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Prediction of Reflux Ratio for Multicomponent Batch Distillation

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The conventional operation of multicomponent batch distillation columns is inherently dynamic and involves charging the reboiler with the initial feed mixture. The fractionation is carried out until a desired amount of top product has been distilled off. Based on the amount of this desired top product and time-dependent temperatures of the condenser and reboiler, an equation is proposed for the estimation of a time-varying reflux ratio in multicomponent batch distillation processes, which, in comparison to measured experimental data, produces good prediction with reasonable accuracy.

INTRODUCTION

The conventional operation of a batch distillation involves charging the feed to be separated into the reboiler and heating up the reboiler until a desired portion of initial feed charge has been distilled off at the top.

The process is inherently dynamic and model equations to describe the system are time-dependent differential and algebraic equations. This makes the analysis of the system complicated, therefore, its steady-state operation has been recently considered [1], which involves steady-state distillation computations with only algebraic equations. Wu et al. [2] used selected effective concentrations in a graphical procedure as a criterion to determine the required reflux ratio for the simulation of the operation steps for various reflux ratios. Reflux ratios are determined, sequentially, from one operation step to the next. Bernot et al. [3] used an iterative calculation for the constant reflux policy and a variable reflux policy for the design and operating targets of batch distillation. At each value of the warped time, they used the procedure to find the reflux ratio that gives the specified product purity and the desired fractional recovery. Logsdon and Biegler [4] predicted unusual and advantageous optimal reflux policies that exploit hold-ups in batch columns through optimization calculations. As analysis of a dynamic batch operation is difficult, in the present work, shortcuts and approximate algebraic solutions are shown to be useful and experiments are conducted to develop a simple relationship for the prediction of a time-varying reflux ratio.

THE ESTIMATED REFLUX MODEL

A material balance on a condenser tray would be as follows [5]:

$$m_C \frac{dx_{Di}}{dt} = V_2 (y_{2i} - x_{Di}), \tag{1}$$

and the energy balance:

$$V_2 (h_2^V - h_1^L) - Q_C = 0. \tag{2}$$

Therefore, by substituting for V_2, one has:

$$m_C \frac{dx_{Di}}{dt} = \frac{Q_C}{h_2^V - h_1^L} (y_{2i} - x_{Di}), \tag{3}$$

or:

$$\begin{aligned} m_C &= \frac{(x_{Di,t_2} - x_{Di,t_1})}{\Delta t} \\ &= \frac{Q_C}{m_2 C_p^V T_2 - m_1 C_p^L T_1} (y_{2i} - x_{Di}), \end{aligned} \tag{4}$$

or, by using average composition for x_{Di} and simplifying, one has:

$$m_C x_{Di} = \frac{Q_C \Delta t}{m_2 C_{pV} T_2 - m_1 C_{p_1}^L T_1} (y_{2i} - x_{Di}). \tag{5}$$

By conducting an energy balance around the column, the following is obtained:

$$Q_R - Q_C - m_1 C_{p_1}^L T_1 = 0. \tag{6}$$

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Hence;

$$m_C x_{Di} = \frac{[Q_R - m_1 C^L_{p_1} T_1]\Delta t}{m_2 C_{p_V} T_2 - m_1 C^L_{p_1} T_1}(y_{2i} - x_{Di}). \qquad (7)$$

By performing an energy balance around the reboiler, one has:

$$Q_R = \Delta m_R C^L_p T_R + V_N C^V_p T_R, \qquad (8)$$

where:

$$\Delta m_R = F - B = B(t_1) - B(t_2). \qquad (9)$$

Therefore:

$$m_C x_{Di} =$$

$$\frac{[\Delta m_R C^L_p T_R + V_N C^V_p T_R - m_1 C_{p_{1L}} T_1]\Delta t}{m_2 C_{p_V} T_2 - m_1 C_{p_{1L}} T_1}(y_{2i} - x_{Di}). \qquad (10)$$

One also has:

$L_1 = rV_2$, $m_D = V_2(1-r)$ and $K_i L_1 x_{Di} = rV_2 y_{2i}$,

which are valid for a very small condenser hold-up. Now, $K_i m_D x_{Di} = V_2 y_{2i}(1-r)$ and $y_{2i} = \frac{K_i m_D x_{Di}}{V_2(1-r)}$. Therefore:

$$m_C x_{Di} = \frac{[\Delta m_R C_{p_L} T_R + V_N C_{p_V} T_R - m_1 C_{p_L} T_1]\Delta t}{m_2 C_p V T_2 - m_1 C_p L T_1} \times \left[\frac{K_i m_D x_{Di}}{V_2(1-r)} - x_{Di}\right]. \qquad (11)$$

