

DIAGNOSIS OF THERMODYNAMIC EFFICIENCY IN HEAT INTEGRATED DISTILLATION

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Abstract—For the systematic diagnosis of the thermodynamic efficiency in dual column distillation with heat integration of practical modes, a computing scheme is developed in conjunction with the method of calculation of exergy and the modularized reduced-order model, which are discussed elsewhere by the present authors [1,2]. The thermodynamic efficiencies of 9 types of heat integration modes are actually calculated and compared with that of a distillation column without heat integration using 3 different kinds of hydrocarbon mixtures, i.e., tertiary, quaternary, and octanary.

The results show that the separation energy demand can be reduced by 15% when single heat integration through the reboiler of the first column is made, and about 20% when dual integration through the condenser and the reboiler of the first column is made.

INTRODUCTION

Separation processes especially such as distillation play a prime role in chemical and related industries. However, as pointed out frequently, these processes consume vast amount of energy for the separation of fluid mixtures and the cost of energy is ever increasing. It is, therefore, urgent to develop energy efficient separation process and/or to improve existing processes. During the past years, many works have been carried out for the development of energy saving schemes in process industries [3-9]. Even though distillation process, as a major energy consumer with no competitive alternative, has been a prime target for further study and improvement, it is still designed with only meager amount of data leaving a lot to be discovered for the ways to energy conservation [3,10,15].

There are many rooms of improvement for the energy conservation within the existing distillation columns. They are the reduction of temperature difference in reboiler and condenser, recovery of heat by side stream withdrawal, application of heat pump and vapor recompression, splitting the column, adjustment of pressure drops, and feed-product-bottom heat integration [10]. Among them, the heat integration by utilizing heat exchanger is accepted as a practical candidate due to its easy installation to existing distillation systems.

The aim of this article is to discuss the systematic method for diagnosis of the degree of efficient energy use in an existing distillation system with or without

heat integration. The principle of the method is based on the modularized reduced-order simulation technique and exergy calculation of equilibrium mixtures [1,2].

THERMODYNAMIC ANALYSIS OF A DISTILLATION COLUMN

1. Thermodynamic Definition of Column

The basic elements of an ordinary distillation column consist of a column internal, reboiler and condenser. A feed preheater and/or product cooler is commonly installed for other process requirements. For the thermodynamic analysis, a distillation system can be defined as being consisted of three subsystems as shown in Figure 1. One is the column internal, and the others are the exergy sink, and source. The exergy sink streams from the column internal are the products, streams to be reboiled, and cooling media. The exergy source streams to the column internal are the feed, streams to be condensed and heating media. Also, exergy loss in the column internal can occur through the fluid mixing, heat transfer with latent and sensible heat, and heat leaks across the boundary.

In comparing the results obtained by the heat integration, the temperatures and pressures of influent and effluent process streams at the column boundary are assumed to be the same as the reference absolute temperature and pressure. When a distillation system is regarded as a composite thermodynamic system, the overall exergy flow can be analyzed in terms of the re-

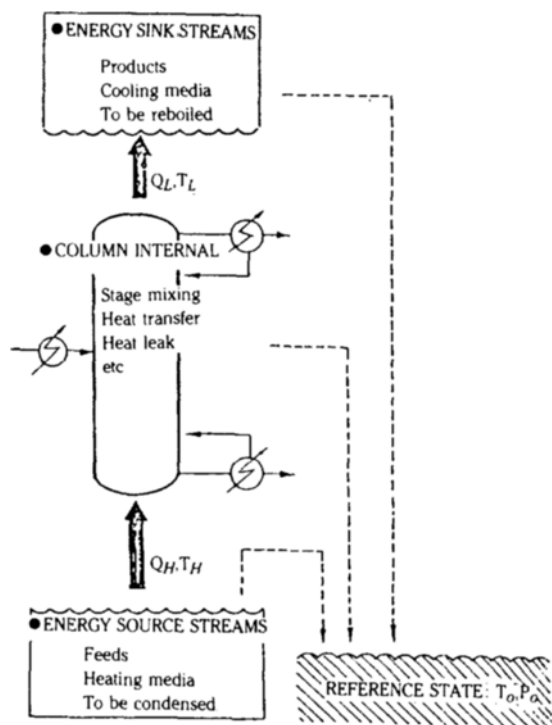


Fig. 1. A binary distillation system and thermal energy transfer subsystems.

quirement of minimum separation energy and total exergy input across the system boundary. Therefore, a distillation system can be redefined as being composed of a system-boundary-reference as shown in Figure 2, and then, a method of exergy balance can be applied to a general control volume.

