Influence of Liquid Redistributors on the Mass-Transfer Efficiency of Packed Columns

M. Schultes

Raschig GmbH, Mundenheimerstrasse 100,
67061 Ludwigshafen, Germany

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In industrial mass-transfer columns liquid distributors and redistributors are used to mix the liquid after a certain random packing or structured packing height before it is applied to the next bed. The purpose of this is to counter a possible deterioration of the mass-transfer efficiency along the height of the column as a result of wall flow tendency of the liquid or to minimize the deterioration of the mass-transfer efficiency as a consequence of maldistribution of the liquid by the liquid distributor located above. However, when planning industrial columns, this influence on the mass-transfer efficiency can only be quantified with some difficulty which is why empirical standard values are mostly used for maximum bed and packing heights. This analysis shows that the height, after which liquid redistribution should take place, depends on numerous further influencing factors. It becomes evident that uneven irrigation of a packing with liquid over the cross section of the column is also a cause for the decline in the mass-transfer efficiency along the packed bed. The resultant decline in the mass-transfer efficiency is influenced, for instance, by the gas/liquid equilibrium behavior of the mixture that is to be separated, by the L/V flow ratio in the rectification, absorption, and desorption columns, by the type of packing used, and by the number of theoretical stages to be executed. A simulation procedure is shown, with whose assistance these factors influencing the mass-transfer efficiency can be recorded quantitatively.

Introduction

The design of rectification, absorption, or desorption columns is usually done, nowadays with the aid of computer programs. Many thermodynamic values, such as physical properties of gas and liquid phases, phase equilibrium, and the number of theoretical stages for a mass-transfer task, can meanwhile be determined by using simulation calculations. This is very limited given only for the fluid dynamic and mass-transfer efficiency of column packings. Thus, when maximum loads, pressure drops, liquid holdup, and above all the mass-transfer efficiency of packings are described, the determination of these values is chiefly based on information supplied by manufacturers' experience and/or results obtained with experimental columns. However, these experimental columns generally have relatively small dimensions which is why the question is always asked as to what degree the results of such experiments can be adopted in the planning of large-scale columns.[1,2] Thus, it has been repeatedly shown in the past that serious differences can exist in the fluid-dynamic and mass-transfer-efficient behavior of laboratory columns in comparison with industrial systems.[3–5]

The discussion, for example, about scaling up of experimental results is often based on the experience available to date that packed columns react sensitively to the maldistribution of the vapor or liquid phase,[5–10] which is why special attention is given on the design of distribution and redistribution systems of the phases.

This is what distinguishes packed columns from tray columns, which already bring about equal distribution of the gas phase over the column cross section because of their large pressure drop.

In addition to this, the liquid phase in tray columns is continuously redistributed from tray to tray, whereas in packed columns it is applied by means of a liquid distributor at the top of the packing and then flows downward solely subject to gravity.[11]

The necessity of liquid redistributors in high packed columns and/or their influence on the mass-transfer efficiency is the subject of discussions time and again.[12] Ultimately, empirical values which state that liquid flow in packed columns should be redistributed if a mass-transfer-efficient column height of 10 theoretical stages is achieved are often assumed.[6] In the case of modern packings, a maximum of 20 theoretical stages is allowed without a liquid redistributor.[13] Moreover, for small column diameters the ratio between the packing heights and the column diameter, which should not exceed a value of H/D = 5–10, is taken into consideration as a further criterion.[14,15] Irrespective of these estimates, there are industrial columns operating which generally redistribute the liquid phase after a column height of 3–5 m.

To minimize the influence of a maldistribution of the liquid phase on the mass-transfer efficiency, modern liquid distributors have been designed by various packing manufacturers in the past few years. Although these designs ensure very even liquid distribution at the beginning of a bed and wall flow tendency is very slight in modern packings, the above rules continue to be observed for liquid redistribution under industrial conditions.

Basis of Calculation

Various research work has repeatedly confirmed that the liquid in a packed bed trickles down with differing intensity over the column cross section.[16] If the column cross section is divided into small sections and the local volume rates of these sections are measured, it will be seen that in some areas large trickles flow downward, whereas other areas only have small trickles or drop flows.[17] Figure 1a shows measuring results in this connection for a structured packing. The maldistribution factor M_f is a characteristic value for the inequality of
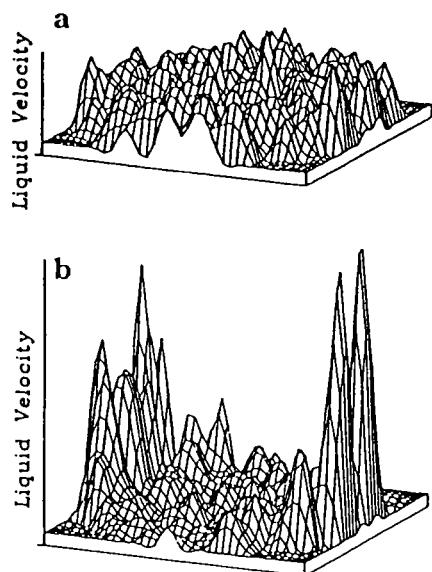

