Saving Energy and Tray Numbers in Ethylene-Ethane Distillation Towers by Vapor Permeation Membrane Load Sharing

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Abstract

Currently the separation of light hydrocarbon binaries, like ethylene-ethane mixture, is achieved almost exclusively by cryogenic distillation. However the process is highly energy intensive because of close relative volatilities (near unity), necessitating increased reflux ratio and number of stages, with the attendant increased energy consumption in the form of condenser and reboiler duties. It was also shown that the use of membrane separators alone will require excessive feed pressure, hence very high compressor power to drive the system, hence are constrained to small scale separations due to the large areas needed. Thus for difficult separations involving mixtures with very close relative volatilities like ethane-ethylene mixture, it was shown that the use of a distillation vapor membrane hybrid to share the load leads to significant reduction in the overall energy expenditure for the process while still achieving the desired petrochemical grades of 99% in each product stream.

Keywords: Distillation, Membrane, Hybrid, Load-sharing, Energy-saving

Introduction

Distillation- the usual method of separating inorganic/organic gaseous/liquid mixtures- is energy intensive because of the condenser and reboiler duties occasioned by the reflux ratio. This is even more pronounced in cryogenic distillation systems where the mixtures are light hydrocarbons with close relative volatilities (near unity), necessitating increased reflux ratio and number of stages, with the attendant increased energy consumption.

Although, there are a number of new developments for alternative separation techniques, they tend to have important limitations that in many cases make them unattractive for practical purposes; e.g. some membrane technologies are constrained to small scale separations (Fuertes, 2002; Teramoto et al, 2002) due to the large areas needed, or they need high compressor power to drive the system (Ohlrogge 2001). Thus hybrid systems (i.e. distillation-membrane) offer an interesting alternative in some difficult separations for distillation with either large number of theoretical trays, large heat loads or both ((like ethane–ethylene mixture) . However because of the stated inherent limitations in each method, there is a need to find the best configuration/ optimum process conditions for the hybrid system that will minimize the costs of energy or maximize energy savings.

Design methods for hybrid distillation membrane process have been proposed by a number of authors (Fahmy, 2001; Fontalvo, 2005.). Their models rely on simple shortcut models for both column and membrane, but important design parameters like reflux ratio of the distillation column were not considered in their works. Pettersen et al (1995) and Callabero et al (2009) presented design models for vapor permeation systems. Their method, based on McCabe diagrams for distillation, is also only valid for binary systems. Also they presented a procedure for screening calculations that allow calculating the break even cost of a membrane above which the hybrid process would be too high to be competitive with distillation systems. All the above mentioned short-cut procedures are restricted to binary systems with ideal vapor liquid equilibrium behavior

In this work we will focus only on a single distillation column and a single membrane module because the separation factor is usually not a problem, but the cost of the membrane and power requirements can be critical (El-Bourawi, 2006). Here energy sharing configuration where the membrane draws from the column overhead product was investigated in terms of its energy efficiency compared to the individual units. By incorporating a membrane in series to draw the overhead Vapor, the number of theoretical stages will obviously be reduced since membrane will supplement the separation (Soave, 2002; Asma, 2014). This translates to reduced energy spending in the distillation column (Soave 2002.) but to increased throughput to the membrane which implies increased feed pressure, hence increased energy spending in the membrane. Thus in the serial hybrid system, the more the number of stages is reduced, the more the energy reduction in the column but the more will be the throughput and hence the increased energy requirement of the membrane.

From the foregone discussion, if follows that decrease in the column energy (by reducing the number of stages and reflux ratio increases the membrane energy requirement (by increasing the compressor power). The reverse is also true. Thus the task is to determine the optimal serial hybrid combination that will give an overall minimum energy while achieving the same required degree of purity/separation. This will involve evaluating and comparing the energy spending in individual units as well as the hybrid configuration

Materials and Methods

Figures.1, 2 and 3 showed the distillation only, membrane only and hybrid arrangements respectively

