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# The Study of Joule Thompson Effect for the Removal of $CO_2$ from Natural Gas by Membrane Process

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**Abstract**
In recent years, membrane gas separation has emerged as an alternative of other available technologies (such as absorption and cryogenic separation) in order to remove $CO_2$ from natural gas. A common phenomenon during gas separation by membrane is the occurrence of Joule-Thomson (JT) effect. The temperature may change to a large extent dependent on the type of gas and the pressure applied. In turn, this temperature change may have a large influence on the permeation properties. Moreover, feed gas may condense due to sudden expansion caused by the decrease in pressure while permeating through the membrane. The effect of different parameters on the temperature change due to JT effect has been investigated under the present study. It is shown that the temperature drop is quite significant for feed gas with high $CO_2$ contents and high pressure, and thus potentially causes condensation across the membrane. It may be prevented by achieving a predetermined dew point before the membrane and then heating the gas to provide a sufficient margin of super heat. This increase in temperature increases the margin between the gas dew point and operating temperature and thus prevents condensation in the membrane.

**Keywords:** Joule Thompson Effect, Gas Separation, Membrane Technology

## 1. Introduction

Carbon dioxide ($CO_2$) separation is very important in many industrial fields such as environmental, petrochemical, agricultural and other related industries, among which the natural gas processing is probably the most important [1, 2, 3]. In some deposits, natural gas may contain high contents of $CO_2$ that must be removed to an acceptable level by the gas producer in order to serve the following purposes; increase the heating value of the gas, prevent corrosion of pipeline and process equipments and crystallization during liquefaction process [4, 5].

There can be different processes for the removal of $CO_2$ considering the factors of; capital and operating costs, gas specifications and environmental concerns. The major processes can be grouped as absorption Processes (chemical and physical absorption), adsorption processes (solid surface), hybrid solution (mixed physical and chemical solvent) and Physical Separations (Membrane. Cryogenic Separation) [6, 7, 8].

Membranes processes are widely used for natural gas processing during the last two decades [9]. For a gas to permeate through a membrane surface, the gas must first dissolve in the high-pressure side of the membrane, diffuse across the membrane wall, and evaporate from the low-pressure side. Gas separation therefore works on the principle that some gases are more soluble in, and pass more readily through polymeric membrane than the other gases [10, 11]

When a gas expands through a restriction from higher pressure to lower pressure, the change in temperature under constant enthalpy is called Joule Thompson (JT) expansion [12]. The change in temperature is dependent on the pressure change and proportional to the JT coefficient [13]. Joule-Thomson effect is known as a special phenomenon in gas separation by membrane. This occurs if a gas is expanded across a membrane, as in the case of a gas permeation process. In the case of such an adiabatic expansion of a real gas, the temperature may change to a large extent dependent on the type of gas and the pressure applied. In turn, this temperature change may have a large influence on the permeation properties, i.e., if the temperature decreases generally the flux decreases and the selectivity increases [14]. Moreover, feed gas may condense due to sudden expansion caused by the decrease in pressure while permeating through the membrane.

Recently, we proposed a cross flow membrane model to be incorporated with Aspen HYSYS in order to design the membrane system for $CO_2/CH_4$ separation [15, 16]. In this work, Joule Thompson model will be included in Aspen HYSYS along with the pre-existing cross flow membrane model in order to study the effect of different feed compositions and pressures of natural gas on the temperature drop across the membrane. It can help to predict the potential occurrence of condensation across the membrane

## 2. Methodology

The study is based on the cross flow model as shown in the detailed flow diagram (Fig. 1). The assumptions that follow the suggested model are:

[1] It holds for the binary gas mixture
[2] No mixing in the permeate side as well as on the high pressure side.
[3] Permeability is independent of pressure and temperature of the gas stream.
[4] It represents the whole membrane module and will not involve the details inside the module.
[5] The concentration polarization is assumed to be negligible.