Figure 1. (a) Liquid velocity profile leaving the bottom of a Mellapak 250 Y structured packing (M_f = 0.26) for gas velocities below the loading point. (b) Liquid velocity profile leaving the bottom of a Mellapak 250 Y structured packing (M_f = 1.74) for a gas velocity above the loading point.

the volume rates over the column cross section and is determined from the various individual volume rates as defined by eq 1. In eq 1 L_i^{vol} describes the liquid volume rate of each individual section, L_{mean}^{vol} the mean volume rate of all sections, and n the number of section elements. The mean volume rate can be calculated by the total liquid rate L_0 divided by n.

$$M_f = \frac{1}{n} \sum_{i=1}^{i=n} \frac{(L_i^{vol} - L_{mean}^{vol})^2}{(L_{mean}^{vol})^2} \qquad (1)$$

The measurements by Stikkelman[17] were carried out in a laboratory column with a liquid distributor which ensured homogeneous liquid distribution. The distribu-

Table 1. Maldistribution Factors for Different Random and Structured Packings[13,17]

packing	material	maldistribution factor	data base
25 mm ralu-ring	plastic	0.55/0.5	Stikkelman/author
38 mm ralu-ring	plastic	0.69	Stikkelman
50 mm ralu-ring	plastic	0.46	author
25 mm pall-ring	metal	0.81/1.1	Stikkelman/author
50 mm pall-ring	metal	0.50	author
25 mm IMTP-ring	metal	0.57	Stikkelman
Raschig super-ring no. 2	metal	0.28	author
Ralu-Pak 250 YC	metal	0.21	author
Mellapak 250 Y	metal	0.24	Stikkelman
Mellapak 500 Y	metal	0.21	Stikkelman
Mellapak BX	plastic	0.22	Stikkelman
Montz B1-250	metal	0.23	Stikkelman
Montz BS-450	metal	0.44	Stikkelman

tor consists of a hollow plate, perforated with vertical gas tubes and provided 760 drip points/m² of column cross section. The variance of the initial liquid distribution of this distributor was less than 0.5%. It can be seen that maldistribution is generated on the packing despite the good liquid distribution at the top of the column.

The degree of maldistribution is for the most part independent of the gas flow as long as the operating point of the two-phase countercurrent flow is below the loading point. Above the loading point, the shearing stress forces of the gas are large enough to hold up the liquid flow so that the maldistribution is intensified, s (Figure 1a,b).

The maldistribution is also dependent on the type of packing used. On a structured packing the liquid films are spread more evenly on the surface than in traditional dumped packings, where in addition to thin films large trickles can occur along the metal strip and drops are formed on the drip edges of the packing elements. Table 1 shows the maldistribution factors of various packings. With the exception of the Raschig super-ring, the random packings all display higher maldistribution factors than structured packings. In the case of Raschig super-ring, this is due to the fact that drops and large trickles are avoided as a consequence of the geometry of the Raschig super-ring;[18] see Figure 2.
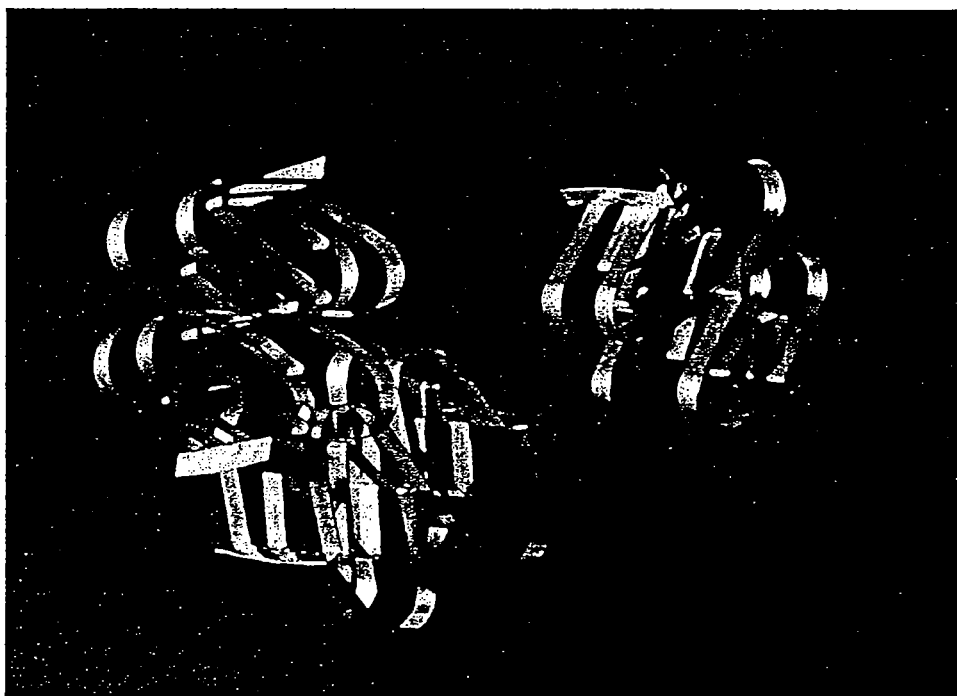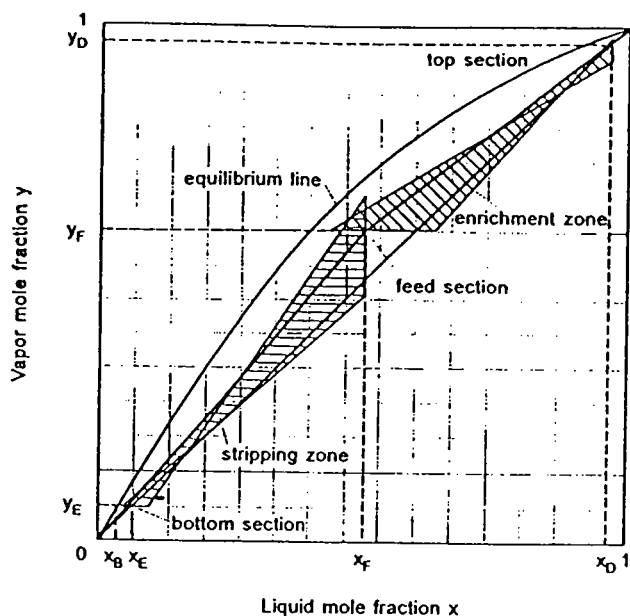

