

Monoethanolamine as an Extractive Solvent for the *n*-Hexane + Benzene, Cyclohexane + Ethanol, and Acetone + Methanol Binary Systems

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The potential of monoethanolamine (MEA) as an extractive solvent for extractive distillation and liquid extraction was assessed. The systems studied were *n*-hexane + benzene, cyclohexane + ethanol, and acetone + methanol binary azeotropic systems with MEA as the solvent. Sub-atmospheric vapor–liquid equilibrium data were determined for all the systems with various amounts of MEA in the still feed. Ternary liquid–liquid equilibrium data were determined for the cyclohexane + ethanol + MEA system at 298.15 K and 1 atm. An equivolatility curve map was determined for the acetone + methanol + MEA system at 67.58 kPa. Results show that MEA cannot be used in extractive distillation for the *n*-hexane + benzene system. A combination of extractive distillation and liquid extraction with MEA as the solvent can be used to obtain pure cyclohexane and pure ethanol from a mixture. MEA works well as a solvent for the extractive distillation of acetone + methanol systems.

1. Introduction

Separation processes are by nature expensive to construct and operate. These costs are elevated when the systems being separated form azeotropes. Several methods exist for the separation of azeotropic compounds, with the use of an extractive solvent being the most popular.

In our previous work¹ preliminary laboratory studies were performed on the solvent monoethanolamine (MEA) to assess its potential as an extractive solvent. The focus of these studies was the determination of selectivity factors from infinite dilution data that can be used to assess a solvent's separating potential.² The selectivity factors indicated that MEA has great potential as an extractive solvent.

This paper assesses the ability of MEA as a solvent in extractive distillation for the *n*-hexane + benzene, cyclohexane + ethanol, and acetone + methanol binary systems. All these binary systems form azeotropes at the conditions at which they were studied. Experimentation involved the determination of vapor–liquid equilibrium (VLE) data for these systems with the solvent MEA. Liquid–liquid equilibrium (LLE) data were determined for the cyclohexane + ethanol + MEA ternary system.

2. Experimental Section

All VLE measurements were made at subatmospheric isobaric conditions. A dynamic recirculating glass still was used to determine the VLE data. The apparatus is a Yerazunis et al. type still³ with modifications by Raal.⁴ Temperature measurement was by a platinum resistance thermometer (Pt-100) submerged in a glass well that is in the packing in the equilibrium chamber. A calibrated Eurotherm temperature display was used for temperature data and is accurate to within 0.02 K. A Fischer pressure controller was used to maintain accurate vacuum pressure.

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This instrument is accurate to within 0.01 kPa. Mole fractions were measured with a calibrated gas chromatograph. These values are accurate to within 0.002 mole fraction.

LLE data were acquired by the stirred flask method as used previously by Letcher and Naicker.⁵ The water bath temperature was maintained constant by a Tronac temperature controller and measured with a Hewlett-Packard quartz thermometer, which is accurate to within 0.002 K. Mole fractions were measured as stated above and are accurate to within 0.002 mole fraction.

3. Source and Purity of the Materials

Table 1 lists chemicals, suppliers, and stated purity.

Purity was verified by GC analysis, and chemicals were not purified further.

4. Theory

VLE data were assessed on a solvent-free basis.^{6,7} In this work VLE curves are plotted for the normalized mole fractions of the binary systems for various concentrations of solvent in the feed to the still. Thus, the following applies:

$$x'_1 = \frac{x_1}{x_1 + x_2} \quad (1)$$

$$x'_2 = \frac{x_2}{x_1 + x_2} \quad (2)$$

$$x'_1 + x'_2 = 1 \quad (3)$$

$$x_1 + x_2 + x_{\text{solvent}} = 1 \quad (4)$$

where x is the component mole fraction in the liquid (y for vapor) phase. Subscripts 1 and 2 refer to the components of the binary system being studied, subscript "solvent"

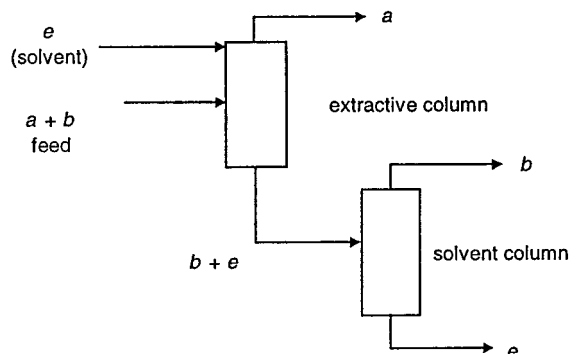


Figure 1. Separation sequence for mixture $a + b$ using heavy solvent e .