2. Exergy Balance

As shown in Figure 2, a distillation column can exchange material and thermal energy with the reference state. As discussed elsewhere by one of the present authors[11,12], an exergy balance can be constructed by combining first and second laws with thermodynamic exergy dissipation. The resulting equations are summarized by

$$\frac{d}{dt} (mb - P_0 v)_{sys} = \sum_i W_{min, ro, i} + \sum_j W_{net, j} - \sum_k L W_k \quad (1)$$

$$W_{min, ro} = \Delta B = \sum (mb)_{prob} - \sum (mb)_{feed} \quad (2)$$

$$W_{net} = \sum_i \left(1.0 - \frac{T_0}{T_i}\right) \cdot q_i + \sum_i W_i \quad (3)$$

$$\Delta B = \Delta (mh) - T_0 \Delta (ms) \quad (4)$$

where b represents molar exergy, m the mass flow rate, W_{min} the minimum separation energy relative to

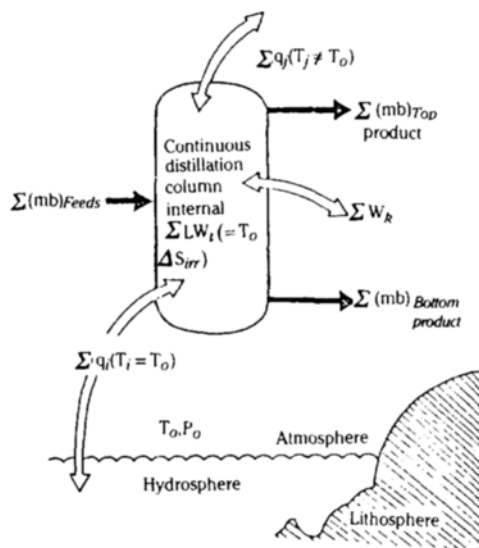


Fig. 2. A distillation system and exergy interaction across the boundary.

the reference temperature, T_0 , W_{net} the overall Carnot work and LW the lost exergy due to irreversible entropy generation $[T_0 \Delta (mS)_{irrev}]$. W_i denotes mechanical work mode such as compression and expansion when a heat pump is installed. In the present study, this work term is neglected. The values of 273.16°K and 1.013 Bar are used for the reference state T_0 and P_0 in the computation of thermodynamic property, respectively.

3. Definition of Thermodynamic Efficiency

Since we are only concerned with a steady state operation, Eq. 1 becomes as follows

$$\sum W_{min, ro} + \sum W_{net} - \sum L W + \sum (mb)_{utilities} = 0 \quad (5)$$

The third term accounts for the exergy losses due to process irreversibilities, and the last terms accounts for process utilities which could be positive or negative depending on whether it is a net exergy input or output. In the next section, various sources of process exergy losses will be briefly discussed.

Based on the exergy balance, we can define a thermodynamic efficiency. Suppose that minimum separation energy is positive, i.e., work must be done on the system to obtain the desired separation as is usually the case. Then, a thermodynamic efficiency may be stated as follows

$$\eta = \frac{\sum W_{min, ro} + \sum (mb)_{utilities}}{\sum W_{net}} \quad (6)$$

Examination of Eqs. 5 and 6 show that for a reversible distillation the lost exergy will be zero giving a pre-

dicted efficiency of unity. For actual distillation, this efficiency will be far less than unity. For the sake of simplicity the process utility term is not included in the computation of thermodynamic efficiency.