Figure 2. Photo of a Raschig super-ring.

Figure 3. McCabe–Thiele diagram with L/V variations.

liquid rate $L_1 = (1 - \delta)L_0/3$, the second section $L_2 = L_0/3$, and the third section $L_3 = (1 + \delta)L_0/3$. Because cross-mixes may occur between the different liquid flow rates, the variation factor δ is not identical with the maldistribution factor M_f.

Influence of Liquid Flow Rate Variation on the Mass-Transfer Efficiency in Distillation Columns

The influence of liquid flow rate variation on the mass-transfer efficiency will be described for a distillation unit with an enrichment zone above the feed section and a stripping zone below. The unequally distributed liquid over the three column cross sections is trickling downward countercurrent to the uprising gas flow. The latest is entering the enrichment zone uniformly distributed and ideally mixed by the column internals located at the feed section. Caused by the material and component balances, different concentrations for the liquid and gas flow leaving the enrichment zone result. For the simulation it was assumed that all parallel sections need the same number of theoretical stages.

At the feed section the liquid from the enrichment zone is assumed to be ideally mixed with the feed before entering the stripping zone. The liquid mixture is then redistributed unequally to the sections of the stripping zone by the variation factor δ. The liquid leaving the stripping zone at the bottom of the column gets again different concentrations because of component and material balances. At the sump of the column the liquid is mixed and fed to the reboiler. The flow of vapor leaving the reboiler is fed to the sections of the stripping zone in the same flow rates and with a uniform concentration. The gas flows upward and is leaving the stripping zone in different concentrations again because of the component and material balances.

The gas leaving the top of the column is mixed and fed to the condenser where the overhead product is generated. In Figure 3 x_D, x_F, and x_W are the distillate, feed, and bottom concentrations of liquid and y_D is the vapor concentration reaching the condenser. y_E and x_E are the vapor and liquid concentrations entering and leaving the packing at the bottom of the stripping zone. It can be concluded from Figure 3 that the areas for the enrichment and stripping zones all become larger, the
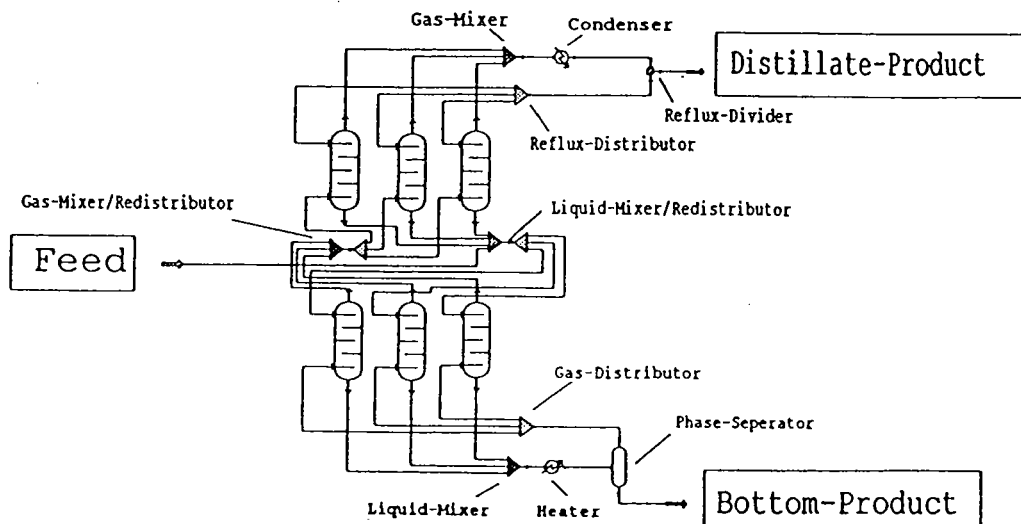
Stikkelman shows that the gas is, in general, relatively evenly distributed over the column cross section if no gas maldistribution is initiated at the gas feed location, so that the maldistribution of the liquid phase results directly in divergences of the L/V flow ratios. To determine the influence of differing L/V flow ratios on the mass-transfer efficiency of a packed column with a process simulator, the packed bed can be subdivided into several parallel bed sections.[19] The mass-transfer efficiency of these bed sections can be calculated according to the number of theoretical stage models.