Table 1. Chemicals, Suppliers, and Stated Purity

chemical	supplier	purity (mol %)
monoethanolamine	ACROS	99%
<i>n</i> -hexane	SAARChem	99%
cyclohexane	ACROS	>99%
benzene	JANSSEN	99.5%
acetone	Romil	99.9%
methanol	Romil	99.8%
ethanol	Fluka	99.8%

refers to the solvent MEA, and superscript ' refers to the normalized values.

Relative volatility plots for the solvent-free basis systems are valuable tools demonstrating the effect of the solvent on the binary system under study and are defined as follows,

$$\alpha'_{12} = \left\{ \left(\frac{y'_1}{x'_1} \right) \right\} / \left\{ \left(\frac{y'_2}{x'_2} \right) \right\} \quad (5)$$

where α'_{12} is the relative volatility of the solvent-free basis binary system and y refers to the mole fraction in the vapor phase.

Equivolatility curves are useful for the comparison of solvents used in extractive distillation. The equivolatility curve, $\Gamma_{\alpha_0}^{ab}$, is a set of points for which the relative volatility of the two chemicals is constant:⁸

$$\Gamma_{\alpha_0}^{ab} = \{ \alpha'_{12} = \alpha_0 \} \quad (6)$$

and

$$\alpha'_{12} = \left\{ \left(\frac{y'_1}{x'_1} \right) \right\} / \left\{ \left(\frac{y'_2}{x'_2} \right) \right\} = \text{const.} \quad (7)$$

The relevant ternary compositions, x_1 and x_2 , for the equivolatility curve are plotted on a right-angled triangle as shown in Figure 2. An important equivolatility curve is the one for which $\alpha_0 = 1$ and is defined as the isovolatility curve. The position of this curve can be used to assess the minimum amount of solvent required to eliminate the azeotrope.

For the separation of azeotropic mixture a and b , using a heavy extractive distillation solvent e , the sequence shown in Figure 1 is used and Figure 2 is an illustrative equivolatility curve map for the system. Only theory pertaining to heavy solvents (i.e., solvents that are much less volatile than a and b) is considered here as MEA falls into this category.

Laroche et al.⁸ state that the criterion for the extraction of a as distillate from the extractive column is that the

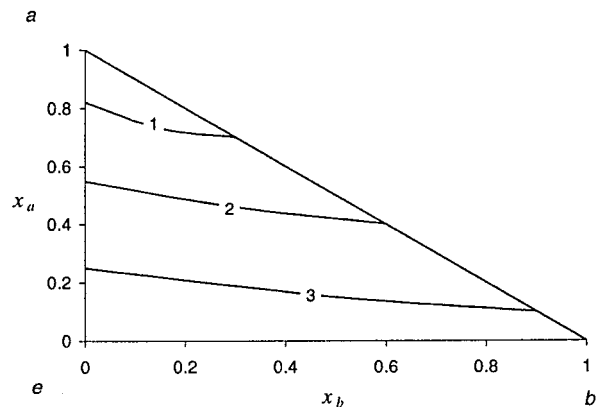


Figure 2. An illustrative example of an equivolatility curve map.

Table 2. Infinite Dilution Separation Factors at $T = 298.15$ K from Harris et al.¹

system	β_{12}^{∞}
<i>n</i> -hexane (1) + benzene (2)	31
cyclohexane (1) + ethanol (2)	148
acetone (1) + methanol (2)	7.7

isovolatility curve intersects the a - e edge as illustrated in Figure 2. If the isovolatility curve intersects the b - e edge, then b is extracted as the distillate from the extractive column.

Determination of the isovolatility curve is important as it defines the separation sequence. Furthermore, the separation sequence is important for the comparison of solvents. Laroche et al.⁸ state that, for an azeotropic mixture of a and b with several possible solvents, only solvents that produce the same separation sequence can be compared. Only separation sequences such as that described in Figure 1 are discussed in this paper.

The following criteria can be used for the comparison of solvents:

(1) the intersection of the isovolatility curve to the a - e (in this case) edge;

(2) the maximum binary relative volatility for a and b .

The intersection of the a - e edge by the isovolatility curve allows the determination of the value x_e , which is the mole fraction of the solvent at the intersection ($1 - x_{a(\text{at intersection})} = x_e$). Laroche et al.⁸ concluded that the best solvent for a particular azeotropic separation is the one that gives the lowest value of x_e and the highest binary relative volatility.

Ternary liquid-liquid equilibrium data are presented on an equilateral triangle.