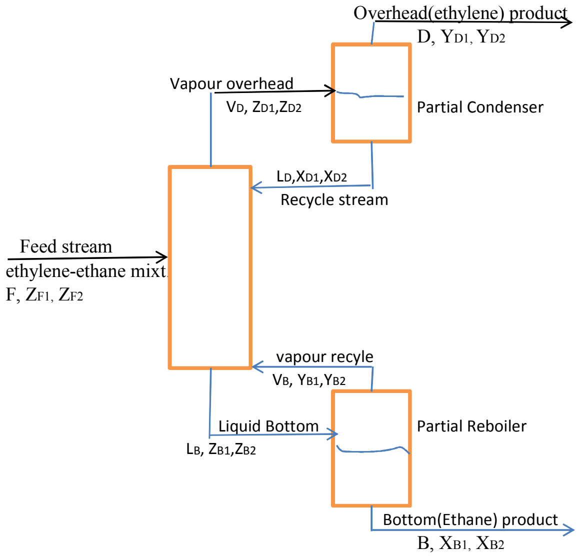


Fig. 1 : Distillation Separator for ethylene-ethane mixture

membrane Separator

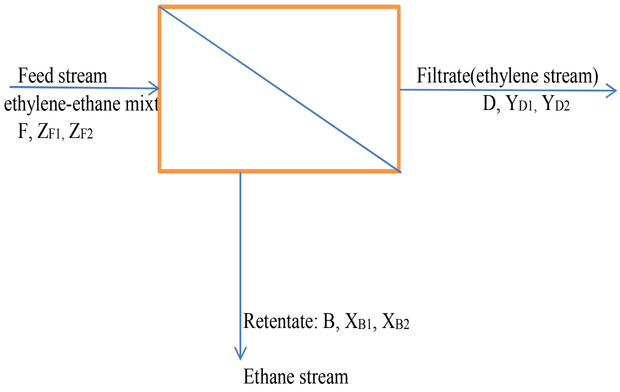


Fig. 2 : Membrane Separator for ethylene-ethane mixture

The membrane is the ideal crossflow Vapor permeation type. There are two types of membranes available namely: glassy polymers and rubbery membrane. In practice, almost all commercial plants use rubbery membranes, predominantly membranes made from silicone rubber (polydimethyl siloxane, PDMS). Rubbery membranes are preferred because they tend to have much higher permeability than glassy polymers, The vapor to be recovered permeates the membrane, leaving a residue stream enriched in raffinate, thus can produce an permeate stream almost completely depleted of the raffinate (Baker, 1994; Ohlrogge, 2001; Pinnau et al, 2004). In other words for a two component stream like ethane-ethylene mixture, it can separate them into pure petrochemical grades (>=99%), hence the choice of the configurations shown in fig.3

Thus compressor is used to feed the unit, and this accounts for the energy input into the unit. The column is broken in two sections. The top section is referred to as the rectifying section. The bottom section is known as the stripping section. The top product stream passes through a partial condenser. This effectively condenses part of the vapor overhead to liquid reflux. The bottom product stream uses a partial reboiler to vaporize part of the bottom product as recycle. Thus both the condenser and reboiler account for the input of energy into the column.

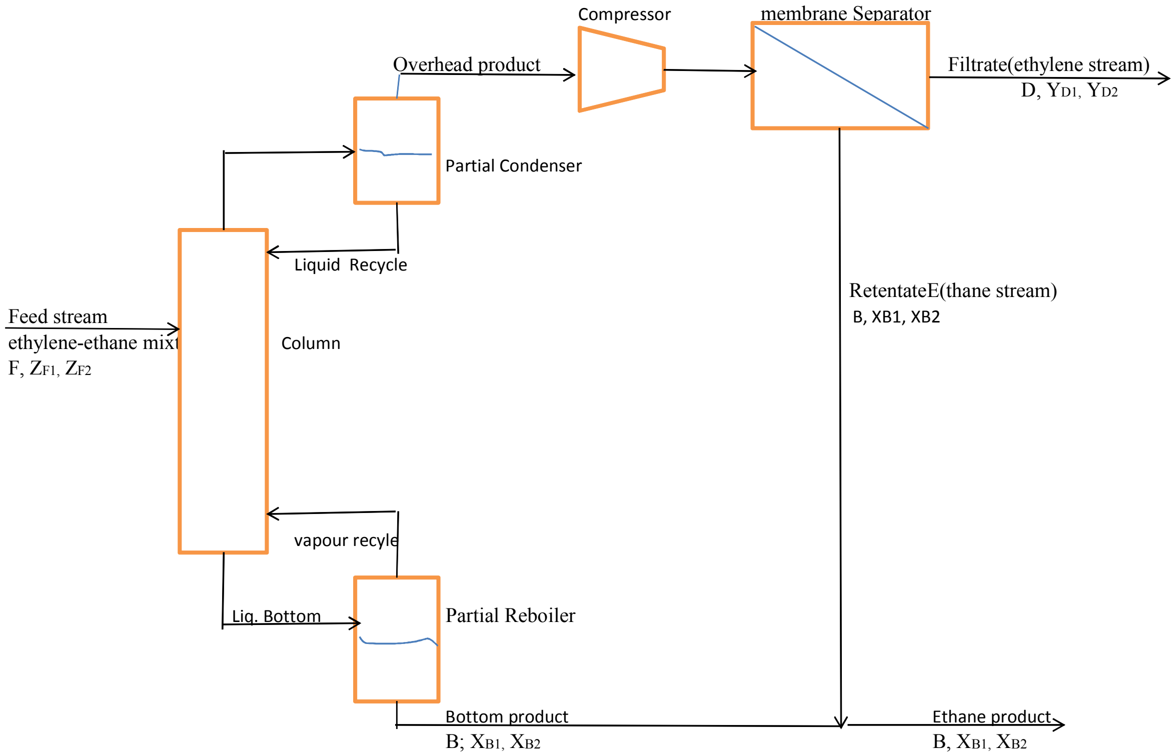


Fig. 3 : Distillation - Membrane Hybrid Separator for ethylene-ethane mixture

Deternination of the number of stages

The process specification/equilibrium are shown in table 1

Table 1: Process specifications for ethylene ethane mixture

Component 1:	ethylene
Component 2:	ethane
Feed composition:	
F(mols/hr):	100
Z1:	0.5
Z2:	0.5
operating pressure, Kpa:	853.157
feed enters as sat'd liquid: q =	1
Overhead product composition:	
Y_{D1}:	0.99
Y_{D2}:	0.01
Bottom product composition:	
X_{B1}:	0.01
X_{B2}:	0.99
relative volatility, α:	1.2
Critical properties:	
P_{C1},Kpa	5,118.33
P_{C2},Kpa	4,885.21
T_{C1}, K	283.06
T_{C2}, K	305.48