The simulation of a column subdivided in such a way can be explained as follows on the basis of the McCabe–Thiele diagram of Figure 3 for the rectification of a two-component mixture and the flowchart of Figure 4.

At the top of the column the liquid flow L_0 is unequally distributed over, for instance, three column sections as the reflux. The influence of unequal distribution on the mass-transfer efficiency can be characterized by the variation factor of liquid flow rate $\delta = \Delta L/L_0$ that typifies a characteristic relative deviation from the mean liquid load L_0. The first section receives the

Figure 4. Flowchart of a rectification process without redistributors in the enrichment and stripping sections.

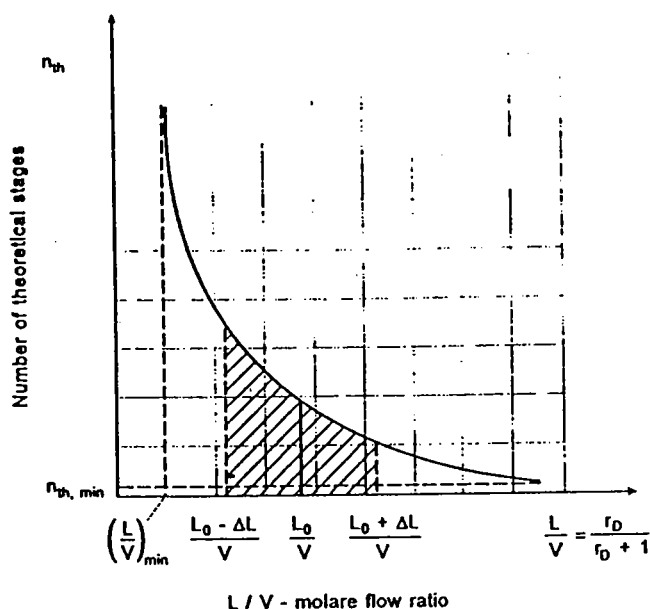**Figure 5.** Dependence of the requisite number of theoretical stages on the L/V ratio of a rectification column.

Figure 6. McCabe–Thiele diagram with redistributors and L/V variations.

greater the variation factor δ chosen as a consequence of the differing L/V ratios in the different column cross sections.

Figure 5 shows the corresponding ratios in the form of the dependence of the required number of theoretical stages over the L/V flow ratio and the reflux ratio r_D of a column. Already before the nominal reflux ratio L_0/V reaches the minimum reflux ratio, a critical operation comes about. This occurs when the column section with the smaller L/V flow ratio reaches the equilibrium curve. This column section consequently loses the mass-transfer efficiency tremendously because the number of theoretical stages is specified by the given packed height of the column. The loss in the mass-transfer efficiency cannot be compensated by the column section with the higher L/V flow ratio. This results in the fact that the mass-transfer efficiency of a rectification column generally decreases with an increasing variation factor δ and declining reflux ratio.

Influence of Liquid Redistributors on the Mass-Transfer Efficiency in Distillation Columns

As described above, the closer a distillation column operates in the vicinity of the minimum reflux ratio, the more theoretical stages that are required to fulfill the separation task. A large number of theoretical stages in the enrichment or stripping zone of a distillation unit results in high packed beds so that liquid redistributors have to be installed in between. Figures 6 and 7 show how these liquid redistributors can be taken into account by the process simulation.

In analogy to Figures 3 and 4, the different L/V flow ratios in a packed bed can be taken into consideration in the calculation as a result of the fact that the portion of column cross section with the lower irrigation density is simulated by a column with a lower L/V flow ratio, whereas the second parallel mass-transfer column is calculated with the middle value and a third one with a higher L/V ratio.

