SEPARATION OF GAS MIXTURES USING HOLLOW FIBRE MEMBRANE PILOT PLANT DATA ANALYSIS

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SUMMARY

The permeation rates and selectivities for a specific membrane permeator are dependent on the type of membrane used and the operating conditions, namely feed pressure, stage cut, temperature and feed composition. This paper presents the experimental study that was carried out on a laboratory scale using hollow fiber *Prism* separator with a calculated area of 383517.76 cm^2, to determine the effects of two of these operating conditions; feed pressure and stage cut upon permeation rates and selectivities. Compressed air was used as the gas mixture. The range of the feed pressure and stage cut utilised were respectively 50 psi to 110 psi and 0.0289 to 0.1823.

INTRODUCTION

In 1979, Monsanto Co. came out with a revolutionary membrane for gas separation where instead of using the separating material, polysulfone as coating, it is used as substrate. This approach increased available flux rate to the extent that the *Prism* separators are 10^2 to 10^3 times faster per unit area than any other systems proposed for commercial gas separation [5]. Development such as this would one day replace conventional gas separation techniques such as Cryogenic system or the Pressure Swing Absorption. In this study, a laboratory scale *Prism* separator system was utilised to evaluate separation of oxygen and nitrogen from a compressed air feed.

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THEORETICAL CONSIDERATION

Each gas entering the separator has a characteristic permeation rate that is also a function of its ability to dissolve in and diffuse through the hollow fibre membrane. *Prism* separators utilise these relative permeation rates to selectively separate a 'fast' gas such as hydrogen from the 'slow' gas component in the process gas stream. The driving force for this separation would be the difference between the stream components' partial pressure on the outside of the fibres (Shell side) and the inside of the hollow fibres (bore side). Relative rates of permeation are shown below :

H_2O, H_2, He, H_2S	CO_2, O_2	Ar, CO, N_2, CH_4
'Fast'		'Slow'

Figure 1.0 : Relative Permeation Rates of a Gas Spectrum through hollow fibre membrane [1]

Generally, permeation involves the following sequences of steps: [2]

[1] Absorption of gas at one interface of the membrane
[2] Solution of gas into the membrane at that interface
[3] Activated diffusion of gas in and through the membrane
[4] Release of the gas from solution at the opposite interface
[5] Desorption from the latter interface.

For permeation through planar membranes under steady state condition, where the concentration of the penetrant at the two interfaces of the membrane are kept at different but constant value; the diffusion of gas into the membrane can be expressed by Fick's first law which stated :

$$J = -D_o\ (dc/dx) \qquad \text{.......(1.0)}$$

where J is the rate of diffusion of the penetrant gas, D_o is the diffusion coeffiecient for a specific penetrant-membrane system and temperature; and c is the concentration of the penetrant in the membrane at a position cordinate x.

Integration of equation 1.0 with respect to x across the thickness of the membrane (l) and c, resulted in :

$$J = D_o\ [(C_h - C_l)/l] \qquad \text{.......(2.0)}$$

where C_h and C_l are the penetrant concentration at the membrane high and low interfaces respectively.

For simple non-interacting gases, the concentration in the membrane matrix is given by the Henry's law expression, linking it to partial pressure as below :

$$C = S_o P \qquad ....(3.0)$$

where S_o is the solubility coefficient. By substituting equation 3.0 into 2.0 gives :

$$J = D_oS_o\ [(P_h - P_l)/l] \qquad ....(4.0)$$

where $\bar{P}_o$ (the permeability coefficient) is equivalent to D_oS_o.

Finally the permeation rate (P_o/l) is given by the equation :

$$Q/A = J = \bar{P}_o\ [(P_h - P_l)/l] \qquad ....(5.0)$$

$$\bar{P}_o/l = Q/(A\Delta P) \qquad ....(6.0)$$

where ΔP is the pressure difference between the two interfaces, Q the rate of gas permeation and A the planar surface area of the membrane.

