# Periodic Cycling of Distillation Columns Using a New Tray Design

Computer simulations of periodically cycled columns predicted improvements in column performance of greater than 200% for systems with nonlinear equilibrium curves. Experiments distilling methanol-water mixtures have verified those predictions for periodic cycling with a single plate. The extension of the theory to multiple-plate columns fails in usual distillation apparatus due to liquid mixing during the liquid flow period. To overcome this problem a new tray design consisting of a sieve plate and inclined surfaces has introduced an effective liquid time delay. Although the predicted efficiencies of 200% were not realized due to vapor movement hindering the tray action, a wide range of operating conditions provided good overall column efficiencies of 140%.

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## SCOPE

Previous theoretical and experimental work has shown that large improvements in performance are possible when a distillation column is operated in the cyclic mode. However, cycled experiments in multistage sieve plate columns have been unable to produce the large improvements predicted by the theory. This failure is attributed to interplate liquid mixing during the liquid flow period (LFP). The scope of this work was to determine whether particular hardware modifications enable the theoretical advantages of cycled columns to be achieved on an industrial scale. Experiments were performed in a 610 mm (24 in.) dia. five-plate cycled column using the methanol-water system. The column internals were modified and an external manifold added to reduce liquid mixing in the LFP, with the hope of achieving approximately double the number of effective plates when a distillation column was operated in the cyclic mode.

## CONCLUSIONS AND SIGNIFICANCE

A numerical solution of the differential mass transfer equations for the methanol-water system predicted double the number of effective plates when a column was operated in the cyclic mode. Mass transfer experiments on a one-plate cycled column showed the one cycled plate to give an equivalent separation of between 1.77 and 2.27 conventional plates. New column modifications provided some improvement over conventional sieve plates. However, further work is required to commercialize the approach. This work showed that the internals could not completely prevent liquid from falling more than one plate in the column. However, it is likely that the poor draining occurred because the external manifold was unable to vent the vapor space below each plate instantaneously at the start of the liquid flow period. However, even with these present limitations, overall column efficiencies of 140% were obtained. Thus it must be concluded that while the theoretical advantages of periodic cycling have been shown to apply to real systems, they have yet to be fully achieved in a large-scale column. The potential exists for further improvements by hardware modification.

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## INTRODUCTION

The concept of controlled cycling in separation processes was first introduced by Cannon in the early 1950s. Experiments on small laboratory-scale cycled columns have shown that controlled cycling provided higher capacities, simpler and cheaper plate design, more flexibility, and greater plate efficiencies than those of conventional columns. McWhirter and Lloyd (1963) were the first to observe overall column efficiencies of over 22%. They performed their mass transfer experiments on a 150 mm (6 in.) dia. column containing five packed plates with 76 mm (3 in.) of protruded package.

The first large-scale experiments were carried out by Schrodt et al. (1967) on a fifteen-plate, 305 mm (12 in.) dia. column with an acetone-water system. They found that while they could obtain throughputs of more than double the conventional operations, they

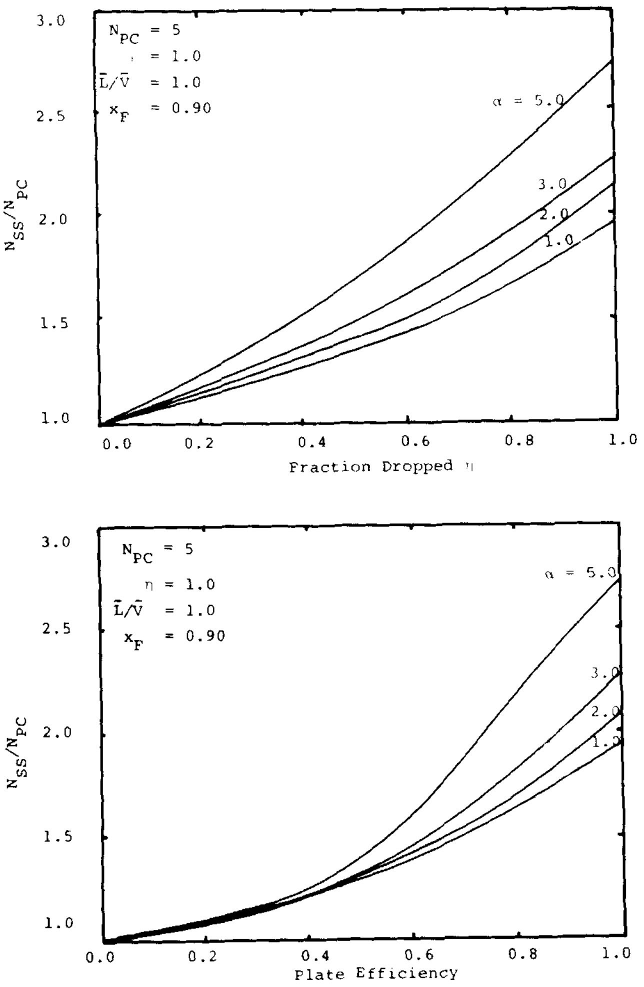

**Figure 1. Stripping section of a distillation column.**
**Effect of relative volatility and fraction dropped on cycled column performance with direct steam injection.**

were unable to achieve any improvements in column efficiency. Schrodt et al. observed that liquid fell more than one plate in the liquid flow period (LFP) and that the lower trays drained completely before any liquid flowed from the upper trays. Simulations by Wade et al. (1969) and Larsen and Kümmel (1979) showed that the poor performance observed by Schrodt et al. was due to a pressure gradient in the column during the LFP. They predicted that stable operation was possible if an external manifold was used.