SOURCES OF EXERGY LOSSES

In an actual distillation, the irreversible exergy loss can arise from numerous sources such as temperature driving force in heat transfer, mass transfer and mixing. And these sources of irreversibilities give rise to direct effect of lowering thermodynamic efficiency. In the following discussions, we attempted to identify and interpret those losses of exergy associated with the distillation processes. This identification of exergy sources associated with subsystems can clearly pinpoint the subsystem to be modified.

1. Exergy Loss Through Heat Leak

As we know well, the column internal temperature is usually much higher than the reference temperature, T_o . The exergy loss due to heat leak from system to surrounding can not be neglected, especially in winter season. Therefore, we need a thermal insulation with an appropriate material of low thermal conductivity. If we neglect the heat loss due to thermal radiation, the exergy loss through heat leak can be defined as

$$LW_{hl} = \sum (1 - \frac{T_o}{T_i}) U \cdot A_T \Delta \bar{T}_{i,m} \quad (7)$$

where U denotes an overall heat transfer coefficient, A the heat transfer external area for each subsystem. The subscript hl represents heat loss.

Whenever appropriate data on A and U are available for the construction material, an accurate analysis of this quantity can be carried out. We did not considered this type of exergy loss in this work.

2. Exergy Loss in Heat Exchanger

For the sake of simplicity, we considered a two stream shell-and-tube heat exchanger involving only two temperature levels, T_h and T_c relative to the reference temperature T_o . With reference to Figure 4, Eq. 1 is simplified as follows

$$LW_{hx} = T_o \{ \Delta (ms)_{out} - \sum (ms)_{in} \} = \{ (1 - \frac{T_o}{T_h}) Q_c - (1 - \frac{T_o}{T_c}) Q_h \} \quad (8)$$

In arriving at Eq. 8, we regard the enthalpy interaction is conserved ($b = -T_o S$). The first term on the right hand side of Eq. 8 represents the Carnot work required to transfer heat Q over unit time to the heat stream T_h from the reference temperature T_o . The second term with the minus sign represents the work derived from

a Carnot engine operating between T_c and T_o to remove heat over unit time from the cold stream at T_c . Eq. 8 is applicable for the case of $T_h > T_c > T_o$.

3. Exergy Loss in Column Internal

If we consider an ordinary adiabatic distillation column which involves no mechanical work, the only heat exchanges with the reference are through the condenser at T_c and the reboiler at T_r . Applying Eq. 1 to such a system,

$$LW_{sm} = LW_{sm}^{net} - \{ \int_0^{q_r} (1 - \frac{T_o}{T_{ri}}) dq_{ri} + \int_0^{q_c} (1 - \frac{T_o}{T_{ci}}) dq_{ci} \} \quad (9)$$

$$LW_{sm}^{net} = (1 - \frac{T_o}{T_c}) Q_c - (1 - \frac{T_o}{T_r}) Q_r \quad (10)$$

Note that LW accounts for all the irreversibilities which occurs in each stage of the column. The first term on the right hand side of Eq. 9 represents the net work required to transfer heat Q_c from the condenser at T_c to the reference at T_o and to transfer heat Q_r to the reboiler at T_r from the reference at T_o . The last two terms in Eq. 9 represent the net work required to transfer heat q_r from the rectifying section integrated over its temperature distribution with respect to T_o and to transfer heat q_c to the stripping section integrated over its temperature with respect to T_o . The difference between these two net work terms measures exactly the cumulative exergy losses over all the stages. Any heat integration which reduces this net work difference will reduce exergy losses due to stage mixing in the column internal.