For constant relative volatility, the equilibrium relationship, relating the vapor phase concentration, Y to the liquid phase concentration, X is given by the relation:

$$Y = \alpha X / (1 + X(\alpha - 1)) \quad (1)$$

and represented in the equilibrium curve of fig 4

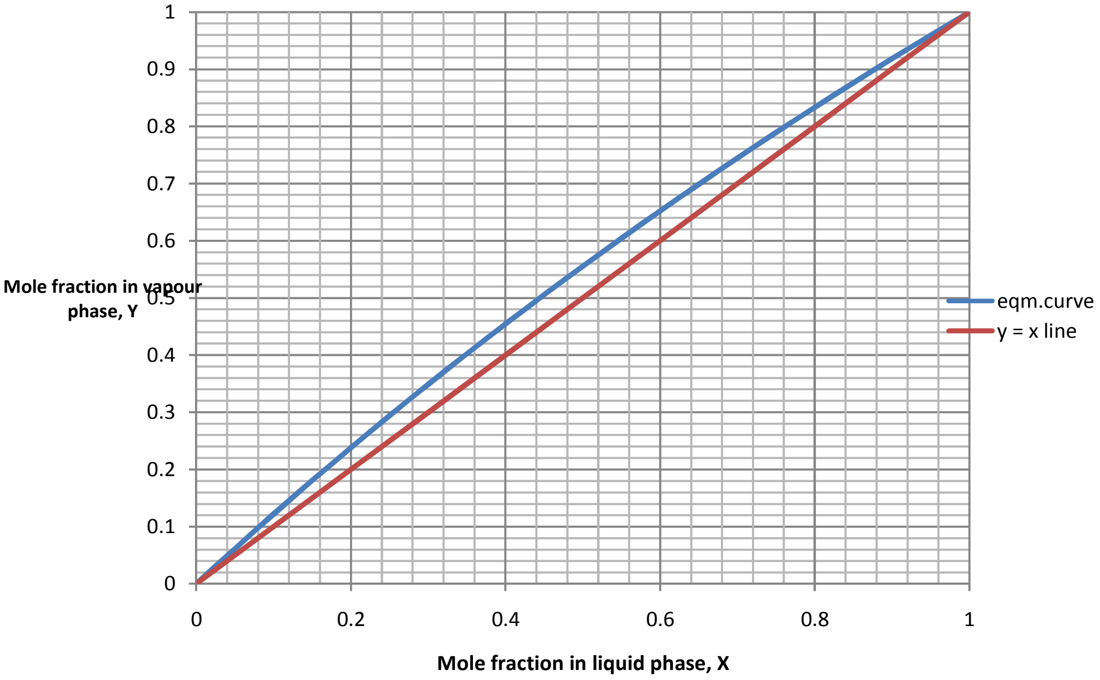


Fig.4: Equilibrium curve for Ethylene-Ethane system

The minimum curvature of the equilibrium curve is as a result of the low volatility ratio of ethylene-ethane mixture marked by their close boiling points(ethane :184.5 K; ethylene :169.4 K), hence the difficulty of separation. Thus with the high conversion desired, the use of McCabe –Thiele graphical method poses a lot of difficulties and clustering leading to inaccuracies. Thus shortcut methods-Modified Fenske-Underwood-Gilliland, FUG (Bausa, 1998) and rigorous stage by stage calculations are used to find the reflux ratio and the number of stages (Caballero, 2005; 2007 and 2009.) as follows:

For constant relative volatility, the Underwood equations allow to calculate the minimum vapor flow, V_{min} in a 'infinitely' large distillation column with a single feed distillate and bottoms streams, as in this case. The two equations are:

$$\Sigma (\alpha_i - Z_{Fi}) / (\alpha_i - \varphi) = 1 - q \quad (2)$$

$$V_{min} = D \, \Sigma (\alpha_i - Y_{Di}) / (\alpha_i - \varphi) \quad (3)$$

Where:
D = molar flowrate of distillate
q = Thermal feed quality (heat to vaporize 1 mol feed / molar latent heat of feed)
V_{min} = Molar flowrate of Vapor at minimum reflux
Z_{Fi} = Mole fraction of i in feed
Y_{Di} = Mole fraction of i in distillate
α_i = Volatility of component i relative to heaviest component
φ = Root of equation ($\alpha_{LK} < \varphi < \alpha_{HK}$)

D, the overhead product(distillate), is found from material balance for the entire unit (fig.3), based on equimolar feed rate of 100 moles, and petrochemical grade (99%) purity requirement for each product Thus minimum reflux ratio, R_{min}, is found by balance around the condenser:

$$R_{min} = L_{min} / D = (V_{min} - D)/D \quad (4)$$

$$L_{min} = V_{min} - D \quad (5)$$

$$R = R_{min} * 1.2 \quad (6)$$

Then rigorous stage-by-stage calculations are used to determine the number of stages (Caballero, 2005). The equations are:

For the rectifying section,
$Y_{N+1} = Y_N + R / (R + 1) * (X_N - X_{N-1})$ (7)
$X_{N+1} = Y_{N+1} / (Y_{N+1} + \alpha * (1 - Y_{N+1}))$ (8)
Starting from the partial condenser with $Y_N(1) = Z_{D1}$, $X_N(0) = X_{L1}$

For the stripping section:
$Y_M = \alpha * X_M / (1 + X_M * (\alpha - 1))$ (9)
$X_{M+1} = V_B / L_B * (Y_M - Y_{M-1}) + X_M$ (10)
Starting from the partial reboiler with $Y_M(0) = Y_{B1}$, $X_M(1) = Z_{B1}$
The iterations are continued until the concentrations equal the feed concentration, to give the number of stages required in both sections.