A detrimental effect of using an external manifold is that the bypass flows in the manifold lead to reduced separations in the cycled column. Furzer (1980) solved the unsteady-state mass transfer equations for a cycled column fitted with an external manifold. He showed that for an exponentially varying area manifold design a favorable increase in separating ability could still be obtained in a cycled column.

Furzer and Duffy (1976) proposed simple stage models to explain liquid draining in cycled columns. The 1S model for nonideal draining assumed a fraction $\eta$ of the liquid holdup was transferred to the plate below in the LFP. In the 2S model fractions ($a$) and ($b$) of the liquid holdup were transferred to the next two stages respectively. Goss and Furzer (1980) used the 2S model to describe successfully the liquid movement in a five-plate, 100 mm (4 in.) dia. cycled distillation column. They studied the linear methylcyclohexane-$n$-heptane system.

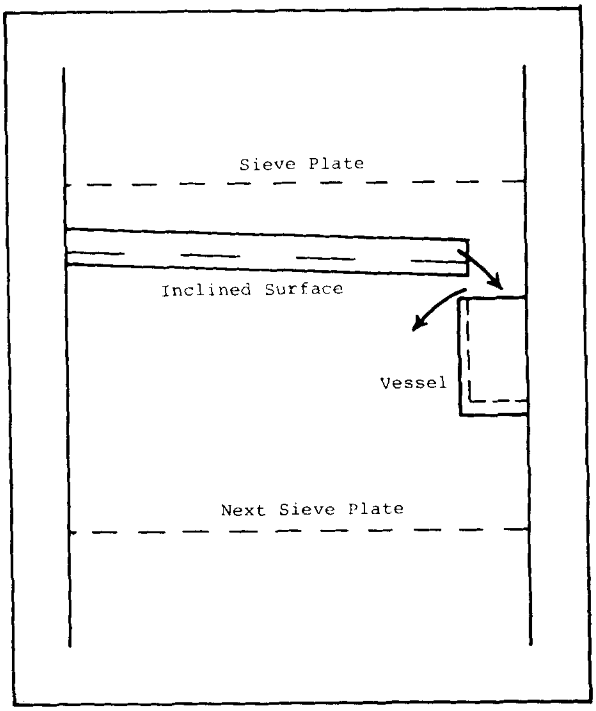

**Figure 2. Column internals, simplified.**
**Sieve plate, inclined surfaces, and an effectively closed downcomer to form a new combined tray. U.S. Pat. Applies.**

Goss and Furzer were only able to obtain moderate improvements in the separating ability of cycled columns with sieve plates and packed sieve plates. They concluded that in order to reproduce the results of McWhirter and Lloyd (1963) it would be necessary to modify the column internals to achieve control over the fractions ($a$) and ($b$) such that ($a$) approached unity and ($b$) approached zero.

Despite the intensive study of controlled cycling in mass and heat transfer operations, no one has yet been able to solve the problems of cycled operation so that the advantages of cycled columns can be realized on an industrial scale.

## THEORY

The unsteady-state mass and energy equations of a cycled column during the vapor flow period (VFP) are given by Szonyi (1981). The liquid phase activity coefficients for the methanol-water system were estimated by the UNIFAC group contribution method as given by Skjold-Jorgensen et al. (1979), leading to a correlation of the estimated VLE data by:

$$y_i^* = 6.387x_i - 29.684x_i^2 + 80.026x_i^3 - 116.693x_i^4 + 86.186x_i^5 - 25.222x_i^6 \quad (1)$$

A numerical solution of these system equations has been used throughout this work.

If the VLE is described by a relative volatility term, $\alpha$, the numerical solutions provide an indication for the separation in other systems. The stripping section of a distillation column with pure vapor input such as direct steam injection in aqueous systems is shown in Figure 1, for a range of the parameters $\alpha$, $\eta$, and $\epsilon$.