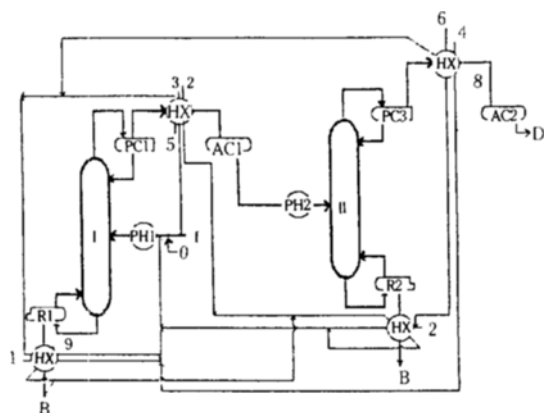
HEAT INTEGRATION

1. Method of Heat Integration

To illustrate the utilization of exergy analysis in process modification studies, we chose a heat integrated dual distillation column as our system as shown in Figure 3. Various multicomponent mixtures were tested as feeds. We considered 10 types of heat integration modes. These heat integrations are based on the idea of integrating the available heat in reboilers and condensers of column I and II. The first 5 types of heat integrations (Type 0-4) represent single heat integration and the other 5 types represent dual heat integration. In the subsequent analysis, it is assumed that the feed enters the column I at saturated liquid state through the preheater.

2. Temperature Gain in Heat Exchanger

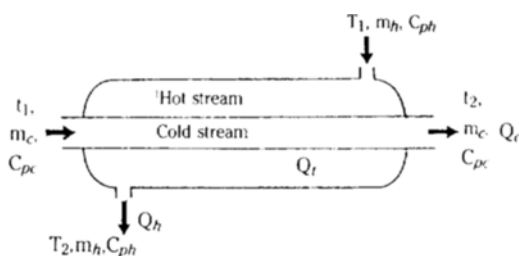
The method of heat integration considered here is based on the heat gain of cold feed stream through



- SYMBOLS: PC-partial condenser AC-accumulator
R -reboiler HX-heat exchanger
- TYPES OF HEAT INTEGRATIONS:
- 0-no heat integration
- 1-reboiler in column I
- 2-reboiler in column II
- 3-condenser in column I
- 4-condenser in column II
- 5-condenser and reboiler in column I
- 6-condenser and reboiler in column II
- 7-condenser in column I and reboiler in column II
- 8-condenser in column II and reboiler in column I
- 9-reboiler in column I and reboiler in column II

Fig. 3. Schematic diagram for various heat integrations (0-4: single integration loops, 5-9: dual integration loops).

reboilers and condensers. With emphasis on practical extension of present approach, we put a limit on the maximum attainable temperature when cold organic liquid mixture pass through the hot stream media. As shown in Figure 4, we assign Q_c to the heat gain by cold stream, Q_h to the heat loss by hot stream and Q_t to overall heat gain through heat transfer area in shell-and-tube heat exchanger. According to Burkhust and Harker[13], the most important variable in the design of heat exchanger is AU (product of heat transfer area and overall heat transfer coefficient). When the magnitude of AU becomes small, the outlet temperature (t_2) of cold stream decreases. Also, if m_h or m_c become small, AU decreases. When a shell-and-tube heat exchanger is used for hydrocarbons fluids, the average value of AU lies in the range of 600 cal/Kw. With this criterion, we first assume the value of AU , then calculate t_2 and T_2 by Gauss-Siedal numerical method using the expressions of Q_h , Q_c and Q_t given in Figure 4. Then, AU is updated with previous values of t_2 and T_2 . The results show that the temperature gain of cold hydrocarbon streams are in the range of 6-9°C within practical temperature range of inlets and outlets. In all



$Q_t = AU \Delta t_m$: amount of heat transferred through the wall
 $Q_c = m_c C_{pc}(t_2 - t_1)$: amount of heat gained by the cold streams
 $Q_h = m_h C_{ph}(T_1 - T_2)$: amount of heat lost by the hot streams

Fig. 4. Specifications of temperature gain in heat exchanger.

types of heat integrations, we adopted the maximum temperature gain of 6°C for single heat integration.

SIMULATION STUDIES

1. Simulation Method

Two of the essential ingredients for rational audit of exergy and related thermodynamic efficiency for a given type of heat integrated distillation column are the accurate knowledges of the required exergy of vapor-liquid equilibrium mixtures and the types of process simulation method utilized.