5. Results

Table 2 represents the infinite dilution separation factors, β_{12}^{∞} , determined in our previous work¹ for the *n*-hexane + benzene, cyclohexane + ethanol, and acetone + methanol binary systems with MEA as the solvent.

The separation factor (for the separation of components 1 and 2) is defined by Tiegs et al.² as

$$\beta_{12}^{\infty} = \frac{\gamma_1^{\infty}}{\gamma_2^{\infty}} \quad (8)$$

where γ_1^{∞} is the activity coefficient at infinite dilution of solute 1 in monoethanolamine and γ_2^{∞} is the activity coefficient at infinite dilution of solute 2 in monoethanolamine.

Table 3. List of Measured Systems

binary system	% MEA in feed	pressure (kPa)
<i>n</i> -hexane + benzene	0	53.33
<i>n</i> -hexane + benzene	2	53.33
cyclohexane + ethanol	0	40
cyclohexane + ethanol	5	40
cyclohexane + ethanol	10	40
acetone + methanol	0	67.58
acetone + methanol	5	67.58
acetone + methanol	10	67.58
acetone + methanol	20	67.58

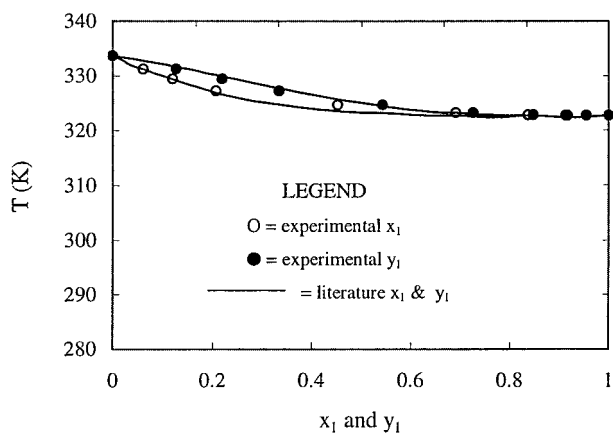
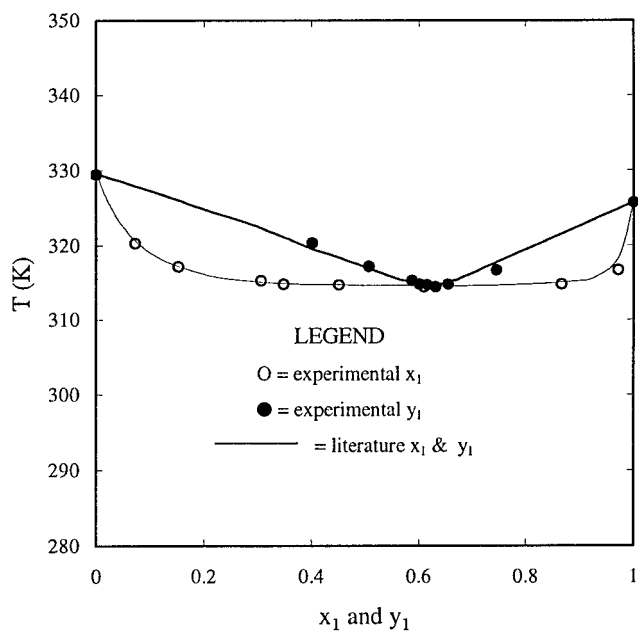
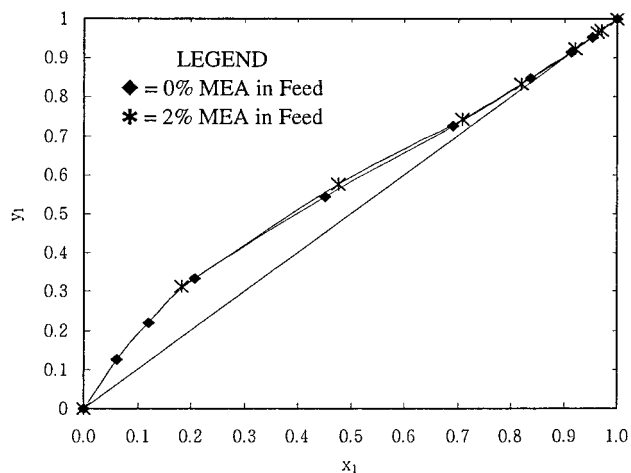
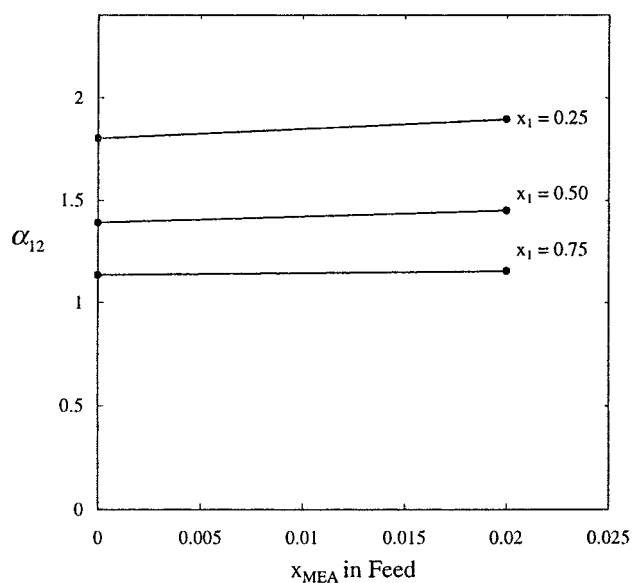
**Figure 3.** VLE T - x - y plot of *n*-hexane (1) + benzene (2) at 53.33 kPa—experimental compared to Gothard and Minea.⁹**Figure 4.** VLE T - x - y plot of cyclohexane (1) + ethanol (2) at 40 kPa—experimental compared to Morachevsky and Zharov.¹⁰