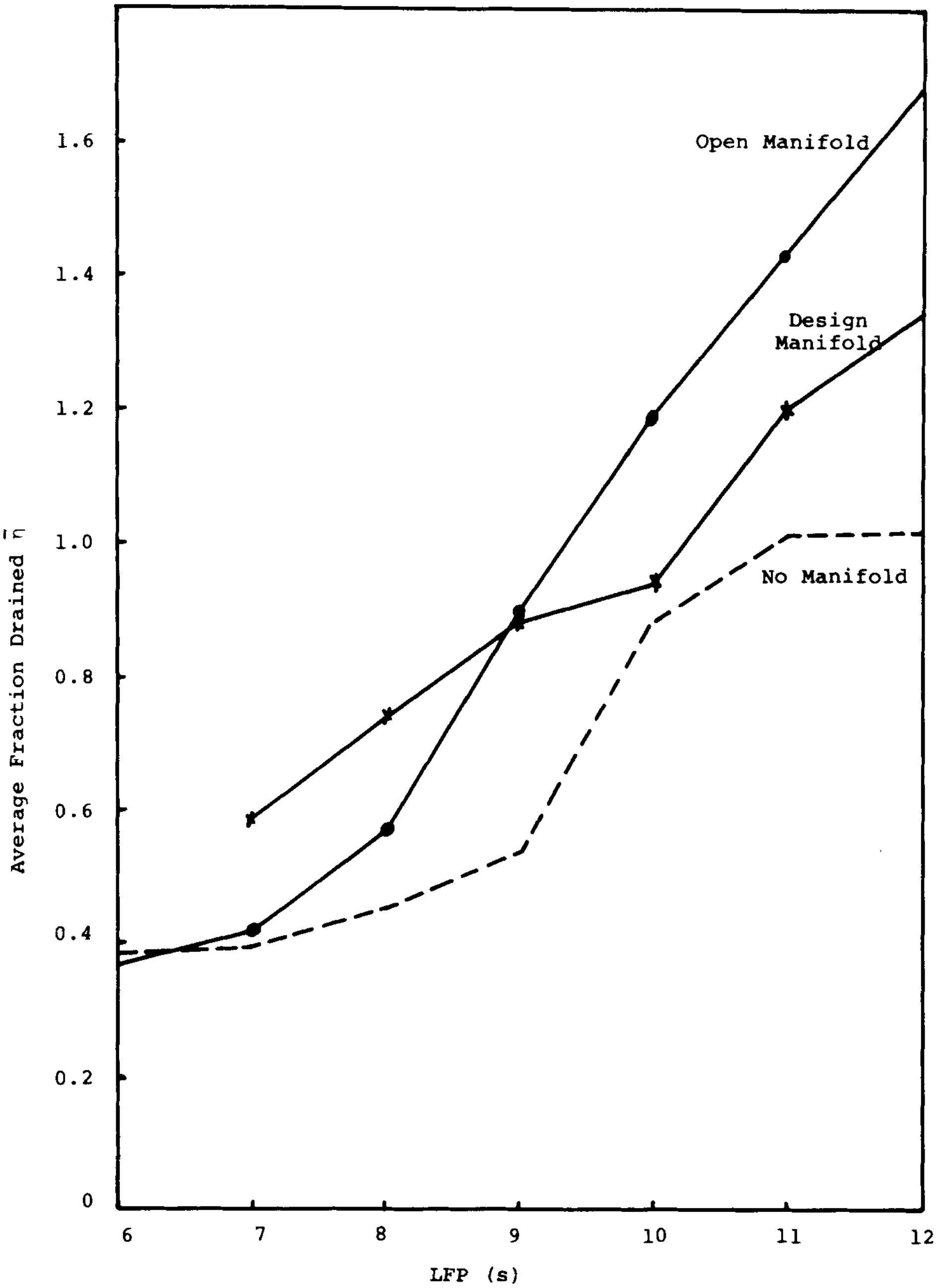

**Figure 3. Effect of manifold design and liquid flow period on fraction drained in a cycled five-plate column.**

## EXPERIMENTAL

### Apparatus

A detailed description of the experimental apparatus is given by Szonyi (1981). Controlled cycling experiments were performed in a five-plate, insulated mild steel column of 610 mm (24 in.) I.D. Trays on 610 mm (24 in.) spacing had a sieve plate section which contained 545 holes, of 6.4 mm (0.25 in.) dia. arranged on a 19 mm (0.75 in.) square grid. The percentage free area based on the total cross-sectional area was 5.9%. The new trays consisted of a sieve tray plus inclined surfaces and a vessel as shown in Figure 2 and described by Furzer (1983). The trays collected liquid from the sieve plate above and directed it to the vessel during the LFP. Liquid on the inclined surfaces and in the vessel experienced a time delay before flowing to the sieve plate below. These trays were shown to be extremely effective in transferring liquid from one plate to another without mixing of liquids from separate plates, in an air-water simulator.

An external manifold constructed from 25 mm (1 in.) I.D. pipe linked the vapor spaces below each plate. Its purpose was to equalize pressures at the start of the LFP and thus stabilize liquid holdup distributions in the column.

Cycling of the vapor was achieved by switching a 51 mm (2 in.) solenoid valve on the steam line. Steam was injected directly into a vapor chamber at the base of the column via two 38 mm (1.5 in.) dia. bayonet tubes. The steam entered the main section of the column by rising through a 305 mm (12 in.) dia. inverted funnel and T-piece. The funnel arrangement prevented liquid falling into the vapor chamber without impeding the flow of steam up through the column. A 25 mm (1 in.) dia. vapor bypass line extended from the vapor chamber at the base of the column to the condenser. The bypass line was fitted with a 25 mm (1 in.) dia. computer-controlled solenoid valve.