Based on these assumptions, the composition of permeate can be determined at any point along the membrane by the relative permeation rates of feed component at that point. The local permeation rate at any point in the stage over a differential membrane area $dA_m$ is

$$ydV = \frac{P_A}{t[p_h x - p_l y]} \qquad (1)$$

$$(1-y)dV = P_B/t[p_h\,(1-x) - p_l\,(1-y) \qquad (2)$$

Dividing eq (i) by eq (ii), we get

$$\frac{y}{1-y} = \frac{\alpha[x - (p_l - p_h)y]}{(1-x) - \left(\frac{p_l}{p_h}\right)(1-y)} \qquad (3)$$

Using ingenious transformations, an analytical solution to the three equations (eq. (1) - eq. (3)) have been obtained.

$$\frac{(1-\theta^*)(1-x)}{(1-Xf)} = \left[ [\frac{U_f - \frac{E}{D}}{U - \frac{E}{D}}]^R [\frac{U_f - \alpha + F}{U - \alpha + F}]^S [\frac{U_f - F}{U - F}]^T \right]$$
(4)

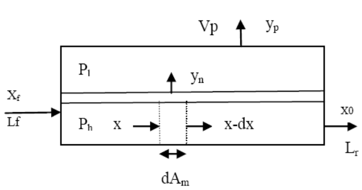


**Fig. 1: Schematic diagram of cross flow membrane separation**

Where

$\theta^* = 1 - \frac{L}{L_f}$ (L as flow rate permeated in the differential element)

$$i = \frac{x}{(1-x)}$$

$$u = -Di + \left(D^2 i^2 + 2Ei + F^2\right)^0.5$$

$$D = 0.5\frac{(1-\alpha)p_l}{p_h} + \alpha$$

E = ($\alpha$ /2)-DF

$$F = -0.5\frac{(1-\alpha)}{p_h}p_l - 1$$ , R = 1/ (2D − 1)

$$S = (\alpha(D-1) + F))/((2D-1) - (\alpha/2 - F))$$

$$T = \frac{1}{1 - D - \left(\frac{E}{F}\right)}$$

The term uf is the value of u at i = if. The value of $\theta^*$ is the fraction permeated up to the value of x. At the outlet where x = $x_0$, the value of $\theta^*$ becomes equal to $\theta$ i.e., the total fraction permeated. The composition of the permeate stream is yp and thus can be calculated from the overall material balance.

$$yp = \frac{xf - x0(i-\theta)}{\theta} \qquad (5)$$

The total membrane area is then calculated using additional transformations of eqs. (1) to (5) in order to obtain

$$A_m = \frac{tL_f}{p_h P_B}\int_{i_o}^{i_f} \frac{(1-\theta^*)(1-x)di}{[(f]_i - i)\left[\frac{1}{1+i} - \frac{P_l}{P_h}\left(\frac{1}{1+f_i}\right)\right]} \qquad (6)$$

Where

$f_i = (Di - F) + (D^2 i^2 + 2Ei + F^2)^{0.5}$

The term if is the value of i at the feed and $i_0$ is the value of i at the outlet. The integral is solved numerically to calculate the value of total membrane area required for the separation [17, 18].

The JT coefficient must be calculated in order to study the JT expansion effect on the gas passing through the membrane [14].

$$\mu_{JT} = \frac{RT^2}{\rho C_{m,p}}\left(\frac{\partial Z}{\partial T}\right)_P \qquad (7)$$

$$C_{m,p} = \frac{C_P}{M} = \frac{C_{PI} - RT(T\phi'' + 2\phi')}{M} \qquad (8)$$

Where: $C_{PI}$: Ideal heat molar capacity

$\phi', \phi''$: First and second derivatives of the gas fugacity coefficient

The first derivative of the compression factor with respect to temperature is:

$$Z' = \frac{R(TZ)^2 \sum_{n=13}^{58} C_n^{*'} D_n^* + pZ\left(TZ_0 \square'_{Z_1}\right)}{R\,(TZ)^2 + pTZ_1} \qquad (9)$$