Table 3 summarizes the systems for which VLE were measured.

MEA was not completely miscible in the *n*-hexane + benzene and cyclohexane + ethanol systems, thus restricting measurements.

The Binary Systems. Literature data exist for the system *n*-hexane (1) + benzene (2) at 53.33 kPa⁹ and for cyclohexane (1) + ethanol at 40.00 kPa.¹⁰ Figures 3 and 4 compare our data to the literature data. No literature data exists for the system acetone (1) + methanol (2) at 67.58 kPa.

**Figure 5.** VLE x - y plot for *n*-hexane (1) + benzene (2) on a solvent-free basis at $P = 53.33$ kPa.**Figure 6.** Binary relative volatility for the system *n*-hexane (1) + benzene (2) versus % MEA in still feed (solvent-free basis) at $P = 53.33$ kPa.

Thermodynamic consistency tests were performed for all three binary systems and are discussed in detail in our other work.¹¹ All three binary systems display good thermodynamic consistency.¹¹

The *n*-Hexane (1) + Benzene (2) System. The results for the *n*-hexane + benzene system are represented as follows:

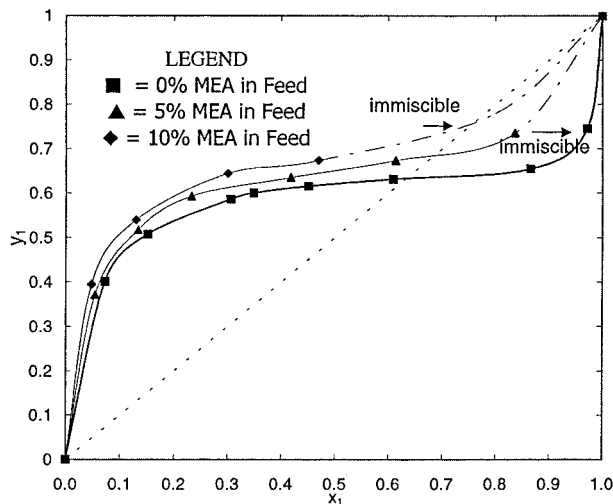
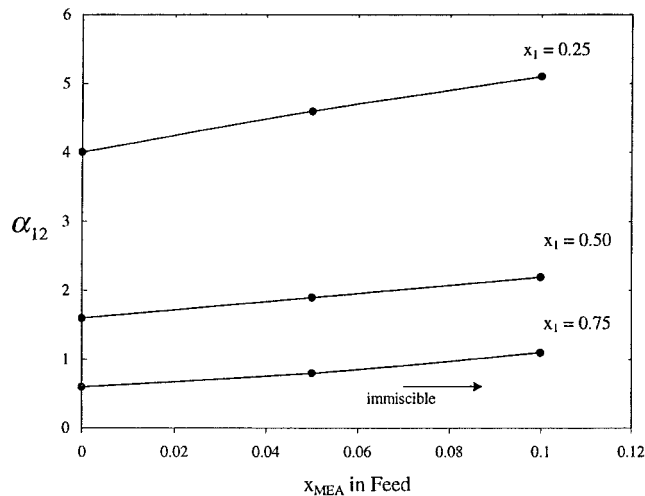
- (1) solvent-free basis x - y plots (Figure 5);
- (2) binary relative volatility versus percent MEA in the feed to the still (Figure 6);
- (3) solvent-free binary VLE data (Table 4).