The overhead vapors from the column passed through a 250 mm (10 in.) dia. pipe to a single-pass inclined condenser. The condensed vapors or distillate were collected in the accumulator tank and recycled or pumped

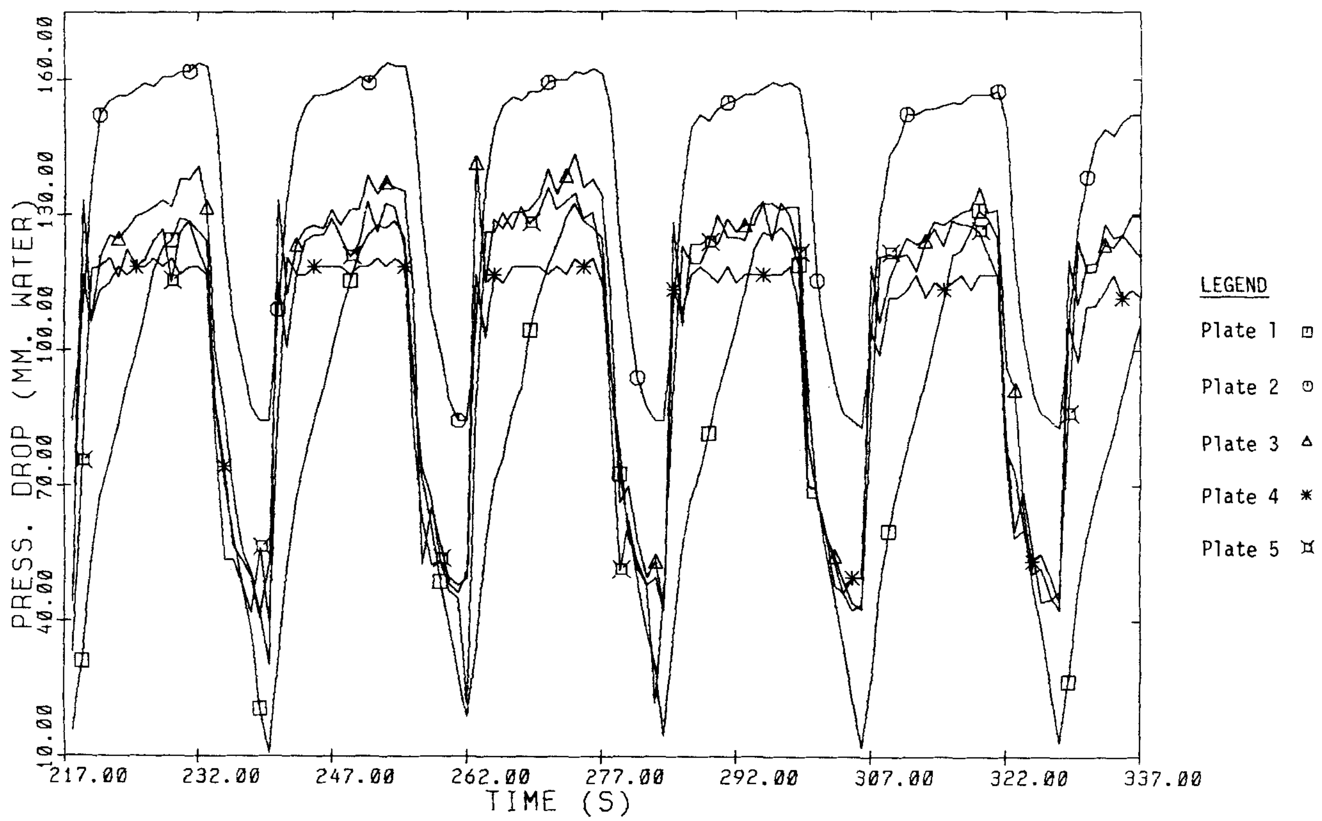

**Figure 4. Holdup distribution in a cycled column fitted with the design manifold. Cycles 11-15, experiment 3, for LFP of 9 s.**

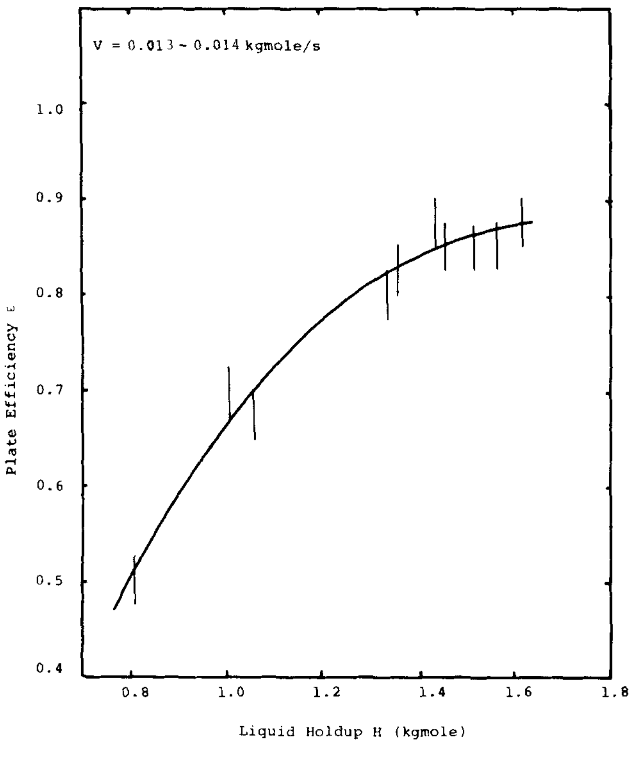

**Figure 5. Effect of liquid holdup on plate efficiency. Experiments 20-29; $V$ = 0.014 kmol/s.**

to the distillate tank. A vent tank connected to the accumulator insured that both the condenser and the column were open to the atmosphere.