Energy Requirements by the column

The energy demand in distillation column is depend upon the reflux ratio and comprise of the combined energy requirements of the reboiler and condenser duties respectively (Marquardt, 2008). Thus the reflux ratio(and the corresponding number of stages) to achieve the desired purity of petrochemical grades(99%) in both top(ethylene) and bottom (ethane) streams are determined as follows:

Determination of Condenser heat loads

Being a partial condenser, as shown in fig 3, the heat load is the energy (latent heat) required to condense the reflux (at its dew point). Thus the condenser load, Q_{cond} is given by:

$$Q_{cond} = L_D.(\Delta HT_{dew}) \quad (11)$$

But th reflux ratio, R is given as:

$$R=L_D/D \quad (12)$$

thus
Thus eq.11 becomes:

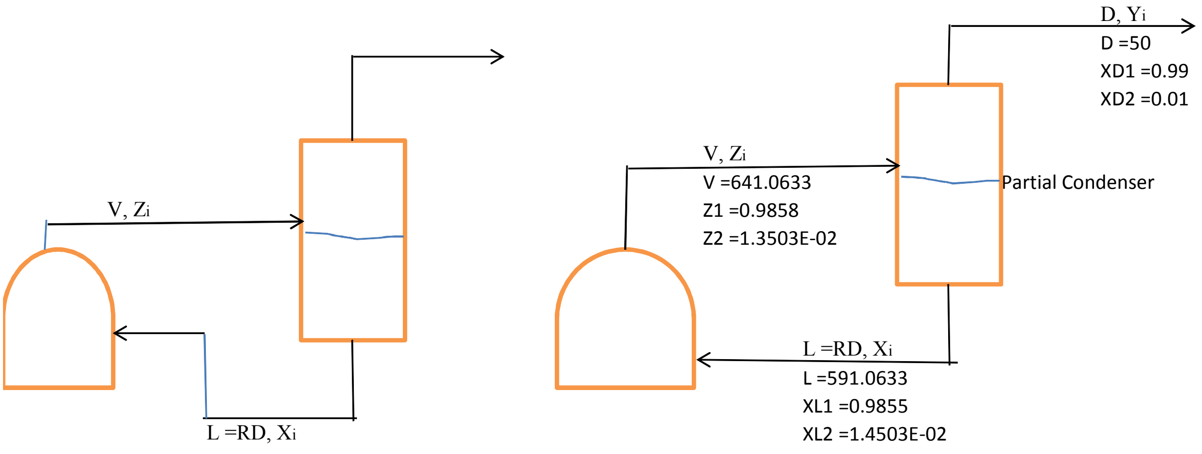
$$Qcond = RD. (\Delta HT_{dew}) \quad (13)$$

Where ΔH_{Tdew} is the enthalpy of condensation (at the dew point temperature, T_{dew}), given by Reidel correlation:

$$\frac{hvi,}{T_{dew}} = \frac{2.7(InP_{ci}-1)}{0.930T_{ri}} \quad (14)$$

Where

$h_{Vi} = \Delta H_{Tdew}$
T_{dew} = the dew point temperature (for condensation) or bubble point temperature (for vaporization)
P_c =critical pressure, atm.
T_c = critical temperature, K
$T_{r,}$ = reduced temperature, T/T_c
T_{dew} is determined from the equation
$\sum Z_i / K_i = 1$ (15)
Z_i, the mole fraction of the Vapor feed to the condenser is found from material balance and equilibrium relation around the unit as follows:



(a) Qualitative (b) Quantitative

Fig.5: Material balance around Partial Condenser

Overall and component balances around the unit, as well as equilibrium relationship gave equations 16, 17 and 18 respectively:

$V = D + L = D + R.D = D\,(1+R)$ (16)

$VZ_i = DY_i + LX_i = DY_i + RDX_i$ (17)

$Y_i = \alpha X_i/(1+X_i(\alpha-1))$ (18)

Combining and rearranging:

$X_i = Y_i/\{Y_i + \alpha(1-Y_i)\}$ (19)

$Z_i = (Y_i + RX_i) / (1+R) = (X_{Di} + RX_{Li}) / (1+R)$ (20)

Thus the balance around the condenser can be determined as shown in fig 5b

Thus eq.15 now becomes:

$(X_{D1} + R.X_{L1}) / \{K_1.(1+R)\} + (X_{D2} + R.X_{L2}) / \{K_2.(1+R)\} = 1$ (21)

Where K_i, the equilibrium constant for each component, is given by:

$K_i = \gamma_i P_i^0 / \varphi_i P$ (22)

where P_i^0 is the vapor pressure of each component, i found from the Antoine equation:

$P_i^0 = \exp(A_i - B_i /(T + C_i)$ (23)

and P is the total or operating pressure of the system; A_i, B_i, C_i are the Antoine constants for each component, i; γ_i and φ_i are the fugacity and activity coefficients, and are unity if the system is ideal in both phases as is the case for low pressures(< 10 atm = 1013.25 kPa), as in this process.

Using eqs.21 to 23, a convergence scheme is set to search for the temperature (the dew point) which satisfies the dew point equation (eq.15), using developed Excel template.