The Cyclohexane (1) + Ethanol (2) System. The results for the cyclohexane + ethanol system are represented as follows:

- (1) solvent-free basis x - y plots (Figure 7);
- (2) binary relative volatility versus percent MEA in the feed to the still (Figure 8);
- (3) solvent-free binary VLE data (Table 5);
- (4) ternary LLE phase diagram (Figure 9);
- (5) ternary LLE data (Table 6).

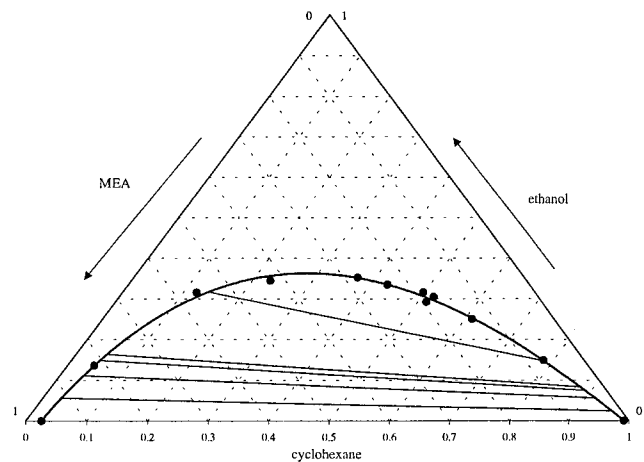
Table 4. Experimental Data for the System *n*-Hexane (1) + Benzene (2) at $P = 53.33$ kPa

x_{MEA} in feed	x_1	y_1
0.00	0.061	0.127
0.00	0.120	0.220
0.00	0.207	0.335
0.00	0.452	0.543
0.00	0.691	0.726
0.00	0.837	0.847
0.00	0.913	0.916
0.00	0.954	0.954
0.02	0.183	0.314
0.02	0.477	0.576
0.02	0.708	0.743
0.02	0.821	0.834
0.02	0.922	0.924
0.02	0.963	0.963
0.02	0.970	0.970

**Figure 7.** VLE x - y plot for cyclohexane (1) + ethanol (2) on a solvent-free basis at $P = 40$ kPa.**Figure 8.** Binary relative volatility for the system cyclohexane (1) + ethanol (2) versus % MEA in still feed (solvent-free basis) at $P = 40.00$ kPa.

The Acetone (1) + Methanol (2) System. The results for the acetone + methanol system are represented as follows:

- (1) solvent-free basis x - y plots (Figure 10);
- (2) binary relative volatility versus percent MEA in the feed to the still (Figure 11);
- (3) solvent-free binary VLE data (Table 7);
- (4) equivalatility curve map (Figure 12);
- (5) equivalatility curve map data (Table 8).

**Figure 9.** Ternary liquid-liquid equilibria for cyclohexane (1) + ethanol (2) + MEA (solvent) system at $T = 298.15$ K.**Table 5. Experimental Data for the System Cyclohexane (1) + Ethanol (2) at $P = 40.00$ kPa**

x_{MEA} in feed	x_1	y_1
0.00	0.072	0.401
0.00	0.152	0.507
0.00	0.305	0.586
0.00	0.348	0.600
0.00	0.450	0.615
0.00	0.608	0.631
0.00	0.866	0.655
0.00	0.972	0.746
0.05	0.054	0.371
0.05	0.134	0.517
0.05	0.233	0.593
0.05	0.418	0.635
0.05	0.613	0.673
0.05	0.837	0.735
0.10	0.048	0.395
0.10	0.130	0.539
0.10	0.300	0.644
0.10	0.470	0.673

6. Discussion

***n*-Hexane + Benzene.** MEA is only slightly miscible in both *n*-hexane and benzene. Because of these solubility restrictions throughout the whole composition range, only 2% MEA was added to the feed of the still. The x - y plot (Figure 5) and relative volatility plots (Figure 6) demonstrate an improvement in the separability of the two chemicals. However, because of solubility restrictions, extractive distillation with MEA as the solvent for this system is not feasible.