Liquid feed was supplied to the column from a feed storage tank. The feed line was branched so that it was possible to add liquid to any plate of the column. The liquid level in the bottom of the column was recycled to the column or pumped to the bottom tank. An on-line PDP 11/45 digital computer and later an LSI/11 microprocessor system was used to control cycling, regulate liquid levels, and collect data.

## Experimental Results

Experiments with water were used to find the optimum manifold design which resulted in equal holdups on all plates. The flow of vapor through the manifold was controlled by means of restriction orifices inserted in the branches to the column. In the best design the deviation from the mean was less than 4% of the total holdup. The optimum orifice diameters were 8.2, 8.1, 6.5, 6.0, and 7.8 mm, respectively, for branches below plates 1 to 5. The vapor below plate 5 was also vented through a 25 mm (1 in.) dia. computer-controlled solenoid value in the LFP.

The average value of the liquid fraction leaving the plates $\bar{\eta}$ was investigated by using three manifold configurations. These were a closed manifold, a manifold with a distribution of branch diameters, and a fully opened manifold. The effect of the length of the liquid flow period on $\bar{\eta}$ is shown in Figure 3.

Typical measurements of the dynamic pressure drops across each plate are shown in Figure 4 for liquid flow periods of 9 s. This figure illustrates the stabilized column operation. The individual plate efficiency, $\epsilon$, was obtained on a single plate column for the methanol-water system. Samples were withdrawn from the plate during the VFP and analyzed by GLC, at 20 s intervals. The liquid holdup was measured at the end of the run by draining the liquid from the plate into a calibrated drum. The results at different liquid holdups are shown in Figure 5. The largest efficiency of 0.90 occurred at the maximum holdup of 1.6 kmol for a steam flow of 0.014 ± 0.005 kmol/s.

TABLE 1. RESULTS OF MASS TRANSFER EXPERIMENTS IN A ONE-PLATE CYCLED COLUMN

| Experiment No. | $\epsilon$ | $\overline{L}/\overline{V}$ | $x_F$ mol fr. | $x_D$ mol fr. | Experiment $x(\theta_v)$ mol fr. | Experiment $N_{SS}$ $\epsilon \leqslant 1.0$ | Theory $x(\theta_v)$ mol fr. | Theory $N_{SS}$ $\epsilon \leqslant 1.0$ |
|---|---|---|---|---|---|---|---|---|
| 30 | 0.85 | 1.47 | 0.166 | 0.211 | 0.0111 | 1.95 | 0.00789 | 2.25 |
| 31 | 1.00 | 1.84 | 0.166 | 0.182 | 0.00950 | 2.27 | 0.0100 | 2.20 |
| 32 | 0.95 | 1.85 | 0.166 | 0.232 | 0.0189 | 1.80 | 0.0123 | 2.07 |
| 33 | 0.90 | 1.78 | 0.149 | 0.164 | 0.00784 | 2.25 | 0.0105 | 2.09 |
| 34 | 1.00 | 1.99 | 0.149 | 0.233 | 0.0179 | 1.77 | 0.0107 | 2.13 |
| 35 | 1.00 | 1.92 | 0.149 | 0.209 | 0.0137 | 1.90 | 0.00939 | 2.20 |
| 36 | 1.00 | 1.56 | 0.149 | 0.194 | 0.00891 | 1.92 | 0.00402 | 2.56 |
| 37 | 0.80 | 1.84 | 0.149 | 0.229 | 0.0203 | 1.80 | 0.01693 | 1.92 |

Plate efficiencies were also determined from the temperature profile during the VFP. The variation in plate efficiencies was less than ±0.025 for the two different methods. The advantage of thermocouple measurements was that sampling errors were eliminated because the temperature was monitored automatically at 1 s time intervals. The operation of a single-plate column in a periodically cycled mode is shown in Table 1. Major increases in separating ability have occurred with the equivalent number of actual plates varying from 1.77 to 2.27. The plate efficiencies were determined from the temperature profile on the plate. The high plate efficiencies were due to an increase in steam flow from 0.014 to 0.016 kmol/s.

Twenty mass-transfer experiments using a methanol-water feed solution were performed in the five-plate column; the details are given by Szonyi (1981). Each run consisted of fifteen cycles, a VFP of 15 or 20 s, and a LFP ranging from 7 to 13 s. The methanol feed concentration ranged from 0.1 to 0.2 mole fraction. The feed was preheated to approximately 70°C and pumped onto the top plate at the start of each cycle. Feed was also added at the end of the fifteenth cycle and the steam flow to the column remained on at the end of the computer run. The steam was then switched off, the column drained, and the total column holdup determined. The experimental results are listed in Table 2.

The composition profiles in the column were determined from dynamic liquid temperature and the total pressure measurements at each plate. The atmospheric pressure was recorded for each run with a barometer. In addition to thermocouple measurements, liquid compositions were also determined from samples taken directly from the plates in the column and the storage tanks. Experiments 38 to 50 were all performed on the column fitted with an external manifold of optimum design. The two extreme cases of a fully open manifold, where all branches were 25 mm in diameter and no manifold, were investigated in experiments 51 to 57.

