeate composition for purge gas system

4.0 cm³/s. Therefore, higher feed flow rate could be recommended for better recovery of the hydrogen subject to the pressure drop considerations.

Effect of temperature—The effect of temperature on permeate composition was studied by varying the temperature from 323 to 773 K. Fig. 4 shows the effect of temperature on permeate composition. It is evident that there is an insignificant effect of temperature on permeate composition. This effect could be due to the fact that the permeability of both the gases increase with rise in temperature. The separation factor is the ratio of permeabilities, therefore, there is a marginal effect on it.

Effect of pressure ratio—The effect of pressure ratio was studied by varying the pressure ratio from 0.1 to 0.6. Fig. 5 shows the effect of pressure ratio on permeate composition. It is evident from this figure that the permeate composition decreases with increase in pressure ratio. However, there is a steep fall in the permeate composition after the pressure ratio of 0.4. It is, therefore, recommended that membranes system should be operated at high feed pressure subject to mechanical strength consideration.

Conclusions

The present simulation study was utilized to study the performance of three membranes, namely, polystyrene, cellulose acetate and polyimide for enriching the hydrogen from the mixture of hydrogen and nitrogen which simulated the purge gas conditions. It was observed that for a fixed non-dimensional membrane area, polystyrene membrane showed the poorest permeate composition compared to cellulose acetate and polyimide membranes.

The performance of the polyimide membrane was found better than cellulose acetate membrane under all operating conditions studied. Further, it was observed that increase in feed flow rate, resulted in higher recovery of hydrogen whereas change in feed temperature was found to have insignificant effect on permeate composition. Moreover the increase in feed pressure was found to be always better for increasing the permeate composition subject to maximum pressure as permitted by the mechanical strength of the system.

Acknowledgement

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Nomenclature

A	= Surface area of membrane, cm ²
D_i	= Inside diameter of hollow-fibre, cm
L	= Local feed flow rate, mol/s
D_{LM}	= Logarithmic mean of inside and outside diameter at hollow-fibre, cm
L_w	= Feed flow rate at reject end, mol/s
L_f	= Feed flow rate at permeator entry, mol/s
P_f	= Inlet feed pressure, Pa
P_1, P_2	= Total pressure on the two sides of membrane, Pa
P_0	= Permeator pressure, Pa
q_i	= Volumetric flow rate of i species, cm ³ /s
R	= Universal gas constant
R_w	= Non-dimensional membrane area ($= AS_2P_f/L_w$)
S_i	= Specific permeability of i species, mol/s cm ² Pa
S_1, S_2	= Specific permeability of more permeable and less permeable component
T	= Absolute temperature, K
V	= Local permeate flow rate, mol/s
V_f, V_w	= Permeate flow rate at feed side and at reject side mol/s
x_{i1}, x_{i2}	= Mole fraction of i species on two side of membrane
x_f, x_w	= Mole fraction of more permeable component on feed side and in reject side
y_f, y_w	= Mole fraction of more permeable component in permeate stream at feed side and at reject side

Greek letters

α	= Separation factor ($= S_1/S_2$)
β	= Constant ($= 128 \mu R T L_f / (\pi^2 D_i^4 D_{LM} S_2 P_f^3 N)$)
τ	= Pressure ratio ($= P_0/P_f$)
μ	= Viscosity of gas, cP
ϕ	= Pressure ratio ($= P_1/P_f$)

Subscript

f	= Pertaining to the permeator axial location where feed enters
w	= Pertaining to the permeator axial location where feed rejects

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