At the locations of liquid redistributors, the down-flowing liquid and the rising vapor are ideally mixed.
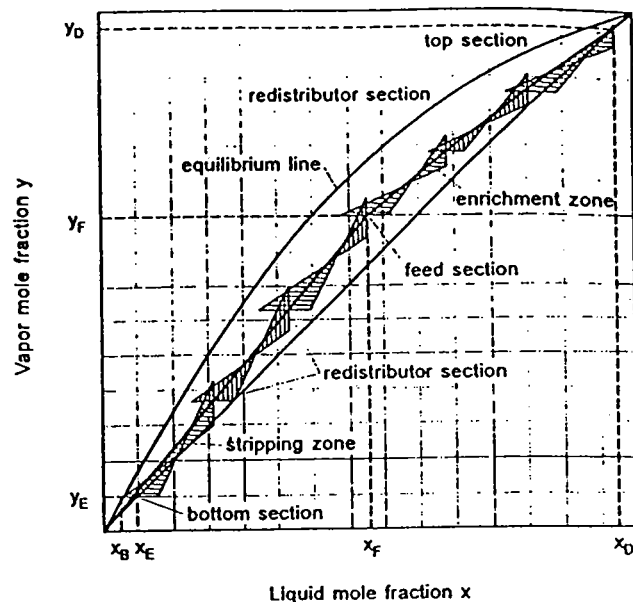
In the simulation the downflowing liquid from the redistributors is again varyingly applied to the subjacent sections by the variation factor δ, whereas the vapor phase flows evenly distributed to the above-lying sections. As a result, the areas around the theoretical enrichment and stripping lines in Figure 6 become smaller in comparison to Figure 3 so that the height segments of the column are operating in a greater distance to the equilibrium line.

Figure 8 shows the changes in the top concentration with an increasing variation factor δ and a varying number of liquid redistributors for the rectification of a chlorobenzene/ethylbenzene mixture under vacuum as an example.

In the evaluation it was presupposed that the chlorobenzene/ethylbenzene mixture flows into the column with a concentration of 50 mol % and that both a concentration of 99 mol % of the low-boiling component at the top of the column and a concentration of 99 mol % of the higher boiling component at the bottom of the column are to be achieved. The separation of the feed mixture is effected under vacuum with a pressure of 66 mbar. From a simulation calculation it follows that the requisite conditions are achieved if the column is operated with a theoretical reflux ratio of $r_D = 37.5$ and if 36 theoretical stages are provided in the enrichment section and 36 theoretical stages in the stripping section. In the case of a structured packing with a specific surface area of 250 m²/m³, the HETP value is calculated as 0.4 m, yielding a packing height of 14.4 m in both the enrichment and stripping sections. Under these conditions the column is operated with 1.8 times the minimum reflux ratio. The influence of the pressure drop along the mass-transfer column was not included in the evaluation.