Determination of Reboiler heat loads

Being a partial reboiler, as shown in fig 3, the heat load is the energy (latent heat) required to vaporize part of the column bottom liquid, L, into Vapor, V_B.. Thus: the reboiler load, $Q_{Reboiler}$ is given by
:

$Q_{cond} = V_B.(\Delta H_{Tbubble})$ (24)

Where $\Delta H_{Tbubble}$ is the enthalpy of vaporization(at the bubble point temperature, T_{bubble}), also given by Reidel correlation.

T_{bubble} is determined fom the bubble point equation, which in this case of a binary mixture becomes:

$\sum K_iZ_i = 1$ (25)

Z_i, the mole fraction of the liquid feed to the reboiler, and V are found from material balance and equilibrium relations around the unit as follows:

Overall and component balances around the unit, as well as equilibrium relationship gave equations 26, 27 and 28 respectively:

$L = V_B + B$ (26)

$LZ_i = VY_i + BX_i$ (27)

$Y_i = \alpha X_i/(1+X_i(\alpha-1))$ (28)

Also to know and relate the Vapor recyle rate to the liquid rate ($V_b = V/L$), one needs the balance for the feed tray. Since the feed is a saturated liquid in this case ($q = 1$), the liquid rate to the bottom of the column is the sum of the feed and the reflux. Thus:

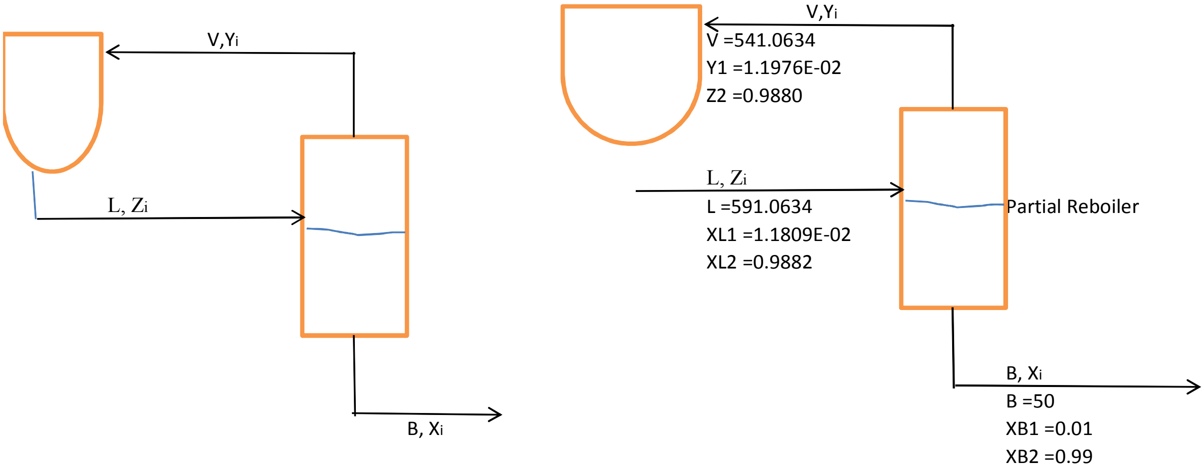
$L = F + RD$ (29)

Combining and rearranging:

$V_B = F + RD - B$ (30)

$Z_i = (VY_i + B\,X_i) / (V+B) = (V_B.Y_{Bi} + B\,X_{Bi}) / (V_B + B)$ (31)

Thus the balance around the condenser can be determined as shown in fig 6b



(a) Qualitative (b) Quantitative

Fig.6: Material balance around Partial Reboiler

Thus the bubble point equation.. eq. 25 becomes:

$K_1.(V_B.Y_{B1} + B\,X_{B1}) / (V_B + B) + K_2.(V_B.Y_{B2} + B\,X_{B2}) / (V_B + B) = 1$ (32)

Using eqs.31 and 32, a convergence scheme was set to search for the temperature (the bubble point) which satisfies the bubble point equation (eq.25), using developed excel template. Thus

$Q_{Reb} = (F + RD - B).\Delta H_{Tbubble}$ (33)

Energy Requirements by the membrane

Fig.7 shows the membrane only process.