Where

$$Z = 1 + B\rho_m\ \rho_r \sum_{n=13}^{18} C_n^* + \sum_{n=13}^{58} C_n^*\ D_n^*$$

$$Z_0 = B - K^3 \sum_{n=13}^{18} C_n^*$$

$$Z_0' = B' - K^3 \sum_{n=13}^{18} C_n^{*'}$$

$$Z_1 = Z_0 + \sum_{n=13}^{58} C_n^*\ D_{1n}$$

- $\rho_m$: Gas mixture molar density
- $\rho_r$: Reduced density
- B: Second virial coefficient
- $C_n^*$: Temperature – Composition dependent coefficient

Therefore, the final analytical equation for Joule-Thomson coefficient is given by

$$\mu_{JT} = \frac{RT^2}{\rho} \times \frac{M}{CPI - RT\,(T\emptyset'' + 2\emptyset')} \times \frac{R(TZ)^2 \sum_{n=13}^{58} C_n^*\ D_n^* + \rho Z\left(TZ_0' - Z_1\right)}{R(TZ)^2 + \rho T Z_1}$$

The temperature drop due to JT effect can thus be calculated as follows

$$T_1 - T_2 = \mu_{JT}\,(P_1 - P_2) \qquad (10)$$

## 3. Results and Discussions

The magnitude of the JT coefficient depends upon the feed composition and changes with temperature and pressure. As a result, temperature drop due to JT effect is proportional to the product of JT coefficient and pressure drop. For feed gas at 80 bar, temperature drop due to JT effect has been studied at different compositions of $CO_2$ as shown in the Fig. 2. Similarly, temperature drop due to JT effect has been calculated for feed gas having 70 % composition of $CO_2$ at different feed pressures as shown in the Fig. 3.

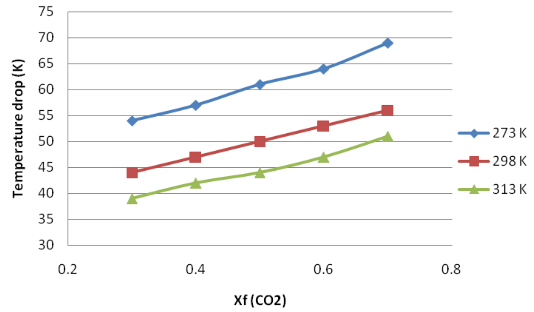


**Fig. 2. Effect of feed composition on temperature drop due to JT effect at various feed temperatures (feed pressure=80 bar)**

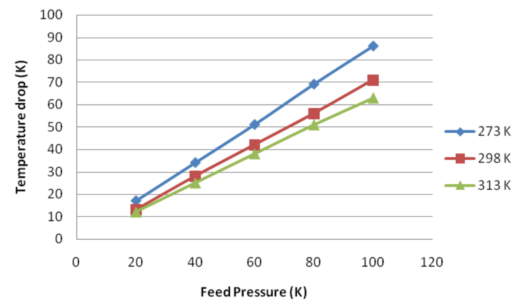


**Fig. 3. Effect of feed pressure on temperature drop due to JT effect at various feed temperatures (feed composition = 70% of $CO_2$)**

It can be observed that temperature drop increases linearly with the increase in $CO_2$ contents as well as feed pressure of the gas. The temperature drop is significant for feed gas having high $CO_2$ contents in comparison to low $CO_2$ contents. It can also be noted that if the feed temperature is high, there is less temperature drop and vice versa.

The temperature drop across the membrane potentially causes condensation across the membrane especially for processes dealing with high $CO_2$ contents and high pressure. It has to be prevented in order to avoid the membrane damage. The damage can be avoided by achieving a predetermined dew point before the membrane and then heating the gas to provide a sufficient margin of super heat. This temperature increase helps to raise the margin between the gas dew point and operating temperature and thus prevents condensation in the membrane. For that purpose, it is recommended to install heater in the process before the membrane to ensure less temperature drop and thus avoid condensation.