In the evaluation of the thermodynamic efficiency, the physical properties and the exergy function of vapor-liquid equilibrium mixtures are calculated based on the modified data bank[1], which is originally proposed by Prausnitz et al.[14] as a conventional process design purpose. For the column simulation, the modularized reduced-order approximation method developed by the present authors[2] as well as the rigorous Newton's method are used for the purpose of compatibility test and accuracy comparison.

The simulation studies were carried out for each type of heat integration modes as shown in Figure 3. The program is made so that the diagnosis of the thermodynamic efficiency is possible for both cases with or without preheater.

1-1 When feed preheater is not installed

- 1) Assign types of heat integration.
- 2) Assume the feed is at room temperature.
- 3) Simulate column I first, and then, subsequently column II. The top product of column I enters column II as feed.

4) Estimate iteratively the values of vapor composition(x), temperature(T), liquid composition(y), flow rate of top and bottom product(V and B) until the feed temperature calculated with these estimated values converge to the assumed feed temperature.

5) Calculate feed temperature gain, exergies for each subsystems, heat duty, and Carnot work with respect to reference state.

6) Calculate overall exergy loss and thermodynamic efficiency.

7) Return to step 3) by changing the type of heat integration mode.

1-2. When feed preheater is installed

All the algorithmic steps are identical with above case 1-1 except the feed to the column I enters a saturated liquid.

2. Sample Mixtures and Normal Operating Conditions

The compositions of three types of multicomponents mixtures and the normal operating conditions are given in Table 1.

Since the main emphasis was given to the analysis of thermodynamic efficiency, we were not much concerned about the degree of separation of top and bottom products. Before feed mixtures are introduced into the feed stage, the feed state is maintained as saturated liquid by passing heat integration loops and preheater. To compare the effect of heat integration mode on the

thermodynamic efficiency, the same values for heat duties of both and reboilers were used for all types of heat integration. The feed flow rate was assigned as 100 mole/hr in convenience. Also, the number of stages for both columns were assumed to be twenty and the reflux ratios for normal conditions for the columns were fixed to two. In each mixture, the columns were assumed to be operated at atmospheric pressure. The reflux ratios of both column are varied combinatorially from two to four as shown in Table 4.

RESULTS AND DISCUSSION

The results of computer simulation are shown in Table 2-4 and Figure 5-7 for all the types of heat integration. The RMS represents the results obtained by rigorous simulation method and MPA the results by modularized polynomial approximation method. The difference between the top and bottom compositions simulated by the two methods is within the range of 1 to 7%. For the efficiency analysis the difference ranges from 1 to 5% for all the three types of mixtures. This indicates that the use of MPA technique is advanta-

Table 1. Feed and column specification at normal operation for simulated mixtures in dual column distillation

Specifications	system number					
	I		II		III	
Feed						
components	acetone	(0.3)	benzene	(0.25)	carbontetrachloride	(0.1)
and mole	methanol	(0.3)	cyclohexane	(0.25)	ethanol	(0.1)
fraction	ethanol	(0.4)	methylcyclohexane	(0.25)	n-propanol	(0.1)
			n-hexane	(0.25)	cyclohexane	(0.1)
					toluene	(0.2)
					acetone	(0.1)
					methanol	(0.1)
					water	(0.2)
temperature (°K)	335.58		352.97		343.68	
phase state	saturated liquid					
flow rate (mol/hr)	100					
Distillate						
flow rate (mol/hr)						
first column	50					
second column	25					
Column condition						
feed stage	20					
reflux ratio						
first column	2					
second column	2					

Table 2. Exergy analysis for heat integrated distillation system of acetone-methanol-ethanol (I)