A detailed description of the temperature changes on each plate in a five-plate column fitted with an optimum manifold is shown in Figure 6, for cycles 11 to 15 with a LFP of 7 s. An important feature of this figure is the stabilized temperature profile over one cycle which is repeated in a regular manner for all plates. The figure shows that during the VFP the temperature rose as the methanol was stripped from the liquid on the plate. When the steam flow to the column was switched off, the plate temperature decreased as colder liquid with a higher methanol concentration drained from the plates above. As the LFP was lengthened, the fraction $\overline{\eta}$ of the mean holdup draining from the column increased. More time was required to refill the feed plate at the start of each cycle, and the rate of mass transfer on the top plate decreased. The fraction $\overline{\eta}$ was approximately equal to one at a LFP of 10 s. At longer LFP's the top plate drained completely, liquid dropped more than two plates, and the value of $\overline{\eta}$ was greater than one.

TABLE 2. RESULTS OF MASS TRANSFER EXPERIMENTS IN A FIVE-PLATE CYCLED COLUMN

| Experiment No. | LFP s | $\overline{\eta}$ | $\overline{L}/\overline{V}$ | $\overline{x}_F$ mol fr. | $\overline{x}_D$ mol fr. | $\overline{x}_B$ mol fr. | $x_5(\theta_v)$ mol fr. | $N_{SS}$ $\epsilon \leq 1.0$ |
|---|---|---|---|---|---|---|---|---|
| | | | | Design Manifold | | | | |
| 38 | 8 | 0.78 | 3.70 | 0.125 | 0.439 | 0.00453 | 0.00221 | 7.0 |
| 39 | 8 | 0.77 | 3.48 | 0.130 | 0.394 | 0.00248 | 0.00138 | 7.0 |
| 40 | 7 | 0.64 | 2.91 | 0.130 | 0.355 | 0.00304 | 0.000452 | 4.6 |
| 41 | 8 | 0.80 | 3.46 | 0.117 | 0.370 | 0.00193 | 0.00114 | 6.5 |
| 42 | 8 | 0.69 | 3.15 | 0.117 | 0.373 | 0.00254 | 0.000794 | 5.0 |
| 43 | 9 | 0.85 | 3.36 | 0.130 | 0.413 | 0.00286 | 0.00155 | 6.1 |
| 44 | 10 | 0.92 | 3.84 | 0.128 | 0.424 | 0.00950 | 0.00434 | 6.3 |
| 45 | 10 | 0.94 | 4.12 | 0.109 | 0.383 | 0.00698 | 0.00283 | 7.1 |
| 46 | 10 | 1.00 | 4.09 | 0.101 | 0.327 | 0.00550 | 0.00241 | 6.4 |
| 47 | 11 | 0.92 | 3.03 | 0.131 | 0.339 | 0.000993 | 0.000697 | 6.3 |
| 48 | 12 | 1.04 | 3.06 | 0.131 | 0.383 | 0.00181 | 0.000845 | 5.7 |
| 49 | 13 | 1.23 | 3.37 | 0.137 | 0.415 | 0.00384 | 0.00182 | 6.0 |
| 50 | 12 | 1.12 | 3.75 | 0.129 | 0.446 | 0.00654 | 0.00346 | 7.2 |
| | | | | Open Manifold | | | | |
| 51 | 8 | 0.78 | 3.29 | 0.129 | 0.421 | 0.00277 | 0.00161 | 5.8 |
| 52 | 7 | 0.64 | 3.20 | 0.119 | 0.397 | 0.00257 | 0.00108 | 5.2 |
| 53 | 9 | 0.83 | 3.37 | 0.119 | 0.394 | 0.00329 | 0.00137 | 5.4 |
| 54 | 8 | 0.76 | 3.39 | 0.132 | 0.406 | 0.00179 | 0.00108 | 7.2 |
| | | | | No Manifold | | | | |
| 55 | 9 | 0.83 | 3.35 | 0.132 | 0.452 | 0.00469 | 0.00138 | 5.3 |
| 56 | 10 | 0.83 | 3.08 | 0.107 | 0.298 | 0.000492 | 0.000407 | 6.6 |
| 57 | 11 | 0.91 | 3.06 | 0.107 | 0.302 | 0.000893 | 0.000546 | 5.8 |