On re-arranging,

$$1 - r =$$

$$\frac{m_D[\Delta m_R C_{pi} L T_R + V_N C_{p_V} T_R - m_1 C_p L T_1]\Delta t K_i}{V_2\left[m_C + \frac{[\Delta m_R C_p L T_R + V_N C_{p_V} T_R - m_1 C_p L T_1]\Delta t}{m_2 C_p V T_2 - m_1 C_p L T_1}\right](m_2 C_p V T_2 - m_1 C_p L T_1)}. \qquad (12)$$

and assuming $\Delta m_R \approx V_2$, since $\Delta m_R = B(t_1) - B(t_2) \approx V_2$, one has:

$$1 - r =$$

$$\frac{m_D[\Delta m_R C_p L T_R + V_N C_{p_V} T_R - m_1 C_p L T_1]\Delta t K_i}{\Delta m_R\left[m_C + \frac{[\Delta m_R C_p L T_R + V_N C_p V T_R - m_1 C_p L T_1]\Delta t}{m_2 C_{p_V} T_2 - m_1 C_p L T_1}\right](m_2 C_{p_V} T_2 - m_1 C_p L T_1)}. \qquad (13)$$

which leads to:

$$1 - r =$$

$$m_D T_R\left[\Delta m_R C_p L + V_N C_{pv} - m_1 C_p L\frac{T_1}{T_R}\right]\Delta t \frac{p_i^{\mathrm{sat}}}{p} \Big/$$

$$\Delta m_R m_C m_2 C_{p_V} T_2 - \Delta m_R m_C m_1 C_p L T_1$$

$$+ \Delta m_R T_R\left[\Delta m_R C_p L + V_N C_{pv} - m_1 C_p L\frac{T_1}{T_R}\right]\Delta t, \qquad (14)$$

m_C is time-independent (see Equation 1) and since $p = 1$ atm and $p_i^{\mathrm{sat}} = f(T_c)$, one has:

$$1 - r =$$

$$\frac{m_D T_R}{f(T_c)\left(\frac{\Delta m_R m_C m_2 C_{p_V} T_2 - \Delta m_R m_C m_1 C_{p_L} T_1}{(\Delta m_R C_{p_L} + V_N C_{p_V} - m_1 C_{p_L}\frac{T_1}{T_R})} + \Delta m_R T_R\right)} \qquad (15)$$

This equation is rather complicated and numerous experimentally measured data plus considerable tabulated physical properties, as well as phase calculation, are needed. However, a simpler equation can be proposed, based on the form of a equation in which fewer experimental data are used and the end-program to computer is quite simple to use for automatic control of batch distillation columns (Equation 15):

$$r = 1 - \frac{M_D T_{R\Delta}}{\Delta m_D(T_R - T_C) + \Delta m_R T_R}. \qquad (16)$$

In this work, this new equation has been tested experimentally and the use of $\Delta m_D T_C$ is justified by the evaluation of experimental data.

In Equation 16, Δm_R = initial feed charge-bottom residue and Δm_D is the difference between current and previously accumulated m_D. m_D is the mass of accumulated distillate, with respect to time, all in g or kg, T_R is the temperature of the reboiler and T_C is the temperature of the condenser in degrees centigrade.

Where there is a change in the operation state of the column [6], the reflux ratio will change and Equation 16 will be useful in the calculation of a new reflux ratio.

In supercritical fluid extraction in a semi-batch mode [7] the reflux ratio was calculated by measurement of the extraction rate at a certain temperature at the top and bottom of the column. Equation 16 can be used for elimination of the extraction rate and, instead, measurement of the quantity of bottom residue would be sufficient.

For short cut design procedures for batch distillation [8], optimum reflux ratio can be calculated using Equation 16. A simulation model for short-cut design manipulation of the reflux ratio can also be demonstrated using Equation 16, which is proposed in this study.

Tray hydraulics have an impact on optimal reflux policies [9] and the equation proposed in this study can be used to predict such impacts.

EXPERIMENTAL WORK

Apparatus

The equipment for separation of various mixtures of methanol-water was a twenty five stage glass column of about 9 cm diameter and 140 cm height, with the condenser and subcooler at the top. The reboiler was 5000 mL and the electrical heater was 5308.34 j/min.