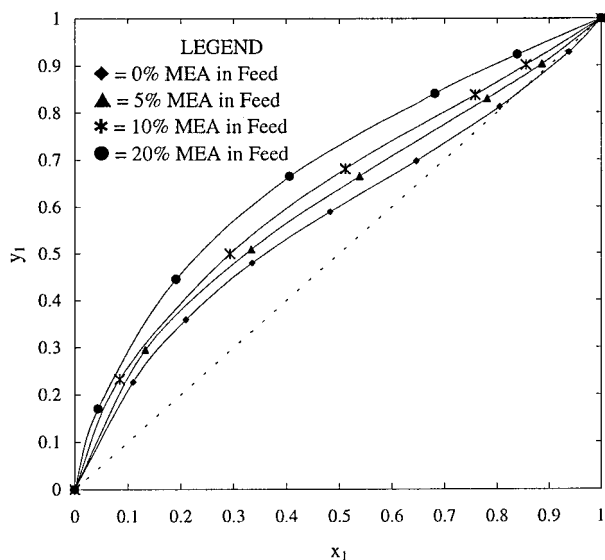
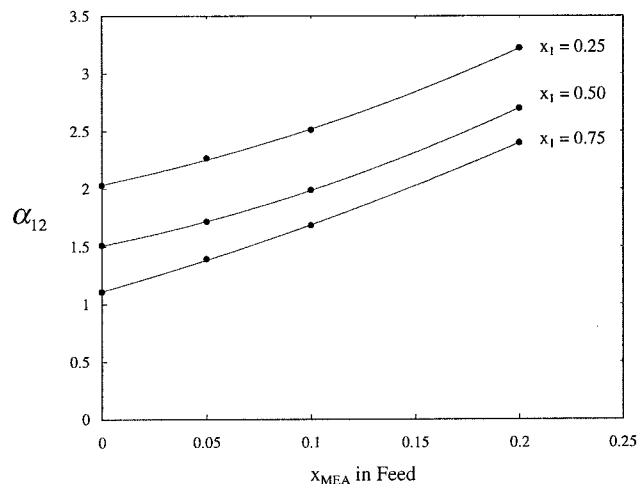
Cyclohexane + Ethanol. The x - y and relative volatility plots (Figures 7 and 8) demonstrate an increase in relative volatility for an increase in MEA in the still feed. Solubility restrictions, however, prevent the addition of sufficient MEA to eliminate the binary azeotrope. MEA is completely miscible in ethanol but only slightly miscible in cyclohexane. Thus, in the cyclohexane-rich regions, a two-phase liquid mixture is formed. This limits the addition of MEA as two liquid phases are not desirable for any liquid flow situations.

The ternary phase diagram in Figure 9 demonstrates that MEA is reasonably selective and would allow relatively good separation of cyclohexane and ethanol. Figure 13 shows a possible process for the separation of cyclohexane and ethanol using MEA as a solvent in extractive distillation and liquid extraction.

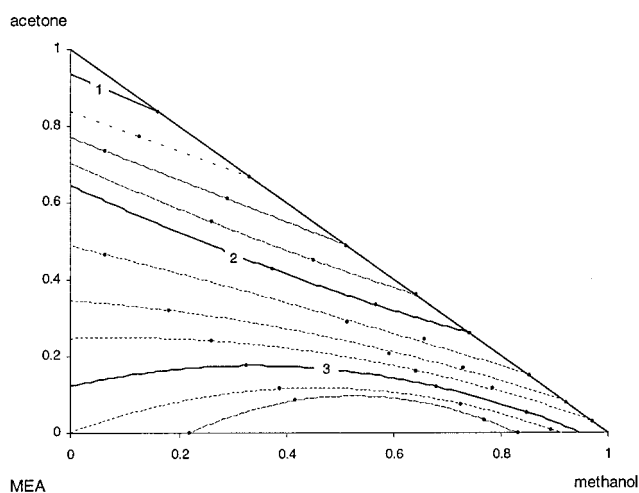
Acetone-Methanol. MEA was added to the feed in the still in 5, 10, and 20% amounts. Results from Figures 10 and 11 illustrate that the azeotrope is eliminated with only

Table 6. LLE Ternary Phase Data for the Cyclohexane (1) + Ethanol (2) + MEA (Solvent) System at $T = 298.15$ K (Two Liquid Phases: α and β)

binodal curve			tie lines					
$x_{\text{cyclohexane}}$	x_{ethanol}	x_{MEA}	α			β		
$x_{\text{cyclohexane}}$	x_{ethanol}	x_{MEA}	$x_{\text{cyclohexane}}$	x_{ethanol}	x_{MEA}	$x_{\text{cyclohexane}}$	x_{ethanol}	x_{MEA}
0.026	0.000	0.974	0.784	0.147	0.069	0.141	0.317	0.542
0.044	0.137	0.819	0.851	0.102	0.047	0.048	0.155	0.797
0.122	0.315	0.563	0.877	0.083	0.039	0.051	0.164	0.785
0.230	0.345	0.425	0.890	0.075	0.036	0.047	0.149	0.804
0.371	0.352	0.277	0.916	0.056	0.028	0.039	0.112	0.849
0.429	0.334	0.237	0.959	0.025	0.017	0.030	0.057	0.913
0.498	0.315	0.187						
0.515	0.292	0.193						
0.521	0.304	0.174						
0.612	0.250	0.138						
0.783	0.149	0.068						
0.991	0.000	0.009						