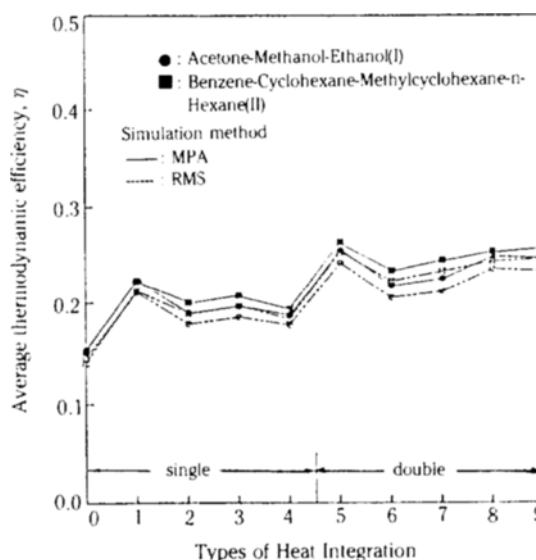
Type of heat integration	no. of heat integration	availability (J/mol) $\times 10^{-6}$		net work (J/mol) $\times 10^{-7}$		preheater heat duty (J/hr) $\times 10^{-6}$		stage mixing (J/mol) $\times 10^{-6}$	
		RMS	MPA	RMS	MPA	RMS	MPA	RMS	MPA
0	0	0.196	0.198	0.272	0.259	0.428	0.428	0.976	0.838
1	1	0.285	0.286	0.270	0.257	0.322	0.323	0.976	0.838
2		0.242	0.245	0.271	0.258	0.375	0.374	0.976	0.838
3		0.252	0.254	0.271	0.258	0.364	0.364	0.976	0.838
4		0.241	0.242	0.271	0.258	0.377	0.377	0.976	0.838
5	2	0.325	0.327	0.269	0.256	0.269	0.270	0.976	0.838
6		0.278	0.281	0.271	0.257	0.331	0.330	0.976	0.838
7		0.287	0.290	0.270	0.257	0.320	0.319	0.976	0.838
8		0.317	0.319	0.270	0.257	0.280	0.281	0.976	0.838
9		0.314	0.316	0.270	0.257	0.285	0.284	0.976	0.838

Table 3. Thermodynamic efficiency for heat integrated distillation

Type of heat integration	system I		system II		system III	
	RMS	MPA	RMS	MPA	RMS	MPA
0	.144	.153	.140	.151	.115	.135
1	.211	.223	.213	.224	.178	.135
2	.179	.190	.189	.201	.160	.178
3	.185	.196	.197	.208	.166	.184
4	.177	.188	.184	.194	.151	.169
5	.242	.255	.253	.264	.215	.232
6	.205	.218	.223	.234	.188	.205
7	.212	.225	.233	.245	.200	.217
8	.235	.248	.244	.254	.205	.221
9	.233	.247	.246	.257	.210	.227

geous over RMS in view point of computing time.

The simulation results of exergy audits for all 10 types of heat integration modes are given in Table 2 for acetone-methanol-ethanol mixture as an illustration. From the results, it is obvious that the heat integration type 1(integrated through the reboiler in column I) for single integration loop and type 5(integrated through the condenser and the reboiler of the column I) for dual loop are the most appropriate candidate for the heat integration to improve thermodynamic efficiency. In Table 3, simulated thermodynamic efficiency for all 10 types of heat integration modes for the three types of mixtures are summarized. Together with the results in Table 2 the result shows that the efficiency mainly depends on the column in-