The reboiler and condenser of the apparatus were equipped with thermometers with temperature measurements displayed on the control panel. The reboiler was heated electrically and a button was available on the control panel for various percentage power inputs.

There are two temperatures which can be pre-specified, namely the reboiler and column temperature cut-offs. For safety purposes, when the respective temperatures reach these pre-specified values, the power input to the heater is automatically cut off. For methanol-water separations, these temperatures were set at 110°C and 129°C, respectively, based on the boiling point of the heaviest component.

There is one side-arm installed in the reboiler for charging feed and withdrawal of bottom products. During experiments, this side-arm was closed with a rubber stopper. When opened for withdrawal of bottom products, the column hold-up flows back into the reboiler (the column hold-up is approximated as 5% of the reboiler residue after the separation). For the authors experiments, the column hold-up was approximately 20 mL, where the bottom product residue for one of the experiments was 175 mL.

Experiments

Several experiments were carried out with various feed compositions of methanol-water. One experiment was conducted with 150 mL methanol and 150 mL water. The measured mixture's specific gravity was $\rho_M = 929$ kg/m^3 and the results obtained are shown in Table 1.

The bottom residue was 127 mL, with a specific gravity of $\rho_B = 959$ kg/m^3 and a distillate specific gravity of $\rho_D = 855$ kg/m^3.

Table 1. Results of experiment using 150 mL methanol +150 mL water.

Time (min)	Reboiler Temp. (°C)	Condenser Temp. (°C)	Distillate (mL)
0	27	23	0
3	56	24	0
5	73	24	0
6	80 starts boiling	24	0
10	84	75	2
11	85	77	10
12	86	78	20
13	86	79	30
14	87	80	40
16	88	80	55
18	90	80	70
20	92	81	87
21	93	82	96
23	94	82	105
25	97	85	123

Another experiment was carried out with 100 mL water and 150 mL methanol with a feed specific gravity of 911 kg/m^3. The specific gravities are average over the entire experiment time and the results are given in Table 2. The final bottom residue was 94 mL, with density of $\rho_B = 982$ kg/m^3 and the specific gravity of the distillate, after the end of the experiment (average), was $\rho_D = 853$ kg/m^3.

Another experiment was carried out with 100 mL acetone and 100 mL water, with a feed specific gravity of 940 kg/m^3, the results for which are given in Table 3.

The final bottom residue was 90 mL with $\rho_B = 996$ kg/m^3. The specific gravity of the distillate (average) was $\rho_D = 811$ kg/m^3.

Several experiments were conducted with mul-

Table 2. Results of experiment using 150 mL methanol +100 mL water.

Time (min)	Reboiler Temp. (°C)	Condenser Temp. (°C)	Distillate (mL)
0	42	32	0
4	75 starts boiling	56	6
5	78	70	6
7	79	78	20
8	80	80	35
10	81	81	62
11	81	81	70
13	82	82	90
15	84	82	105
17	88	82	120
19	92	82	135
20	94	82	140

Table 3. Results of experiment using 100 mL acetone +100 mL water.

Time (min)	Reboiler Temp. (°C)	Condenser Temp. (°C)	Distillate (mL)
0	35	21	0
3	59 starts boiling	21	0
5	66	21	0
7	69	21	0
9	74	22	0
10	79	58	10
11	82	58	20
12	85	58	30
13	90	58	40
14	94	58	48
15	98	58	55
16	100	58	67
17	101	58	77
19	103	57	84

Table 4. Results of experiment using 110 mL acetone +110 mL methanol +80 mL water.

Time (min)	**Reboiler Temp. (°C)**	**Condenser Temp. (°C)**	**Distillate (mL)**
0	41	29	0
4	62 starts boiling	49	18
5	69	56	18
7	70	61	46
8	71	64	70
9	72	66	83
10	73	66	120
11	75	65	97
13	119	60	112
14	128	62	135
15	138	63	

ticomponent systems. The results of one of these experiments, which was carried out with 110 mL acetone, 110 mL methanol, 80 mL water and $\rho_F = 891$ kg/m^3, are given in Table 4.

The final bottom residue was 161 mL, with a specific density of $\rho_B = 942$ kg/m^3. The average specific density of the distillate was $\rho_D = 812$ kg/m^3.