For a binary mixture where gas 1 and gas 2 are present, the permeation rate of gas 1 is given by :

$$\frac{\bar{P}_1}{l} = \frac{Q_1}{(A\ (P_h\ x_1 - P_l\ y_1))} \qquad ....(7.0)$$

where x_1 and y_1 are the mole fraction of gas 1 in the feed and permeate respectively. Similarly for gas 2, its permeation rate is given as :

$$\frac{\bar{P}_2}{l} = \frac{Q_2}{(A\ (P_h\ (1-x_1) - P_l\ (1-y_1)))} \qquad ....(8.0)$$

Ideal selectivity ($\alpha_{1/2}$) defined as the ratio of the permeation rate of fast gas over the permeation rate of the slow gas can be given as :

$$\alpha^*_{1/2} = \bar{P}_1/\bar{P}_2 \qquad ....(9.0)$$

EXPERIMENTAL

The system utilised is a laboratory scale *Prism* separator system, model number MSS - 1x4, containing four 1'' diameter by 5' long hollow fibre bundles with membrane area of 383517.76 cm^2, which is determined by utilising equation 6.0 and taking $\bar{P}/l$ to be 20 x 10^{-6} cm^3/sec cm^2 cmHg [3]. (Figure 2.0) A pressurised feed gas (air in this case) is introduced into the separator and through the bundle of the hollow fibres where the faster gases (oxygen, carbon dioxide) will selectively permeates through the fiber wall. This permeate stream is collected at reduced pressure in a manifold at the bottom of the separators. The non-permeate (retentate) stream exit through the top of the separators at essentially the same pressure as the entering feed gas. The composition of oxygen in the permeate stream (XO2) and retentate stream (YO2) can be read off the Oxycheck oxygen monitor directly.

RESULTS

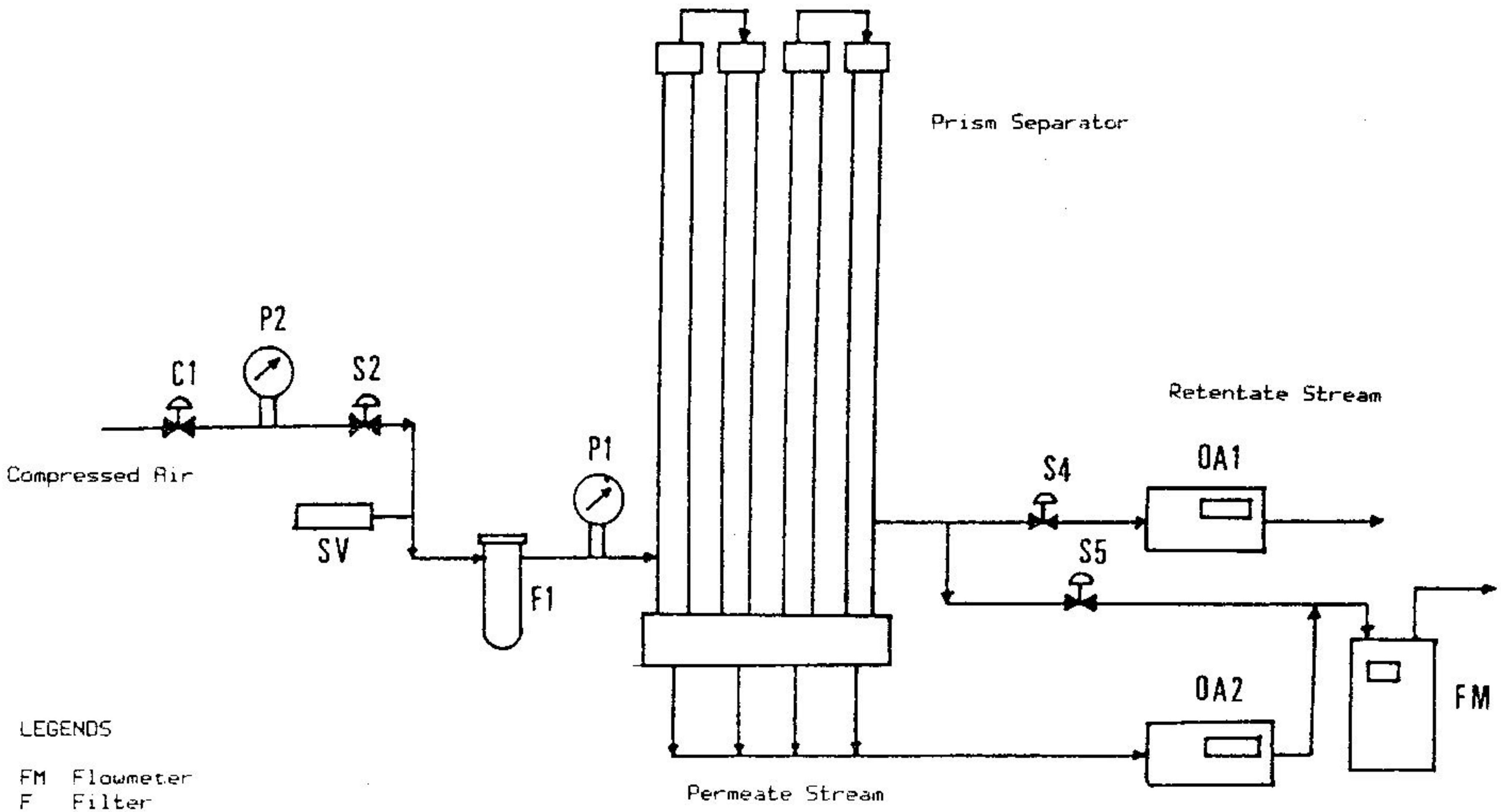
The result of the permeation rates measurement for different feed pressure at constant stage cut and vice-versa are given in table 1.0, 2.0 whereas that of constant stage cut and different feed pressure is given in table 3.0. Stage cut can be calculated through :