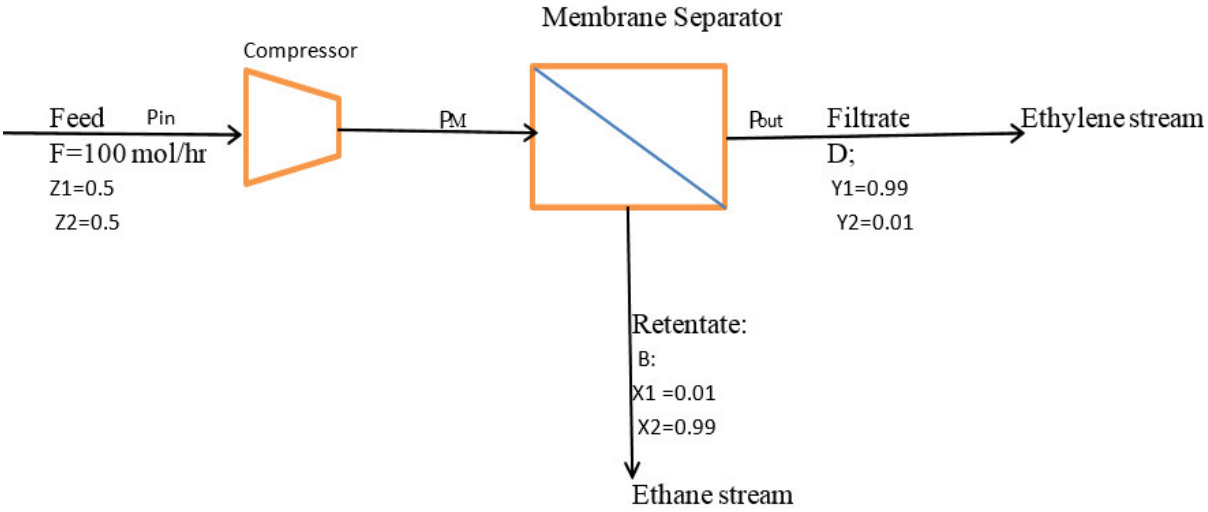


Fig.7: Membrane only process

Creation of the pressure difference between the feed and permeate sides is the only work /the total energy requirement for membrane separation (Caballero, 2007; 2009; 2014). This compressor work is determined as follows:

For a fixed area of the membrane we now want to determine the feed pressure, hence the energy requirements for the membrane (in terms of compressor power) to achieve the same purity. The equations are(Caballero, , 2009):

$P_M = \{F.Z_1\,(Perm^*\,Area) + Y_1.\,P_{out}\}/X_1$ (34)

.

$$W_{comp}\left(\frac{KJ}{Kmol}\right) = RT.\frac{k}{\eta(k-1)} \times \left[\left(\frac{P_M}{P_{in}}\right)^{\frac{(k-1)}{k}} - 1\right] \qquad (35)$$

where
R =gas constant, $J/(kg.K)$
T =inlet gas temperature, K
P_M is membrane feed pressure;
P_{in}= absolute inlet pressure, kPa; and P = absolute discharge pressure, kPa.; $k = C_p/C_v$
Therefore, the adiabatic power, Qm becomes:
$Q_M = F.W_{comp}$ (36)

Thus to determine the power requirement of the membrane separator requires knowing the vapor feed rate to the membrane, Vm, and is estimated as follows:
Recall that using a distillation only method of separation to achieve the desired purity of 99% requires a high reflux ratio of 8.7 (designated as $R_{99\%D}$), and that implies no feed to the membrane (Vm = 0). Alternatively using only the membrane to separate the mixture, implies the entire feed stream enters the membrane (Vm = F), thus no feed to the column, hence no refux ratio ($R_F = 0$). Therefore there is an inverse relationship between them as shown in fig.3.45 below:

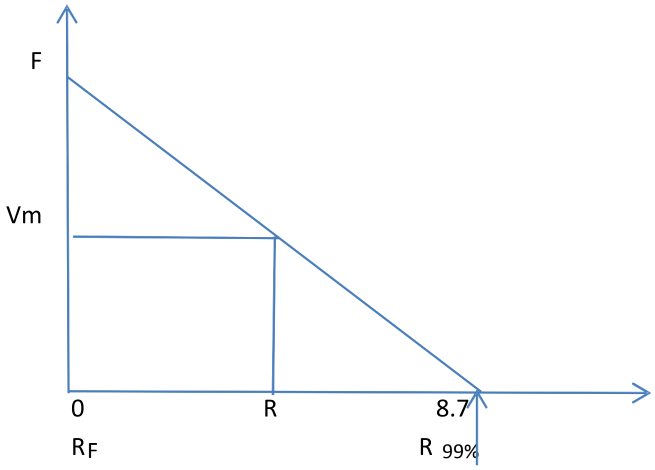


Fig.8: Vapor flow to the membrane vs reflux ratio

Thus
$(F - V_M)(R_F - R) = (F- 0)/ (R_F - R_{99\%})$ (37)
Where:
V_M is the molar Vapor feed to the membrane
R_F is the reflux ratio when the membrane feed is F. Since it corresponds to membrane-only process, $R_F = 0$
$R_{99\%}$ is the reflux ratio at 99% distillate purity and corresponds to distillation-only process when no feed to the membrane
Thus eq. becomes:
$V_M = F (1- R / R_{99\%})$ (38)
Thus using eq. 38, in collaboration with eqs. 34 to 36 will give the pressure needed by the compressor as well as the corresponding power requirement.

Simulations
By varying the distillate compositions between the feed composition (Z_1=0.5) and the desired purity of $X_{D1} = 0.99$), eqs. 1 to 38, along with the process specification/thermodynamic data (table 1), are used to calculate the corresponding parameters namely: reflux ratio, number of stages, distillate flow, bottom product flow/compositions, condenser duty, reboiler duty, membrane compressor feed power, and hence the total energy requirement (Verhoef et al, 2008; Caballero, 2006; Kookos, 2003).

Results and Discussion

The simulation results are represented in Figs.9 to 14.