It can be seen in Figure 8 that the requisite product purities are no longer achieved with an increasing variation factor δ. Even a variation of liquid flow of 10% brings about a fall in concentration to 93% for the low-boiling component at the top of the column if no liquid redistributor is used. With an increasing variation factor δ product impurities increase considerably. Product impurities can be significantly reduced if a redistributor
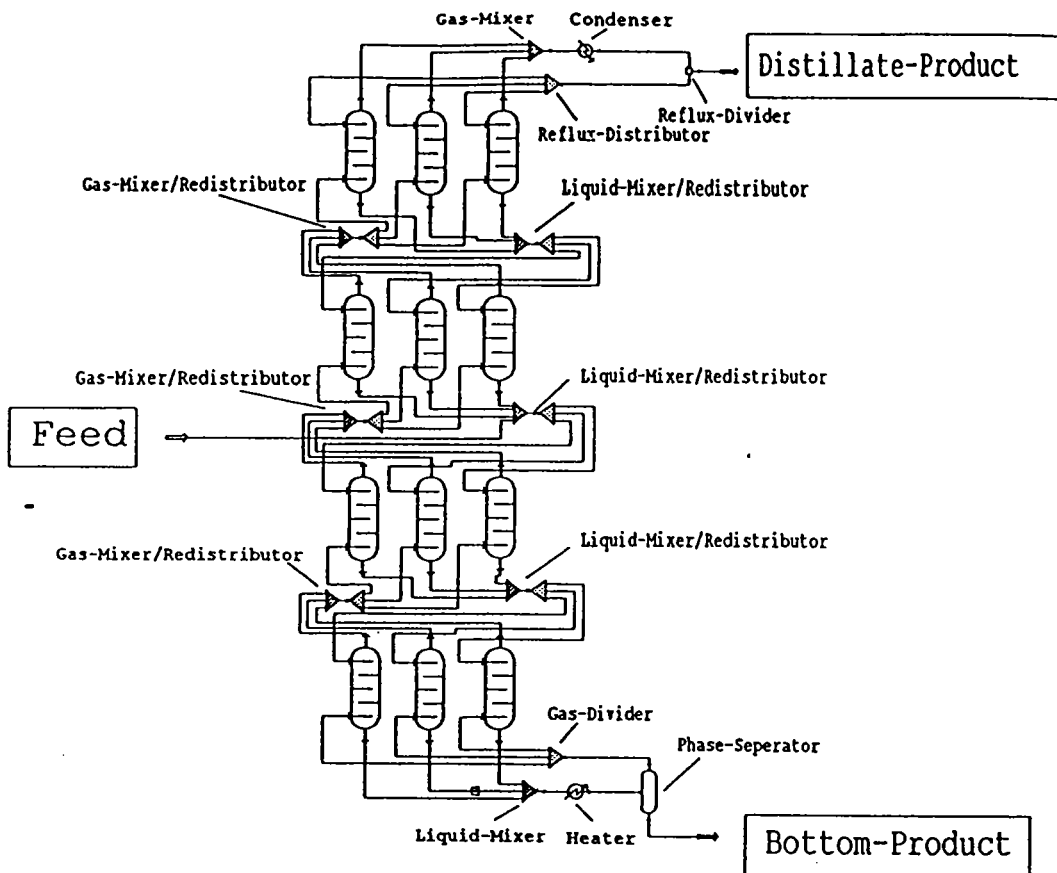
Figure 7. Flowchart of a rectification process with redistributors in the enrichment and stripping sections.
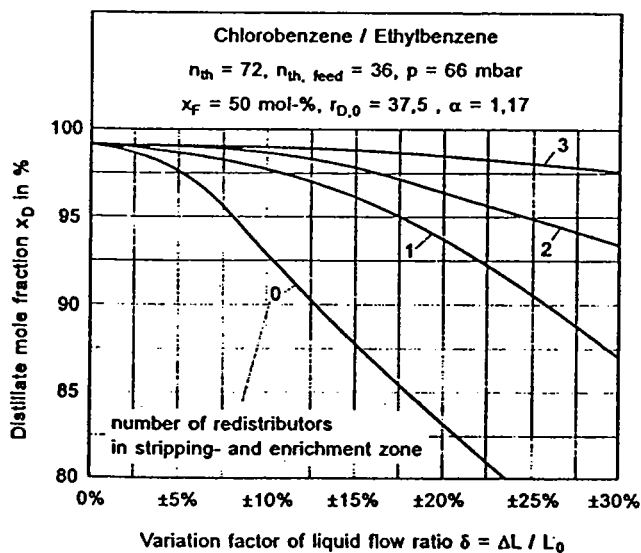

Figure 8. Change in the top concentration with an increasing variation factor $\delta = \Delta L/L_0$ and a varying number of liquid redistributors.

Figure 9. Change in the number of theoretical stages with an increasing variation factor $\delta = \Delta L/L_0$ and a different number of liquid redistributors.

is used in both the enrichment section and the stripping section in each case after 18 theoretical stages. This continues if in each case two redistributors are used at a distance of 12 theoretical stages or three redistributors at a distance of 9 theoretical stages. Figure 8 shows that with two redistributors in the enrichment and stripping sections and with a variation of liquid flow of 15% in the overhead product, a concentration of 98% of the low-boiling component is still achieved.

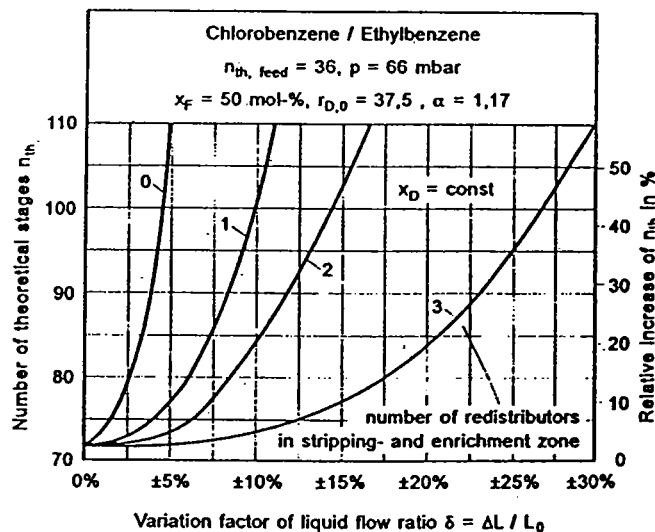If the number of theoretical stages are determined, which would be necessary to achieve the requisite