$$\theta = Q_P/Q_F \qquad \text{.......(10.0)}$$

Refering to the tables, permeation rates for the oxygen enriched stream (PPO2) and the nitrogen enriched stream (PPN2) are determined from equation 7.0 and 8.0 respectively, while ideal selectivity, as known as 'Alpha' is determined through equation 9.0.

EFFECT OF STAGE CUT ON PERMEATION RATES AND SELECTIVITIES

The general relationship between permeation rate and stage cut had been established; that at lower stage cut, permeation rate of the fast gas is higher whereas the reverse is true for the slow gas [4]. Similar results were obtained in this study. Refering to table 1.0, at a constant feed pressure, that is P_H = 310.2 cmHg and P_L = 75.99 cmHg, flux (F1) is mostly constant at 46 cm /sec but the mole percentage of oxygen gas in the permeate stream increases when stage cut is decreased. This gives an indication that as stage cut decreased, the permeation rate of the fast gas



LEGENDS

FM Flowmeter
F Filter
OA Oxygen Analyser/Monitor
P Pressure Gauge
S Valves
SV Relief Valve

FIGURE 2.0 : LAYOUT OF SEPARATION UNIT

(oxygen) will be faster. A comparison with the calculated oxygen permeation rate (PPO2) proves this to be true where at stage cut 0.1389, PPO2 gives a value of 1.732E-06 $cm^3/cm^2 sec\ cmHg$ while at stage cut 0.0448, PPO2 is found to be 2.220E-06 $cm^3/cm^2 sec\ cmHg$. The nitogen permeation rate, on the other hand, decreases as stage cut is decreased. Thus from the above deduction, it can be concluded that the dependency of oxygen and nitrogen permeation rate correspond with the general relationship above.