**Figure 10.** VLE x - y plot for acetone (1) + methanol (2) on a solvent-free basis at $P = 67.58$ kPa.**Figure 11.** Binary relative volatility for the system acetone (1) + methanol (2) versus % MEA in still feed (solvent-free basis) at $P = 67.58$ kPa.

5% MEA in the feed. As more MEA is added to the still, feed separability improves steadily. MEA has a higher affinity for methanol (because of the very polar OH group) and thus decreases its volatility in the mixture, allowing the separation of the two chemicals. Figure 14 illustrates the possible separation process for the separation of acetone and methanol using MEA.

**Figure 12.** Equivolatility curve for acetone (1) + methanol (2) + MEA (solvent) at $P = 67.58$ kPa.**Table 7. Experimental Data for the System Acetone (1) - Methanol (2) at $P = 67.58$ kPa**

$x_{\text{MEA in feed}}$	x_1	y_1
0.00	0.000	0.000
0.00	0.110	0.227
0.00	0.209	0.359
0.00	0.335	0.480
0.00	0.482	0.589
0.00	0.646	0.698
0.00	0.805	0.812
0.00	0.937	0.929
0.00	1.000	1.000
0.05	0.133	0.295
0.05	0.333	0.509
0.05	0.538	0.665
0.05	0.781	0.829
0.05	0.886	0.904
0.10	0.084	0.233
0.10	0.293	0.499
0.10	0.511	0.681
0.10	0.758	0.838
0.10	0.856	0.902
0.20	0.043	0.923
0.20	0.191	0.665
0.20	0.406	0.445
0.20	0.682	0.170
0.20	0.839	0.840

Only two distillation columns are required to produce relatively pure acetone, methanol, and MEA (which can be recycled). The solvent column would be much smaller than the extractive column because of the large difference in boiling points of methanol and MEA. Methanol is much

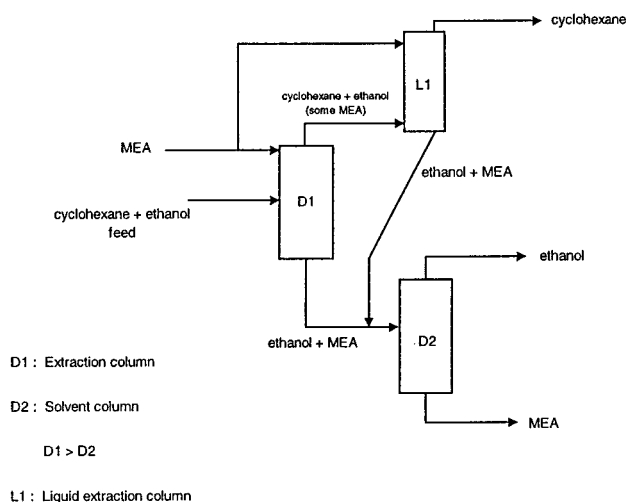


Figure 13. Completed separation process for cyclohexane + ethanol mixture using MEA as the solvent.

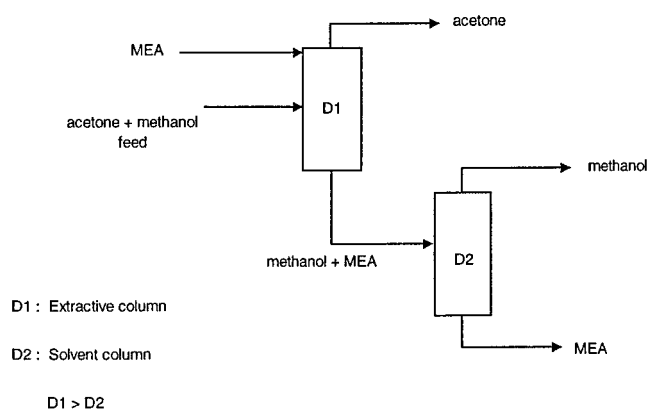


Figure 14. Separation process for acetone + methanol mixture using MEA as the solvent.