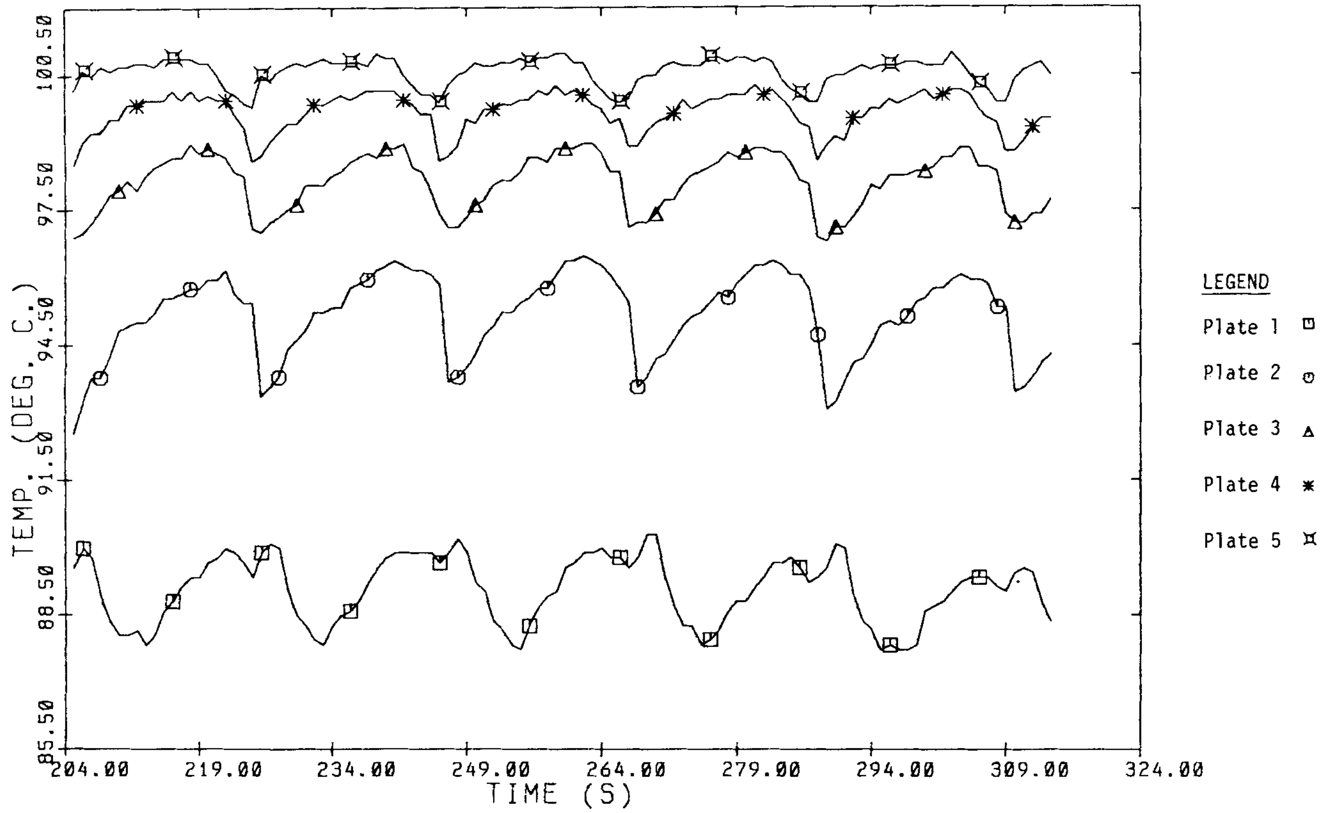

Figure 6. Temperature profiles in a cycled column fitted with the design manifold. Cycles 11-15, experiment 40, for LFP of 7 s.