## 4. Conclusions

The proposed model for Joule Thompson effect is included in the process simulation (Aspen HYSYS) along with pre-existing cross flow membrane model. The design sensitivity has been investigated by changing the operating conditions such as feed composition and pressure at different temperatures. For feed gas with high $CO_2$ contents and high pressure, the temperature drop is quite significant and thus potentially causes condensation across the membrane. In order to avoid condensation, heater could be installed before the membrane in the process. The Aspen HYSYS user defined unit operation along with Joule Thompson model, proposed under present study has potential to be applied for design, optimization and scale up of complex membrane systems.


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## REFERENCES


[1] I. Songa, H. Ahna, H. Jeona, H. K. Jeonga, Y. Leea, S. H. Choib, J. H. Kimb, S. B. Leeb, "Optimal design of multiple stage membrane process for carbon dioxide separation" Desalination Vol. 234, pp 307–315, 2008.

[2] E. F. Vansant and R. Dewolfs, "Gas Separation Technology", Elsevier Science, Amsterdam, p. 450, 1990.

[3] J. Shen, L. Wu, L. Zhang, Y. Dong, H. Chen and C. Gao, "Selective permeation of CO2 through a composite membrane with a separation layer of 2-N,N-dimethyl aminoethyl methacrylate and acrylic acid copolymer", Desalination, Vol. 193, pp. 327–334, 2006.

[4] J. Hao, P. A. Rice and S. A. Stern, "Membrane processes for the removal of acid gases from natural gas. II. Effects of operating conditions, economic parameters, and membrane properties", *Journal of Membrane Science*, Volume 81, Issue 3, pp. 239-252, June 1993.

[5] M. H. Safari, A. Ghanizadeh, and M.M Montazer-Rahmati, "Optimization of membrane-based CO2-removal from natural gas using simple models considering both pressure and temperature effects" *International Journal of Greenhouse Gas Control*, Vol. 3, pp. 3-10, 2008.

[6] S.A. Ebenezer, "Removal of Carbon dioxide from natural gas for LPG production", *Semester project work.* Institute of Petroleum Technology, Norwegian University of Science & Technology, Trondheim, Norway. 2005.

[7] R.N. Maddox, *Gas and Liquid Sweetening*, 2nd ed., Campbell Petroleum series, 1974.

[8] W.J. Koros and R.T. Chern, "Separation of gaseous mixtures using polymer membranes, in: R.W. Rousseau (Ed.)", *Handbook of Separation Process Technology*, Wiley, New York, 1987, pp. 863–953

[9] X. He, J. A. Lie, E. Sheridan, M. B. Hägg, "CO2 Capture by Hollow Fibre Carbon Membranes: Experiments and Process Simulations" Energy Procedia vol. 1, pp.261-268, 2009.

[10] R. W. Baker, "Membrane Technology and Application", 2nd ed., John Wiley & Sons, Chichester, UK, 2004.

[11] S. A. Ebenezer., "Removal of Carbon dioxide from natural gas for LPG production". Semester project work. Institute of Petroleum Technology, Norwegian University of Science & Technology, Trondheim, Norway, 2006.

[12] I. Maric, "The Joule Thompson effect in natural gas flow measurements", Flow Measurement and Instrumentation, Vol. 16, pp. 387-395, 2005.

[13] M. Mulder, Basic principles of membrane technology. 2nd edition, John Wiley, New York. Young, *The Technical Writer's Handbook"* Mill Valley, CA: University Science, 1989.

[14] F. Ahmad, K.K. Lau and A.M. Shariff "Removal of CO2 from Natural Gas Using Membrane Separation System: Modeling and Process Design", *J. Applied Sci*., Vol. 10, Issue 12, pp 1134-1139, June 2010.

[15] F. Ahmad, K.K. Lau and A.M. Shariff "Modeling and Parametric Study for CO2/CH4 Separation using Membrane Processes", *World Academy Of Science, Engineering And Technolog*, Issue 72, pp 994-999, Dec. 2010.

[16] C. J. Geankoplis, "Transport processes and separation process principles" fourth edition, Prentice Hall, New Jersey, 2003

[17] Weller, S. and Steiner, W.A., Separation of gases by fractional permeation through membranes. *Journal of Applied Physics*, Vol. 21, pp. 180-184. 1950.