**Fig. 5. Effect of heat integration on the average thermodynamic efficiency for reflux ratio (2,2).**

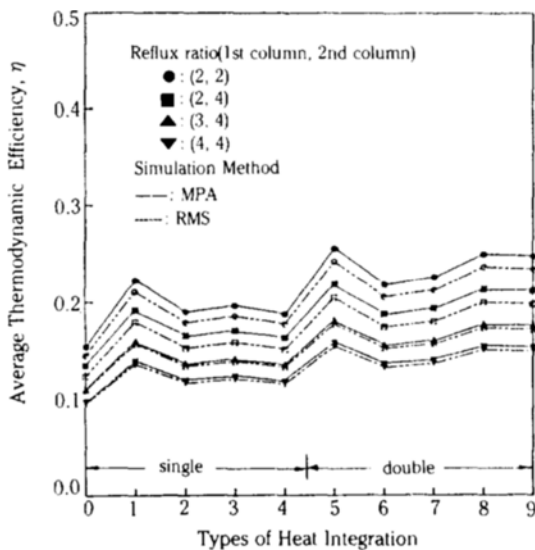
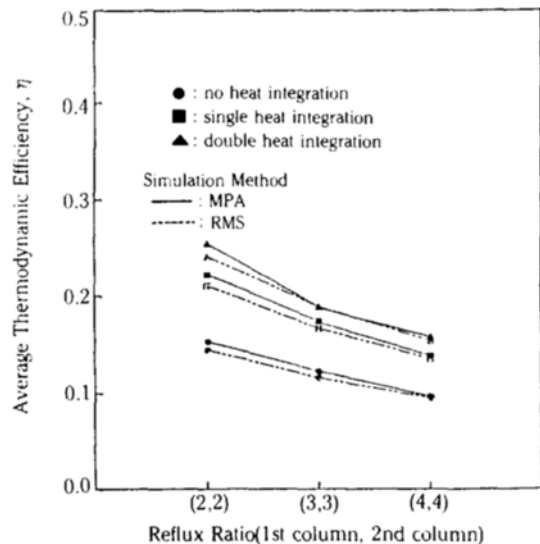
ternal temperature distribution regardless of the types of mixtures. As shown in Figure 5, the single loop heat integration type 1 increases the efficiency about 15% and the dual loop type 5 increases the efficiency about 20% compared with the case of no heat integration (type 0). From this, it seems that there are no significant improvement of efficiency in dual loop integration compared to single loop integration.

The effect of reflux ratio on the thermodynamic efficiency in heat integrations was shown in Table 4 and in Figures 6-7 for the ternary mixture. As we

Table 4. Effect of variation of reflux ratio on the thermodynamic efficiency for system acetone-methanol-ethanol in dual column

Type of heat integration	Method	Variation of reflux ratio								
		R(2,2)*	R(2,3)	R(2,4)	R(3,2)	R(3,3)	R(3,4)	R(4,2)	R(4,3)	R(4,4)
0	RMS	.144	.133	.123	.124	.116	.109	.106	.100	.095
	MPA	.153	.140	.134	.132	.122	.110	.108	.102	.096
1	RMS	.211	.194	.179	.178	.166	.156	.152	.143	.135
	MPA	.223	.203	.192	.188	.174	.159	.156	.147	.138
2	RMS	.179	.164	.152	.152	.142	.133	.129	.122	.116
	MPA	.190	.173	.165	.161	.150	.136	.133	.126	.119
3	RMS	.185	.171	.157	.157	.147	.138	.134	.126	.120
	MPA	.196	.179	.170	.167	.155	.141	.138	.130	.123
4	RMS	.177	.163	.150	.151	.141	.132	.128	.121	.115
	MPA	.188	.171	.163	.159	.148	.134	.132	.124	.117
5	RMS	.242	.222	.205	.203	.189	.177	.172	.162	.154
	MPA	.255	.233	.218	.213	.198	.182	.178	.167	.158
6	RMS	.205	.189	.174	.174	.162	.152	.148	.139	.132
	MPA	.218	.195	.188	.184	.171	.156	.153	.144	.136
7	RMS	.212	.195	.180	.179	.167	.156	.152	.143	.136
	MPA	.225	.206	.194	.190	.176	.161	.158	.148	.140
8	RMS	.235	.216	.199	.198	.185	.173	.168	.159	.150
	MPA	.248	.227	.213	.208	.193	.177	.173	.163	.154
9	RMS	.233	.214	.197	.196	.183	.171	.166	.157	.148
	MPA	.247	.225	.212	.207	.192	.176	.172	.162	.153

* R(x,y): reflux ratio for first and second column

**Fig. 6.** Effect of reflux ratio on the average thermodynamic efficiency for various heat integrations for system acetone-ethanol-methanol.**Fig. 7.** Effect of heat integration and reflux ratio on the thermodynamic efficiency for dual column distillation of acetone-methanol-ethanol.

might expect, under given conditions, the thermodynamic efficiency decreases as the reflux ratios increases.