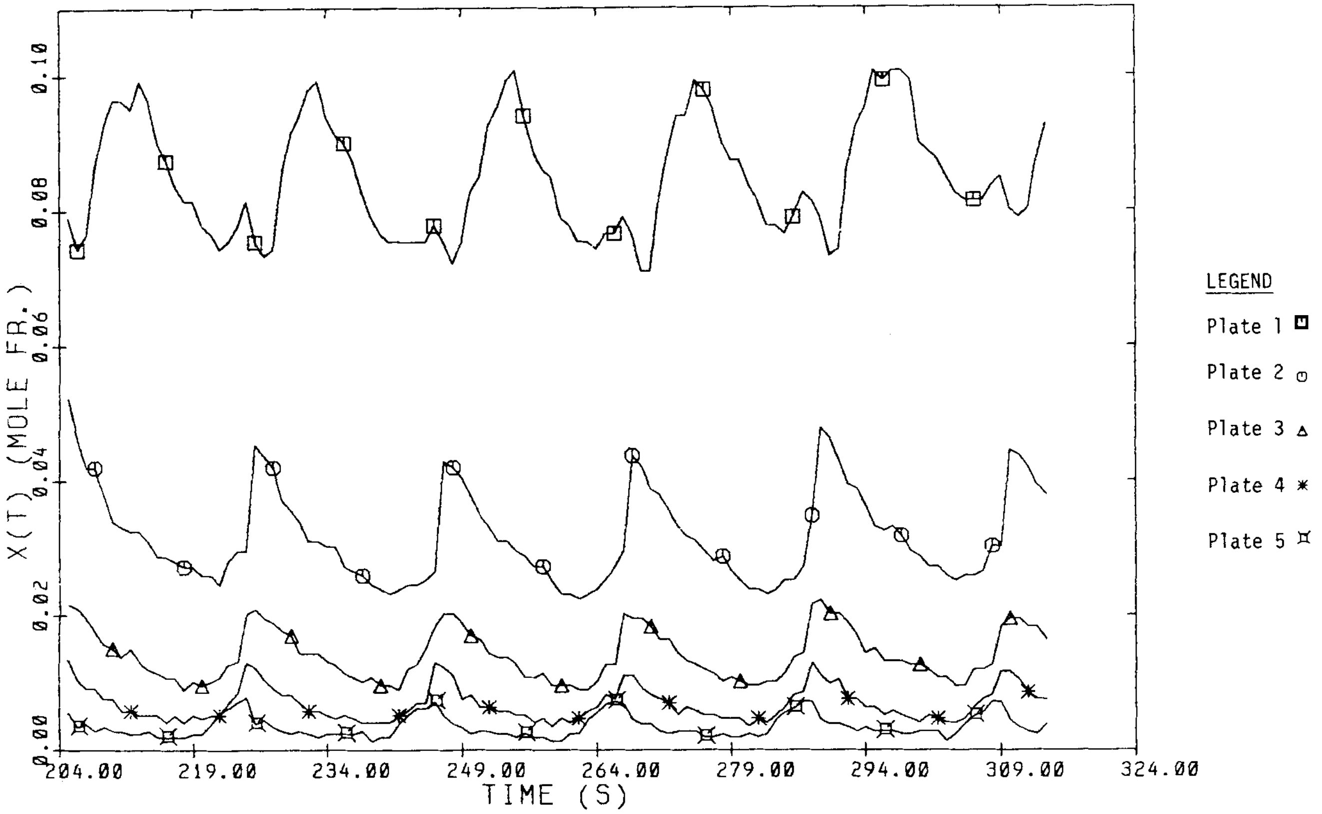

Figure 7. Composition profiles in a cycled column fitted with the design manifold. Cycles 11-15, experiment 40, for LFP of 7 s.

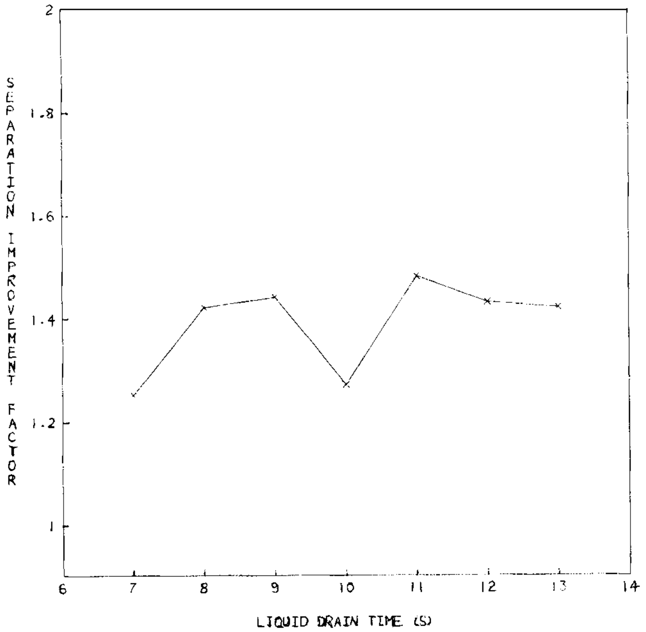

**Figure 8. Separation improvements in a five-plate periodically cycled plate column.**

The composition on each plate was determined as a function of time from the temperature profile and the total pressure above each plate. The temperature-composition equilibrium curve at a given column pressure was calculated by the UNIFAC group contribution method as given by Skjold-Jorgensen et al. (1979). A plot of the composition profiles for cycles 11 to 15 of experiment 40 is shown in Figure 7. The stabilized composition profile per cycle should be noted.

## RESULTS

The mean values of $N_{SS}$ for each particular value of the LFP have been calculated using a McCabe-Thiele program. The separation improvement factor, $N_{SS}/N_{PC}$ varied with the LFP as shown in Figure 8. This factor shows a rise from 1.25 to 1.42 as the LFP increased from 7 to 9 s. The next result at 10 s corresponds to a high value of $\bar{L}/\bar{V}$ leading to a near approach to a pinch condition at the top of the column. In this case the overhead distillate was highly enriched in methanol. Higher values above 1.40 were obtained at higher values of the LFP. We might summarize these results by stating that an overall column efficiency in excess of 140% was obtained over a wide range of LFP using the new tray design for a five-plate column.

The theoretical column performance of 200% was not obtained because of liquid draining more than one plate in the liquid flow period. The long liquid flow periods were necessary because draining did not start immediately at the beginning of the LFP. These results indicate that the venting arrangements were inadequate. Further expansion of manifold dimensions and the introduction of vapor flow control through the manifold in the VFP are necessary to fully utilize the potential of the new tray design.

## ACKNOWLEDGMENT

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## NOTATION

$L$ = instantaneous liquid flow rate, kmol/s
$\bar{L}$ = mean liquid flow rate, kmol/s
$N_{PC}$ = actual number of plates in a periodically cycled column
$N_{SS}$ = actual number of plates in an equivalent steady-state column with the same plate efficiency, $\epsilon$
$V$ = instantaneous vapor flow rate, kmol/s
$\bar{V}$ = mean vapor flow rate, kmol/s
$x$ = liquid concentration, mol/mol
$X_F$ = composition of liquid feed pumped into the column, mol/mol
$y^*$ = vapor composition in equilibrium with liquid on a plate, mol/mol

### Greek Letters

$\alpha$ = relative volatility defined as $y^* = x\alpha/[1 + (\alpha - 1)x]$
$\epsilon$ = Murphree plate efficiency
$\bar{\eta}$ = mean fraction of plate holdup draining from the column in the LFP

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