TEST OF THE SIMPLIFIED EQUATION 16

Equation 16 was applied to the data of the first experiment and calculated values of m_D were compared to experimental values of m_D. The calculated values of r from Equation 16 were put into the following equation:

$$m_D = V_2(1 - r), \tag{17}$$

where V_2 is approximated by conducting a material balance around the column by assuming that the column hold-up is almost constant at the initial and final times. Then, from Figure 1, one obtains:

$$V_N + L_1 = V_2 + L_{N-1}. \tag{18}$$

Using the equimolar overflow assumption, it is concluded that $V_N = V_2$.

Let the amount of bottom residue be $B(0)$ at the initial time and $B(t)$ at time t. By doing a mass balance around the reboiler, one has:

$$B(t) - B(0) = L_{N-1} - V_N. \tag{19}$$

At the initial time, $L_{N-1} = 0$, so:

$$V_N = F - B(t) \quad \text{and} \quad V_2 = F - B(t). \tag{20}$$

Considering a mass balance around the column at any time, one has:

$$F = B(0) + H + m_D, \tag{21}$$

$$B = F - H - m_D, \tag{22}$$

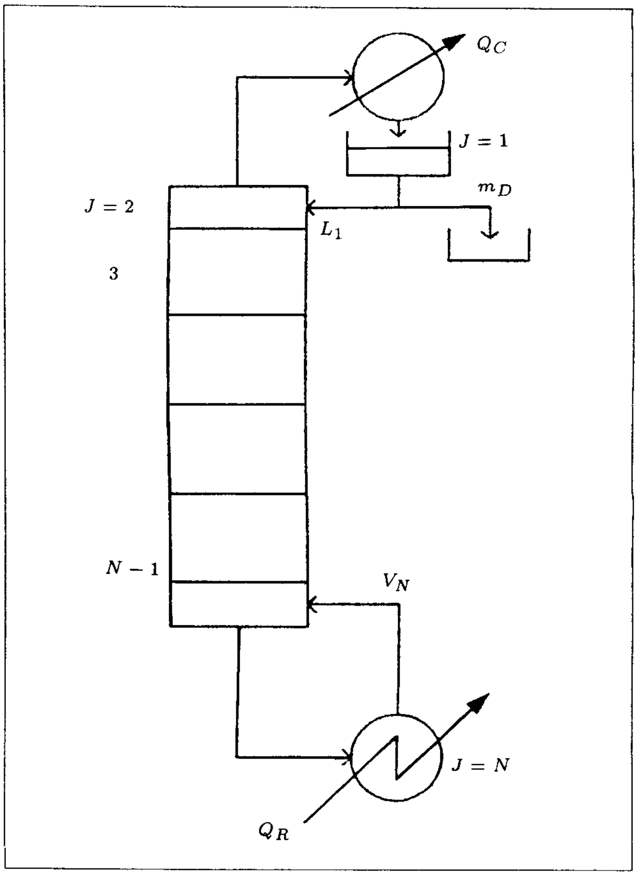

Figure 1. Equipment variables.

where B is the reboiler residue at time t, H is the column hold-up, which was assumed to be constant for the entire experiment time and m_D is the accumulated distillate amount at time t.

Since $\Delta m_R = F - B(t)$ (the amount evaporated from the reboiler up to time t), from Equations 20 and 22, one obtains:

$$\Delta m_R = V_2 = H_C + m_D = F - B, \tag{23}$$

which is valid for equimolar overflow.

The calculated and experimental m_D's are shown in Tables 5 to 8.

DISCUSSION

All the predicted values of m_D are slightly lower than measured. The experimental errors are not judged to be the explanation for this difference, so, the inadequacy lies with Equation 16. Further work on this equation is needed to try to reduce the discrepancy.

Equation 16 can be used for the calculation of a reflux ratio profile or for the calculation of a reboiler temperature profile for a fixed value of reflux ratio and a minimum operation time requirement. A reboiler temperature profile is a useful tool for smooth, computerized and automatic control of distillation

Table 5. Comparison of experimental data using 150 mL methanol +150 mL water.