Table 8. Equivolatility Curve for Acetone (1) + Methanol (2) + MEA (Solvent) at $P = 67.58$ kPa

α'_{12}	x_1	x_2	x_{solvent}	α'_{12}	x_1	x_2	x_{solvent}
1	0.84	0.16	0.00	2.5	0.17	0.73	0.10
1	0.91	0.04	0.05	2.5	0.241	0.59	0.20
1.25	0.67	0.33	0.00	2.5	0.32	0.18	0.50
1.25	0.77	0.13	0.10	2.75	0.03	0.97	0.00
1.5	0.49	0.51	0.00	2.75	0.12	0.78	0.10
1.5	0.61	0.29	0.10	2.75	0.16	0.64	0.20
1.5	0.74	0.06	0.20	2.75	0.24	0.26	0.50
1.75	0.36	0.64	0.00	3	0.05	0.85	0.10
1.75	0.45	0.45	0.10	3	0.12	0.68	0.20
1.75	0.55	0.26	0.19	3	0.18	0.33	0.50
2	0.26	0.74	0.00	3.25	0.01	0.89	0.10
2	0.33	0.57	0.10	3.25	0.08	0.72	0.20
2	0.43	0.37	0.20	3.25	0.12	0.39	0.50
2.25	0.15	0.85	0.00	3.5	0.00	0.22	0.78
2.25	0.24	0.66	0.10	3.5	0.00	0.83	0.17
2.25	0.29	0.51	0.20	3.5	0.03	0.77	0.20
2.25	0.47	0.06	0.47	3.5	0.09	0.42	0.50
2.5	0.08	0.92	0.00				

more volatile than MEA and would flash out of the mixture. It is important to note that the MEA feed (to the first distillation column in the above diagram) is introduced at several trays, all of them above the feed tray, to ensure a sufficient amount of MEA in the mixtures on each tray. MEA is fed to the higher trays as it is much less volatile than the other chemicals and descends to the bottom of the column.

Laroche et al.⁸ compare several solvents for the separation of acetone and methanol (all of which yield acetone as

Table 9. x_e Values and Maximum Binary Relative Volatility Values for Several Solvents Used for the Separation of Acetone (1) and Methanol (2)

solvent	x_e	α'_{12} maximum	pressure
MEA	0.07	3.5	67.58 kPa
water	0.10	3.0	101.325 kPa
ethanol	0.20	2.1	101.325 kPa
2-propanol	0.29	2.5	101.325 kPa

the distillate from the extraction column as does MEA). Table 9 compares the x_e and maximum binary relative volatility values for the various solvents used for acetone (1) + methanol (2) separation.

The x_e values are indicative of the amount of solvent used in the separation. The lower the x_e value, the lower the amount of solvent necessary. From Table 9 it is obvious that MEA requires the least amount of solvent to produce the required separation. The maximum binary relative volatility is related to the minimum reflux ratio required to produce the desired separation. From Table 9 it can be seen that MEA has the highest maximum binary relative volatility. This indicates that it requires the lowest minimum reflux ratio to produce the desired separation compared to the other solvents.

It is, however, important to note that all the other solvent results are simulated values for atmospheric pressure while the results for MEA are based on experimental data at 67.58 kPa. This difference in pressure does affect the relative volatility. To accurately conclude which solvent is best would require experimental data for all the solvents at the same pressure. However, the results shown here indicate that MEA would probably be the best solvent to use in the extractive distillation of acetone + methanol mixtures as mixtures of methanol + water or ethanol or 2-propanol are themselves not easy to separate.

7. Conclusions

MEA was evaluated as an extractive distillation solvent for the following three azeotropic binary mixtures:

- (1) *n*-hexane + benzene;
- (2) cyclohexane + ethanol;
- (3) acetone + methanol.

MEA cannot be used as a solvent for the extractive distillation of *n*-hexane + benzene because of immiscibility of the mixture.

For the cyclohexane + ethanol system, MEA improves vapor–liquid separability but is immiscible in cyclohexane-rich regions. MEA has reasonable selectivity as a liquid extractive solvent in this system. A possible process involving extractive distillation and liquid extraction with MEA as the solvent (Figure 13) produces pure cyclohexane and pure ethanol.

MEA works extremely well as a solvent for the extractive distillation of acetone + methanol. Figure 14 shows the separation process using MEA as the extractive distillation solvent to produce pure acetone and pure methanol. Comparison of equivolatility curve maps shows that MEA could be the best solvent for the separation of acetone + methanol mixtures.

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