A functional relationship was also established between permeation rate and stage cut which takes the form : [4]

$$\bar{P}O_2 = Exp\ (R\bar{P}O_2 + S\bar{P}O_2\ [\theta]) \qquad ....(11.0)$$

$$\bar{P}N_2 = Exp\ (RP\bar{N}_2 + SP\bar{N}_2\ [\theta]) \qquad ....(12.0)$$

where $R\bar{P}O_2$ and $R\bar{P}N_2$ are the intercepts while $S\bar{P}O_2$ and $S\bar{P}N_2$ are the gradients in a semilogarithm plot of permeation rate (P) versus stage cut. Most of the correlation coefficient (R^2) was found to be greater than 0.7 indicating significant relationship between the two variables. As seen from figure 3.0, the permeation rate of oxygen and nitrogen increased and decreased respectively at lower stage cut and the curve fitting using equation 11.0 and 12.0 gave good results.

Ideal selectivities ($\alpha^*_{1/2}$) also show an increase at lower stage cut confirming the results obtained from similar studies [3]. All the calculated selectivities values are greater than 4.5 showing good separation between oxygen and nitrogen. A functional relationship was also established between stage cut and selectivities, taking the form : [5]

$$\alpha_{O_2/N_2} = R\alpha\ Exp\ (S\alpha\ \theta) \qquad ....(13.0)$$

where the values of $R\alpha$ and $S\alpha$ can be obtained by preforming linear regression on the equation below :

$$Ln\ \alpha = C\alpha + S\alpha\ \theta \qquad ....(14.0)$$

where $R\alpha = Exp\ (C\alpha)$, $S\alpha$ is the regression coefficient and $C\alpha$ is the intercept.