bottom and top concentrations of 99 mol % with the various variation factors, Figure 9 follows. It can be seen that even with a variation factor of 5% the requisite concentration is virtually no longer attainable without liquid redistributors. With a variation factor of 10% and with one redistributor in the enrichment and the stripping sections, 43% more theoretical stages are needed, and with two redistributors in each case, 20% more theoretical stages are needed. Only with three redistributors and a variation factor of 20% can the requisite concentration be achieved still with 20% more theoretical stages. In analogy to this, the concentration
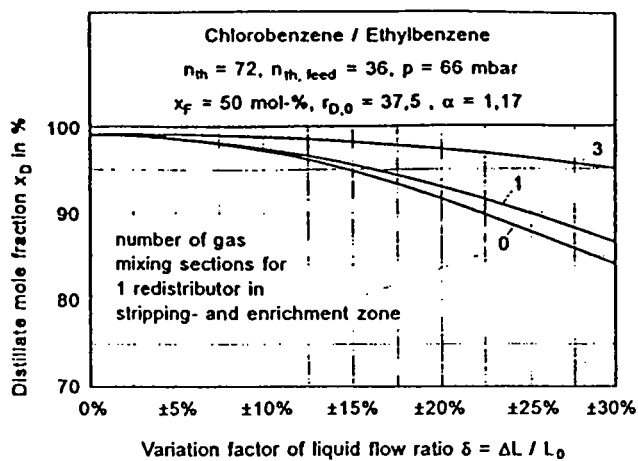
Figure 10. Influence of the gas-phase mixing within the column sections with one redistributor in stripping and enrichment zones.
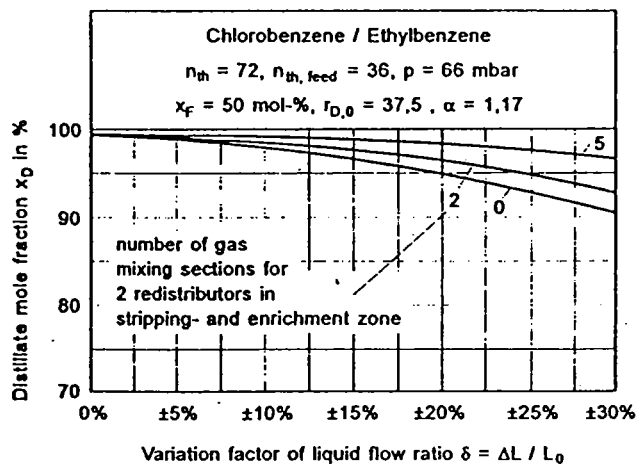

Figure 11. Influence of the gas-phase mixing within the column sections with two redistributors in stripping and enrichment zones.

at the bottom of the column changes with increasing variation factors.

In the evaluations presented, a homogeneous gas flow over the column cross section was taken into consideration. It was also assumed that the gas flow will be ideally mixed at each liquid redistributor prior to re-entry into the column sections above. Cross-mixes of gas flow between the column sections were not taken into consideration but are possible in reality because of the turbulence in the continuous gas flow. A cross-mix in the continuous flow of gas between the sections can be taken into account in the simulation by having the gas phase within the sections partially or fully drawn off, ideally mixed, and fed back. The result of such a calculation is shown in Figures 10 and Figure 11 in the case that one or two redistributors are installed in the stripping and enrichment zones. In this sample calculation a fully drawn off, i.e., complete, mixing of the gas phase was assumed.

It can be seen that as a result of additional homogenization of the flow of gas within the sections the differences in the liquid concentrations between the sections also become less and that the drop in concentration of the distillate product is reduced. Whereas with small column diameters an effective cross-mix within the gas flow can be assumed, the residence time of the gas phase in the bed sections between the height of the redistributors will not be sufficient in large column diameters.
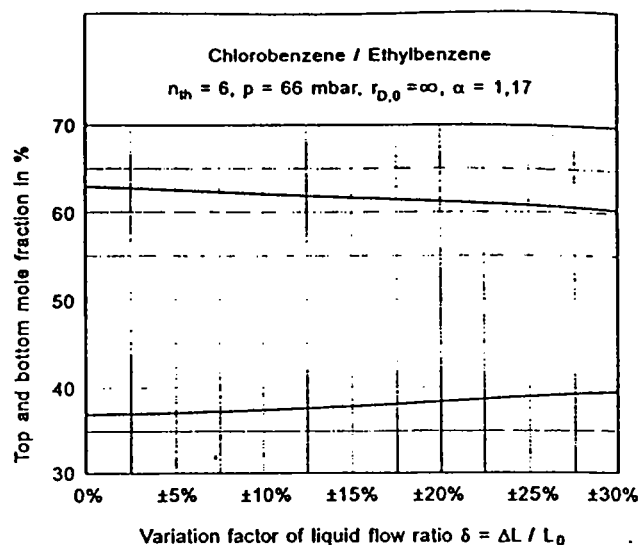