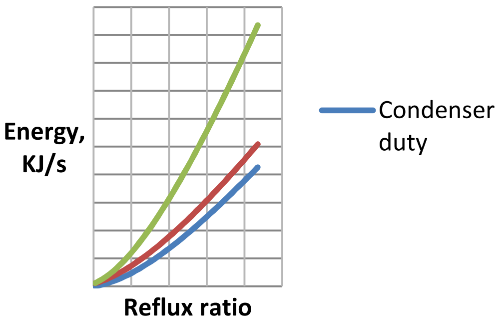


Fig. 9: Column energy requirements vs. Reflux ratio

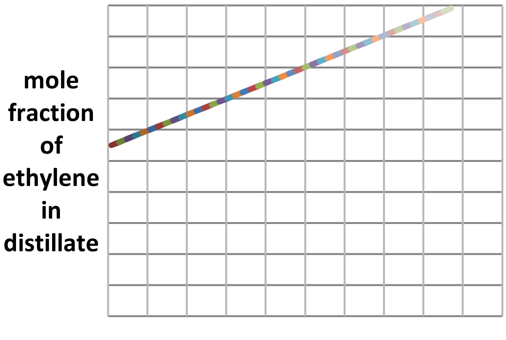


Fig. 10: mole fraction of ethylene in distillate vs. Reflux ratio

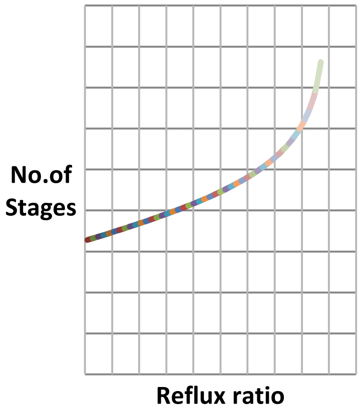


Fig. 11: No.of stages vs. Reflux ratio

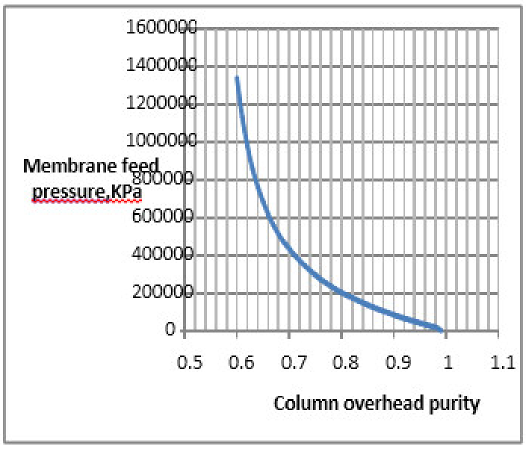


Fig.12 Membrane feed pressure vs. column overhead purity

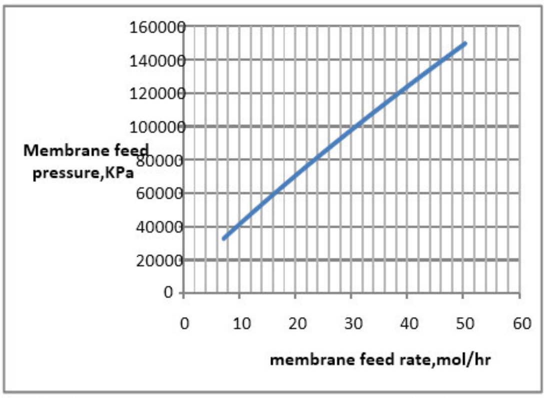


Fig.13 Membrane feed pressure vs .membrane feed rate(Vapor load)

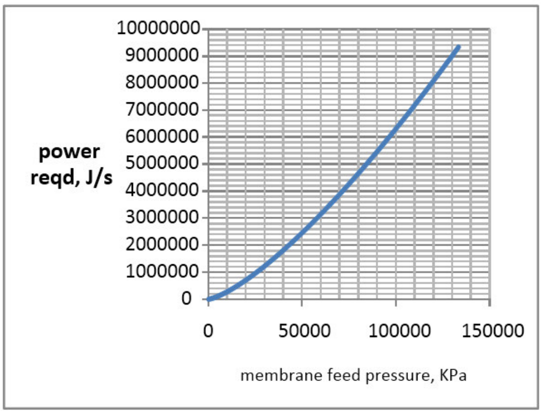


Fig.14 Membrane feed pressure vs vapor load

Fig 9 shows that as the reflux ratio increases, both the condenser and the reboiler duties increase, and that implies that the energy required by the column also increases. This energy increase implies increased purity of product (fig.10) and also increase in the number of stages (fig.11). Similarly fig 12 shows the inverse relationship between the membrane feed pressure and the column overhead product purity. Since the membrane shares the load with the column, this implies as the purity increases, less load is required by the membrane, until the desired purity of 99% when there will be no load to the membrane, hence no gauge pressure feed. However as the load (feed rate) increases, the feed pressure also increases (fig.13), and this also increases the power requirement of the membrane (fig.14)

Of particular interests are figs. 11 and 12, showing number of stages vs. reflux ratio and membrane feed pressure vs. column overhead purity. Fig 12 shows the excessive pressure needed for a membrane only process, that is, when the purity is low and tends towards the original feed composition(50% in this case), which translates to more compressor power(fig14). Similarly fig.11 showed that the most difficult separation is usually performed at the end of the column sequence, implying more energy spending as purity tends towards the desired petrochemical grade (99%). The above analysis showed that both the distillation-only and membrane-only processes lead to very high energy and power requirements respectively. Thus if the distillation process can be truncated midstream and membrane used to complete the remaining separation, then we should expect overall less energy spending., and that is exactly what happened as shown in the analysis below.

Fig. 15 is the combined energy expenses for the hybrid process.