Time (min)	Predicted Reflux Ratio	Predicted m_D (g)	Experimental m_D (g)	% Deviation
0				
3				
5				
6				
10	0.966	1.69	1.71	-0.36
11	0.852	8.45	8.55	-1.16
12	0.743	16.89	17.1	-1.19
13	0.658	25.41	25.65	-0.92
14	0.591	33.91	34.2	-0.82
16	0.514	46.45	47.02	-1.2
18	0.456	59.07	59.85	-1.29
20	0.404	73.34	74.38	-1.39
21	0.377	81.51	82.08	-0.69
23	0.356	89.13	89.77	-0.70
25	0.325	103.83	105.1	

Table 6. Comparison of experimental data using 150 mL methanol +100 mL water.

Time (min)	Predicted Reflux Ratio	Predicted m_D (g)	Experimental m_D (g)	% Deviation
4				
5	0.765	4.96	5.11	-2.93
7	0.487	16.98	17.06	-0.45
8	0.350	29.85	29.85	0.0
10	0.233	52.88	52.88	0.0
11	0.212	59.71	59.71	0.0
13	0.173	76.77	76.77	0.0
15	0.155	89.30	89.56	-0.28
17	0.142	101.61	102.36	-0.73
19	0.132	113.94	115.15	-1.04
20	0.123	118.94	119.42	-0.4

Table 7. Comparison of experimental data using 100 mL acetone +100 mL water.

Time (min)	Predicted Reflux Ratio	Predicted m_D (g)	Experimental m_D (g)	% Deviation
10	0.825	7.73	8.11	-4.65
11	0.703	15.51	16.22	-4.34
12	0.614	23.33	24.33	-4.09
13	0.546	31.12	32.44	-4.04
14	0.498	37.67	38.92	-3.20
15	0.463	43.35	44.6	-2.79
16	0.425	51.97	54.33	-4.32
17	0.388	60.32	62.44	-3.38
19	0.362	66.49	68.12	-2.37

Table 8. Comparison of experimental data using 110 mL acetone +110 mL methanol + 80 mL water.

Time (min)	Predicted Reflux Ratio	Predicted m_D (g)	Experimental m_D (g)	% Deviation
4	0.750	6.14	6.49	-5.25
5	0.573	13.95	14.61	-4.47
7	0.360	35.46	37.34	-5.01
8	0.260	55.46	56.89	-2.50
9	0.22	66.69	67.38	-1.01
10	0.185	87.60	89.32	-1.92
11	0.165		97.44	-0.92

columns. Equation 16 can be used to calculate reboiler temperatures for given values of m_D and reflux ratios. These values of the reboiler temperatures calculated from Equation 16 can be used to determine the amount of steam required to reach such reboiler temperatures. Therefore, Equation 16 is very useful in computerized automatic control of the steam rate to the reboiler, for given values of m_D. In other words, for a specific distillate product, the steam required can be calculated by use of Equation 16. It should be emphasised that for almost pure component recovery at the top, T_C is constant and, if the reflux ratio is fixed, $m_D(t)$ can be calculated by mass balance and, then, Equation 16 can be used to calculate T_R (reboiler temperature) for different values of $m_D(t)$. The T_R profile can be used by means of computerized automatic control to manipulate the steam rate to the reboiler, in order to follow up the predicted temperature profile by Equation 16 for minimum operation time.

CONCLUSION

Equation 16 offers an easy means of predicting the reflux ratio for the simulation and design of batch distillation columns for a proposed separation task. The equation is very simple to apply and the deviations are within engineering accuracy.

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NOMENCLATURE

B	final bottom residue, kg
C_p	heat capacity, kJ/kg°C
F	initial feed charge, kg
h_1^L	enthalpy of liquid phase at stage 1 (condenser), kJ
h_2^V	enthalpy of vapor phase at stage 2, kJ
m_1	total mass in Δt, kg
m_2	total mass in Δt, kg
m_C	mass flow from the condenser in time interval Δt, kg
m_D	accumulated mass of distillate up to time t, kg
Q_C	condenser heat duty, kJ
Q_R	heat duty of reboiler, kJ
r	reflux ratio
T_1	temperature at stage 1 (condenser), °C
H	column hold up
T_c	condenser temperature, °C
T_2	temperature at stage 2, °C
T_R	temperature of the reboiler, °C
V_2	amount of vapor flow from stage 2 at time interval of Δt, kg
V_N	amount of vapor flow from stage N (reboiler) at time interval of Δt, kg
x_{Di}	mole fraction of component in the distillate
x_{Di,t_2}	compositon of component in distillate at time t_2
y_{2i}	mole fraction of component i in stage 2
Δt	$t_2 - t_1$ entire experiment time, min.

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