CONCLUSION

A systematic diagnostic method for energy conservation through simple external heat integration was developed by a thermodynamic analysis incorporated with the exergy concept. The distillation efficiency in an ordinary dual column can be improved up to 15 to 20% with practical single or dual heat integration through the reboilers and the condensers utilizing heat exchanger. In case of atmospheric distillation, the single heat integration through reboiler of the first column in multicolumn distillation seems the most practical candidate to improve thermodynamic efficiency in view point of additional installation cost of heat integrating devices. Also, as one might expect, in appropriate control of reflux ratio is important factor for the thermodynamic efficiency in heat integration.

The approach discussed here made it possible for the thermodynamic efficiency of any multicolumn distillation system with various types of heat integration to be analyzed on an unified quantitative exergy basis.

The algorithm proposed in this paper was programmed by FORTRAN 77 for IBM-PC-XT. The program listing can be obtained from the authors upon request.

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NOMENCLATURE

A	: heat transfer area in heat exchangers, m ²
B	: total exergy function, KJ
b	: molar exergy function, KJ/mol
C _p	: heat capacity, KJ/mol-K
F	: feed flow rate, mol/hr
LW	: lost energy, KJ/mol
m	: flow rate, mol/hr
P	: pressure, bars
Q	: total heat flux, KJ/hr
q	: molar heat flux, KJ/mol-hr
T	: absolute temperature, K
U	: overall heat transfer coefficients
V	: system volume, m ³
W	: various types of energies, KJ

Greek Letter

η	: thermodynamic efficiency
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Subscripts

c	: condenser or cold stream
irrev	: irreversibilities
hl	: heat leak
hx	: heat exchanger
lm	: logarithmic mean
min	: minimum separation
net	: net work requirement
o	: reference state
r	: reboiler or hot stream
sm	: stage mixing
sys	: system

REFERENCES

1. Choi, K.D. and Yoo, K.P.: "Calculation of Availability Functions for Multicomponent Vapor-Liquid Equilibrium Mixtures", *Kor. J. Chem. Eng.*, **5**(1), (1988).
2. Kim, H.S., Lee, K.S., Yoo, K.P. and Lee, W.H.: "Modularized Reduced-Order Model for Multicomponent Multistage Distillation Column", accepted No-E-80 to *J. Chem. Eng. Japan.*, June (1987).
3. Fair, J.R. and Bolles, W.L.: *Chem. Eng. Apr.*, **22**, 156 (1968).
4. Coward, I.: *Chem. Eng. Sci.*, **22**, 503 (1967).
5. Mix, T.W., Dweck, J.S., Weinberg, M. and Armstrong, R.C.: *Chem. Eng. Progr.*, **74**(4), 49 (1978).
6. Mah, R.S.H., Nicolas, J.J. and Wodnik, R.B.: *AIChE J.*, **23**(5), 651 (1977).
7. Fitzmorris, R.E. and Mah, R.S.H.: *AIChE J.*, **26**(2), 265 (1980).
8. Umeda, T.K., Niida, K. and Shiroko, K.: *AIChE J.*, **25**(3), 423 (1979).
9. Elaahl, A. and Luyben, W.L.: *Ind. Eng. Chem. Process Des. Dev.*, **22**, 80 (1983).
10. Stephenson, R.M. and Anderson, T.F.: *Chem. Eng. Progr.*, **76**(8), 68 (1980).
11. Yoo, K.P. and Choi, C.S.: *Kor. J. Chem. Ind. Tech.*, **3**, 3 (1985).
12. Yoo, K.P. and Choi, C.S.: *Kor. J. Chem. Ind. Tech.*, **4**, 3 (1985).
13. Buckhurst, J.R. and Harker, J.H.: *Process Plant Design.*, p 65, Heinemann Educational Books Ltd. London (1973).
14. Prausnitz, J., Anderson T., Grens. E., Eckert, C., Hsieh, R. and O'connell, J.: *Computer Calculations for Multicomponent Vapor-Liquid and Liquid-Liquid Equilibria*, Prentice-Hall Inc., (1980).
15. Naka, Y.: *Proceedings of The 1st Korea-Japan Symposium on Separation Technology* p256, October, Kyungju Korea (1987).