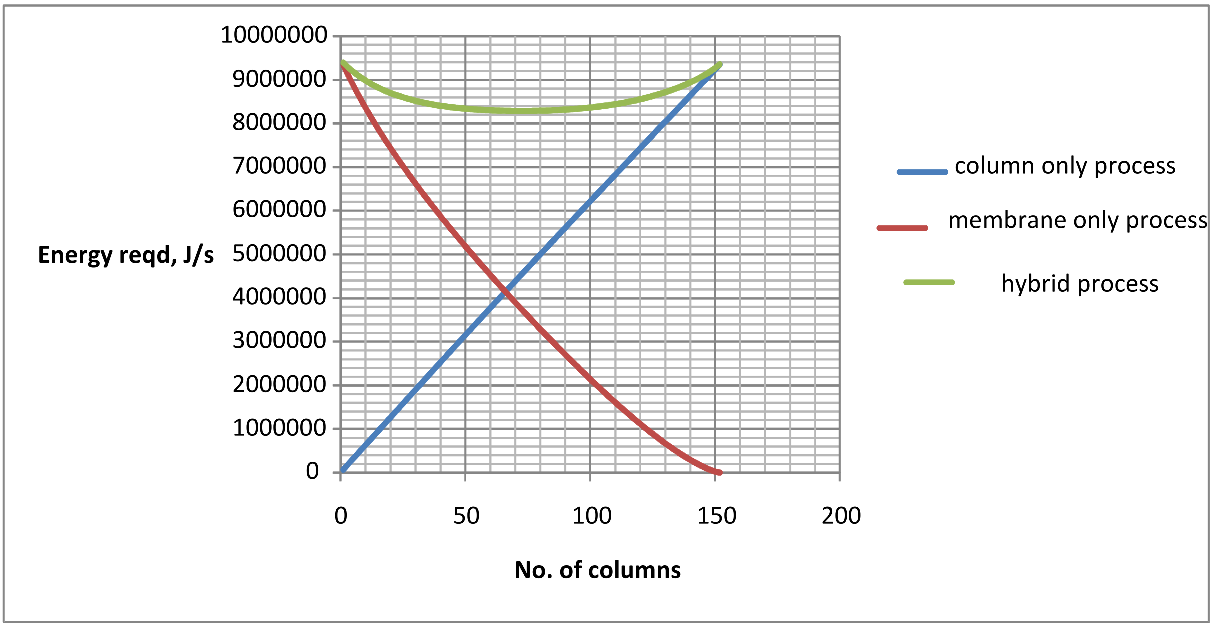


Fig. 15: Energy requirements for the column, membrane and hybrid processes vs total column stages

By regression analysis, the following model was fitted to the hybrid energy requirement as a function of the number of column stages:

$Y = 9406538 - 48377.32 X + 785.3503 X^2 - 5.909527 X^3 + 1.855742E\text{-}02 X^4$ (39)

Y=hybrid energy requirement, X = number of column stages, with regression coefficient = 0.9996374 showing a good fit to the simulated results as shown in fig, 16

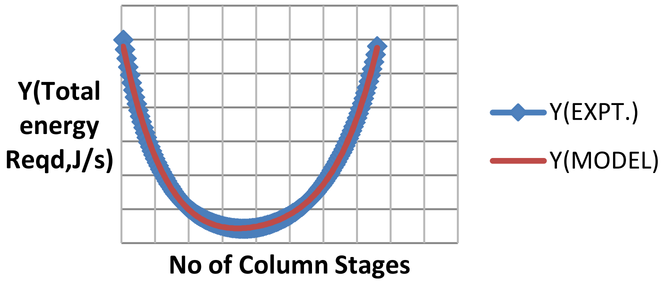


Fig.16: Hybrid energy vs number of stages(expt. vs. model)

The model goes through a minimum, thus we need to determine this optimum energy requirement for the hybrid configuration.

Optimizing the Model

Using Newton's gradient method given by the convergence scheme::

$$X_{n+1} = X_n - Y'(X_n) / Y(X_n) \quad (40)$$

The optimum point occurred at 69 Column Stages corresponding to a total energy spending of 8287KJ/s

Recall that the model for the process is:

$$Y = 9406538 - 48377.32 X + 785.3503 X^2 - 5.909527 X^3 + 1.855742E\text{-}02 X^4 \quad (39)$$

Using only a distillation column to achieve the desired purity, requires152 columns, corresponding to X = 152 in the model, which on evaluating, gives a Y or total energy spending of 9350KJ/s

However by using only a membrane to achieve the desired purity, corresponding to X = 0 in the model, which on evaluating, gives a Y or total energy spending of 9407KJ/s

From the foregone analysis, it has been shown that using a novel column-membrane hybrid configuration consumes lesser energy than using individual units alone to achieve the same degree of purification, thus more economical.

Conclusion

The conventional separation of light hydrocarbon mixtures like ethane-ethylene mixture by cryogenic distillation is very high energy consuming, as a result of high reflux ratio (and large number of stages) required, thus creating a high load on the condenser and reboiler (the energy consuming units of the column), especially when the distillation goes to extreme end to produce pure petrochemical grades. However using a work driven vapor permeation membrane only is also not feasible since this will require excessive membrane feed pressure (and large areas of membrane), thus very high power requirement. Hence hybridization of the membrane with distillation column -by load sharing- offers a better alternative.

Thus in this case of ethane/ethylene binary system, in order to separate the mixture in equal mass fraction to get 99% distillate and 99% at bottom, while normal cryogenic distillation will require 152 columns, corresponding to a total energy spending of 9351KJ/s, a membrane-only process requires a total energy spending of 9407KJ/s. But using the hybrid requires only 69 column stages and a total energy spending of 8287KJ/s. Hence hybridization of the membrane with distillation column is quite satisfactory in reducing the energy consumption.

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