The Study of Joule Thompson Effect for the Removal of CO$_2$ from Natural Gas by Membrane Process

*Faizan Ahmad, Lau Kok Keong, Azmi Mohd. Shariff, Ghulam Murshid
Universiti Teknologi PETRONAS, Bandar Seri Iskandar, 31750, Tronoh, Perak, Malaysia
*Corresponding author E-mail: faizan615@gmail.com

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

$$y_dV = \frac{P_A}{t[H - x - P_i y]}$$

$$y dV = P_i B / t[P_i h(1-x) - P_i l(1-y)]$$

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

$$\frac{y}{1-y} = \frac{a[U - (P_i - P_h)y]}{(1-x) - \frac{P_i}{P_h}(1-y)}$$

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

$$\frac{(1 - \theta^*)(1-x)}{(1-x)} = \left[\frac{U_i - \theta^* F}{U - \theta^* F}\right] \left[\frac{U - \alpha + F}{U - \alpha + F}\right] \left[\frac{U_i - \alpha + F}{U_i - \theta^* F}\right]$$

$$\frac{(1 - \theta^*)(1-x)}{f_i} = \int_{x_f}^{x_0} \frac{(1 - \theta^*)(1-x)}{(1-x)} dx$$

The term $u_f$ is the value of $u$ at $x = x_0$. 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 $y_p$ and thus can be calculated from the overall material balance.

$$y_p = \frac{xf - xo(i - \theta)}{\theta}$$

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

$$A_m = \frac{tL_f}{P_h P_i} \int_{x_f}^{x_0} \frac{(1 - \theta^*)(1-x)}{(1-x)} dx$$

Where

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

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].

$$h_{JT} = \frac{RT^2}{\rho C_{c,p} \frac{\partial Z}{\partial T}}$$

$$C_{c,p} = \frac{C_P}{M} = \frac{C_H - RT(T \phi + 2 \phi)}{M}$$

Where: $C_P$: 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:

\[
Z' = \frac{R(TZ)^2 \sum_{n=1}^{58} C_n^* D_n^* + pZ(TZ_0 - 1)}{R(TZ)^2 + pTZ}
\]  
(9)

Where

\[
Z = 1 + B \rho_m
\]
\[
Z_0 = B - R^2 \sum_{n=1}^{58} C_n^*
\]
\[
Z_0' = B' - R^2 \sum_{n=1}^{58} C_n^*
\]
\[
Z_1 = Z_0 + \sum_{n=1}^{58} C_n^* D_{1n}
\]

- \( \rho_m \): 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

\[
\mu_{JT} = \frac{1}{\rho} \left( \frac{M}{m} \right) \left( \frac{RT^2 \sum_{n=1}^{58} C_n^* D_n^* + pZ(TZ_0 - 1)}{R(TZ)^2 + pTZ} \right)
\]

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

\[
T_1 - T_2 = \mu_{JT} (P_1 - P_2)
\]  
(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 CO₂ as shown in the Fig. 2. Similarly, temperature drop due to JT effect has been calculated for feed gas having 70% composition of CO₂ at different feed pressures as shown in the Fig. 3.

It can be observed that temperature drop increases linearly with the increase in CO₂ contents as well as feed pressure of the gas. The temperature drop is significant for feed gas having high CO₂ contents in comparison to low CO₂ 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 CO₂ 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 CO₂ 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.

ACKNOWLEDGMENT

This work was done with the financial support from Universiti Teknologi PETRONAS.

REFERENCES