FIGURE 3.0 : EFFECT OF STAGE CUT ON

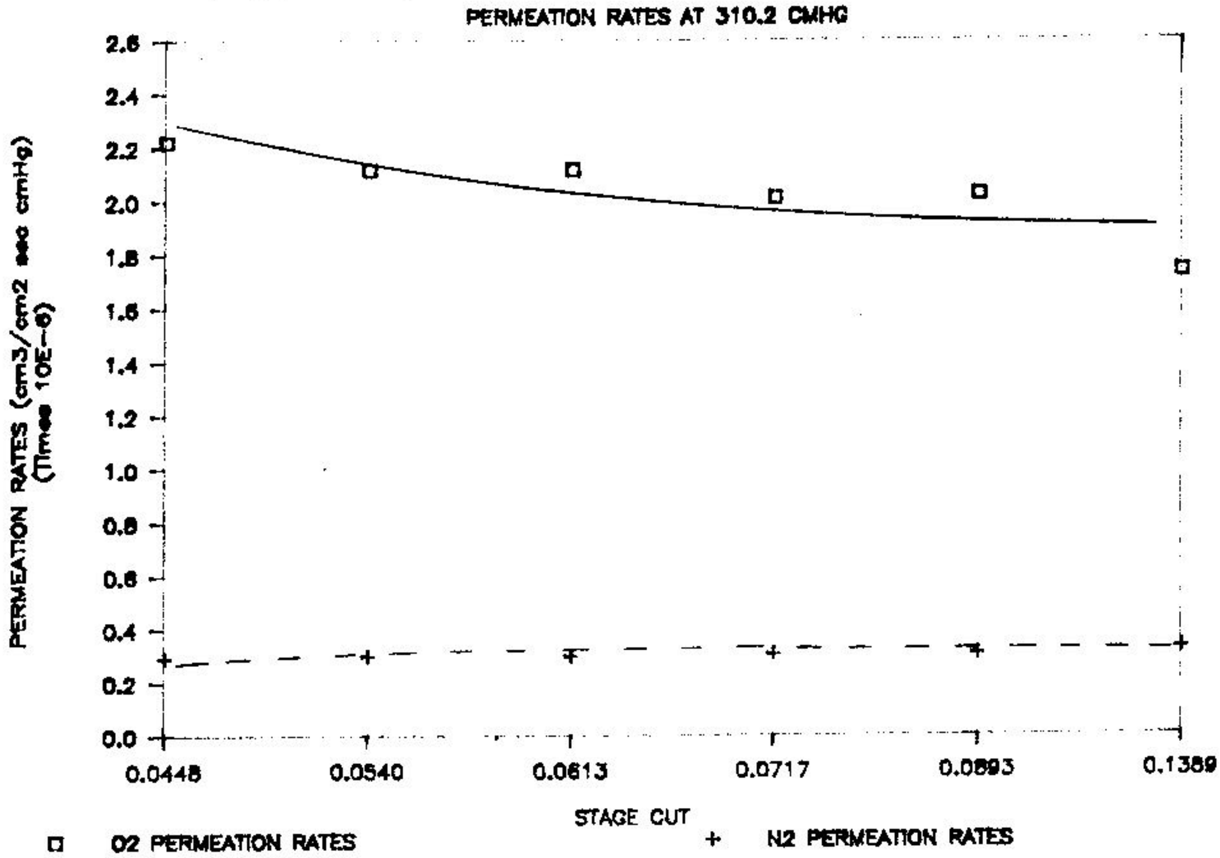


FIGURE 4.0 : EFFECT OF STAGE CUT ON

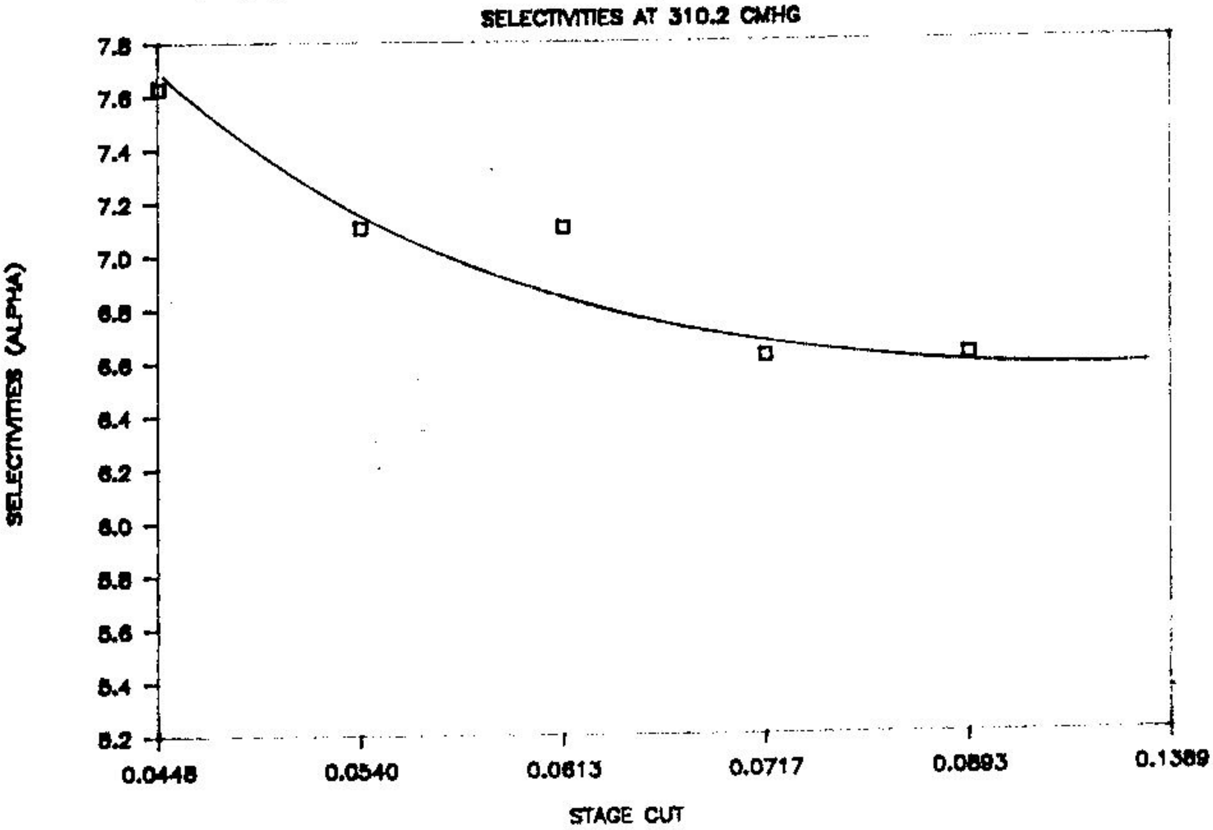


FIGURE 5.0 : EFFECT OF FEED PRESSURE ON

PERMEATION RATES AT STAGE CUT 0.1329

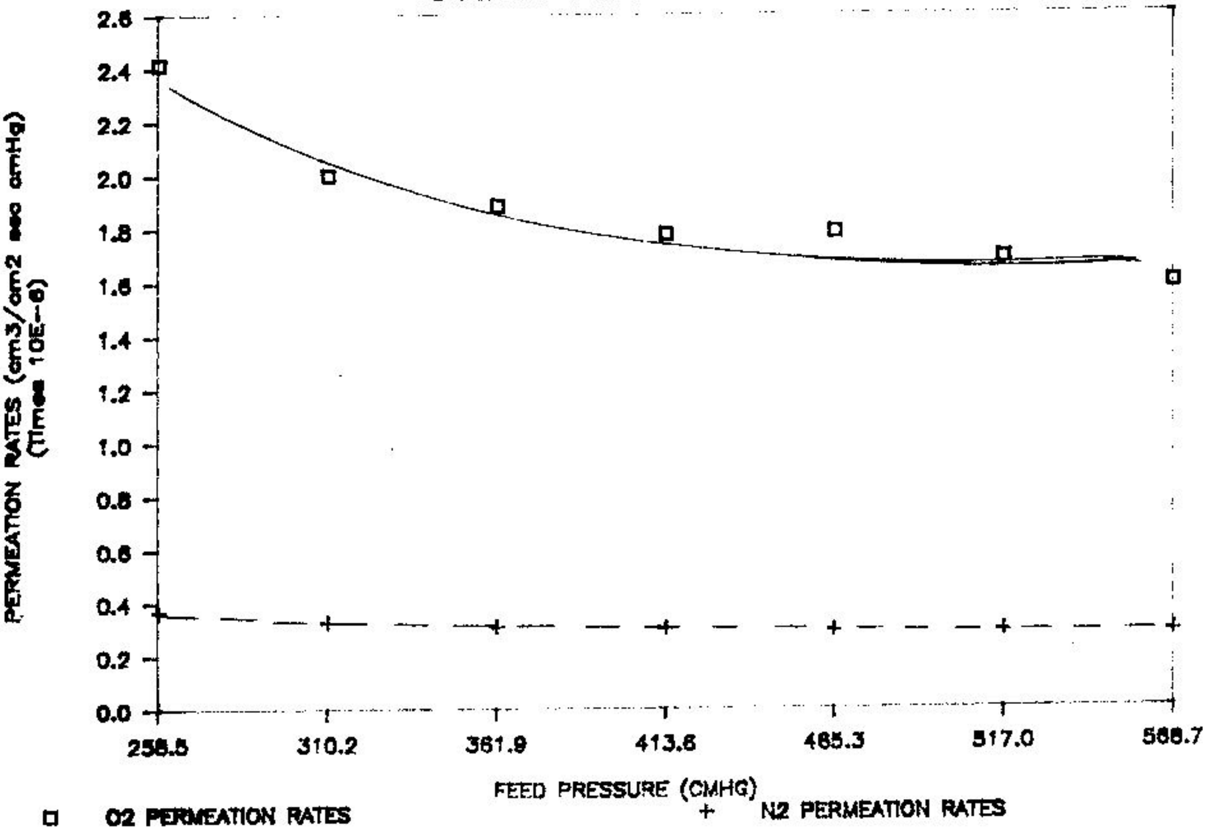
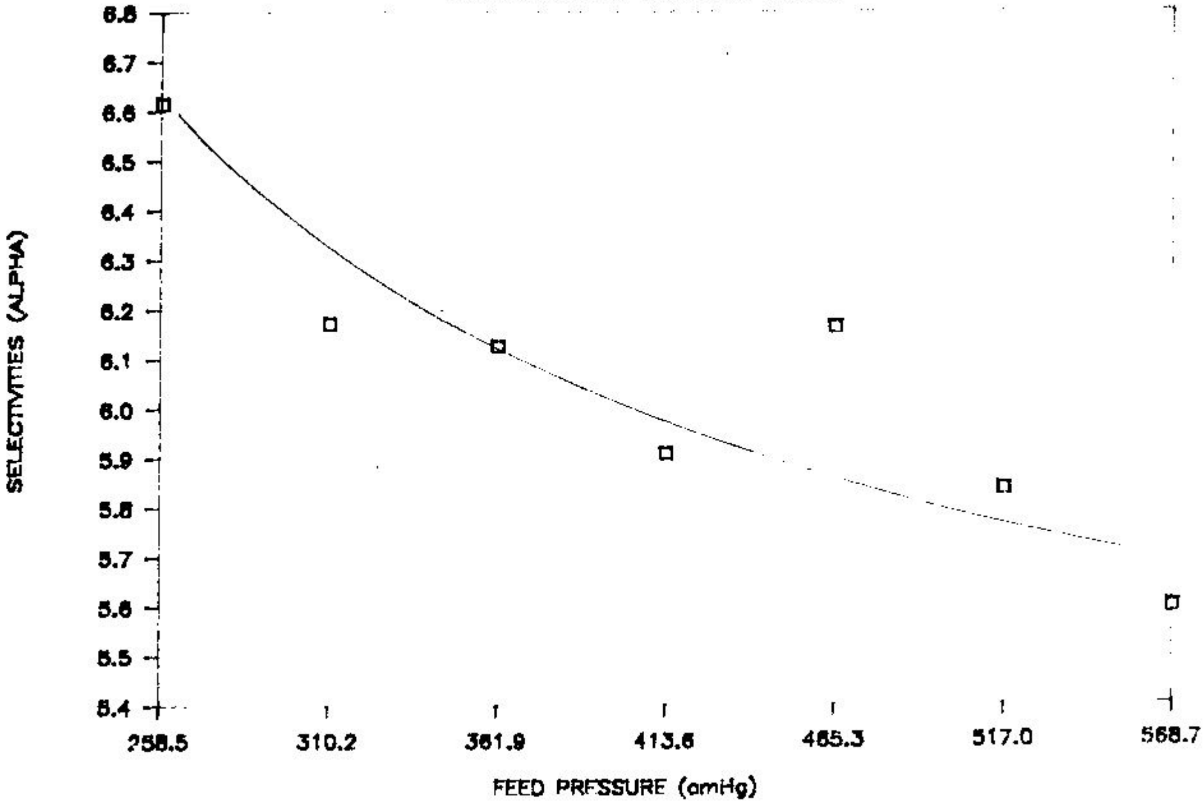


FIGURE 6.0 : EFFECT OF FEED PRESSURE ON

SELECTIVITIES AT STAGE CUT 0.1329



EFFECT OF FEED PRESSURE ON PERMEATION RATES AND SELECTIVITIES

At higher feed pressure, the permeation rate of both oxygen and nitrogen was found to decrease where oxygen decrement seems to be more distinct giving rise to lower selectivities (figure 5.0 and table 3.0). This result does not correspond with the normal established relationship which states that higher feed pressure should give rise to higher selectivities [4]. This is possibly due to temperature fluctuation within the membrane system and the limited range of feed pressure utilised. Operating at higher pressure is still preferred as purity of oxygen in the permeate stream is higher at higher feed pressure although there is a significant drop in oxygen permeation rate.

CONCLUSION

A more oxygen enriched stream can be obtained at lower stage cut. This is bacause as stage cut is decreased, the permeation rate for oxygen increased significantly while there is a drop in the permeation rate of nitrogen. The study also shows that operating at higher pressure is preferred where higher purity of oxygen in the permeate stream was obtained. It would be appropriate to carry out further study on the effect of temperature on permeation rate and selectivities. This would enable us to obtain a more complete picture of the whole permeation process.

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Table 1.0 : Results of Permeation Rates and Selectivities Measurements for Compressed Air at Constant Feed Pressure but Different Stage Cut.

PH (cmHg)	THETA	F1 (cm3/sec)	F2 (cm3/sec)	XO2	YO2	PPO2 (cm3/cm2 sec cmHg)	PPN2 (cm3/cm2 sec cmHg)	ALPHA
				Pressure Drop Across Membrane = 234.70 cmHg				
310.2	0.0448	46.2367	985.043	0.50	0.21	2.220E-06	2.911E-07	7.6275
310.2	0.0540	46.1990	809.143	0.49	0.20	2.115E-06	2.978E-07	7.1026
310.2	0.0613	46.2367	708.000	0.49	0.20	2.117E-06	2.980E-07	7.1026
310.2	0.0717	46.0488	596.211	0.48	0.20	2.010E-06	3.038E-07	6.6185
310.2	0.0893	46.2745	472.000	0.43	0.19	2.020E-06	3.053E-07	6.6185
310.2	0.1389	45.6774	283.000	0.45	0.18	1.732E-06	3.223E-07	5.3740

Table 2.0 : Results of Permeation Rates and Selectivities Measurements for Compressed Air at Constant Feed Pressure but Different Stage Cut.

PH (cmHg)	THETA	F1 (cm3/sec)	F2 (cm3/sec)	XO2	YO2	PPO2 (cm3/cm2 sec cmHg)	PPN2 (cm3/cm2 sec cmHg)	ALPHA
				Pressure Drop Across Membrane = 389.30 cmHg				
465.3	0.0574	72.6154	1192.421	0.53	0.20	1.747E-06	2.681E-07	6.5155
465.3	0.0602	72.6154	1132.900	0.52	0.20	1.692E-06	2.745E-07	6.1635
465.3	0.0714	72.6154	944.000	0.52	0.20	1.692E-06	2.745E-07	6.1637
465.3	0.1034	72.6154	629.333	0.51	0.19	1.638E-06	2.809E-07	5.8317
465.3	0.1444	70.8000	419.556	0.49	0.17	1.496E-06	2.863E-07	5.7249

Table 3.0 : Results of Permeation Rates and Selectivities Measurements for Compressed Air at Constant Stage Cut but Different Feed Pressure.

PH (cmHg)	THETA	F1 (cm3/sec)	F2 (cm3/sec)	XO2	YO2	PPO2 (cm3/cm2 sec cmHg)	PPN2 (cm3/cm2 sec cmHg)	ALPHA
				Stage Cut = 0.1329				
258.5	0.1329	41.3931	269.714	0.45	0.19	2.415E-06	3.650E-07	6.6149
310.2	0.1729	48.0000	314.667	0.47	0.18	1.999E-06	3.239E-07	6.1713
361.9	0.1329	57.2121	372.622	0.49	0.18	1.886E-06	3.078E-07	6.1251
413.6	0.1329	66.6353	435.692	0.50	0.18	1.778E-06	3.009E-07	5.9093
465.3	0.1329	76.5402	501.239	0.52	0.19	1.783E-06	2.893E-07	6.1577
517.0	0.1329	85.8182	560.793	0.52	.18	1.635E-06	2.888E-07	5.9357
568.7	0.1329	93.6190	615.652	0.57	0.17	1.388E-06	2.838E-07	5.3961