Figure 12. Change in the top concentration with increasing variation factor $\delta = \Delta L/L_0$ in a laboratory column.
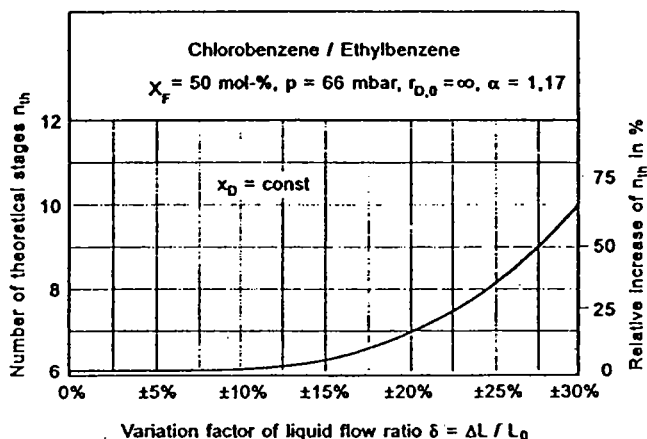

Figure 13. Change in the number of theoretical stages with increasing variation factor $\delta = \Delta L/L_0$ in a laboratory column.
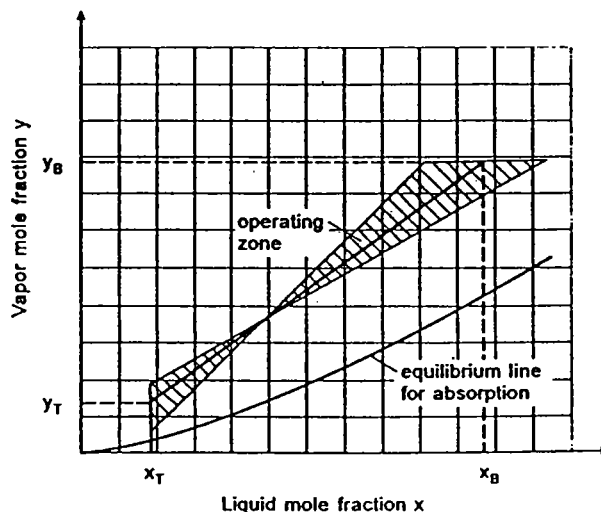

Figure 14. McCabe–Thiele diagram with L/V variations for an absorption column.

The relationships shown usually mainly occur under industrial conditions because mass transfer in laboratory columns or pilot-plant facilities takes place with an infinite reflux ratio with a limited number of theoretical stages and a medium concentration range. Figure 12 shows a corresponding evaluation for a
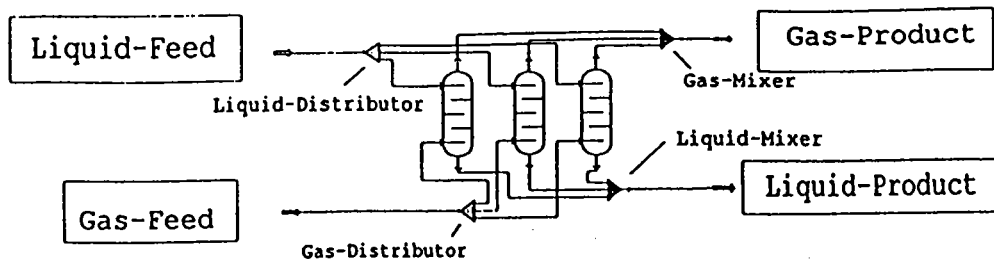
Figure 15. Flowchart of an absorption/stripping column without redistributors.
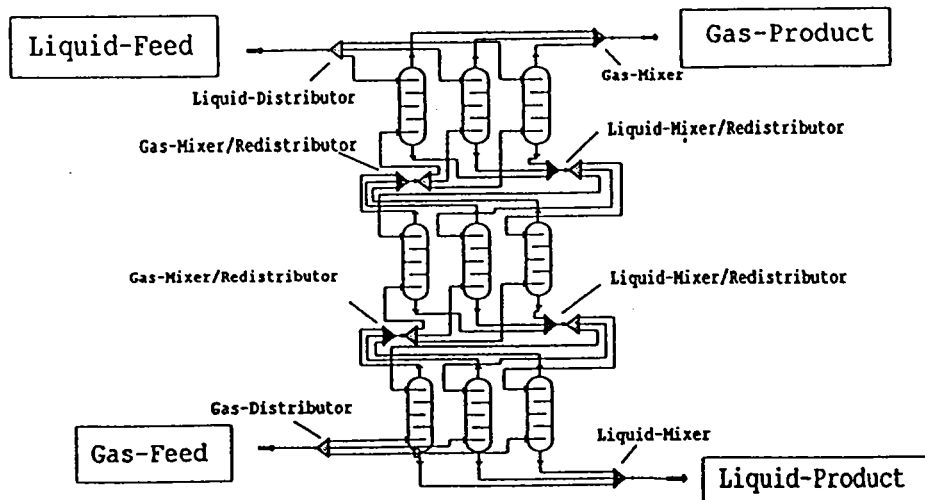

Figure 16. Flowchart of an absorption/stripping column with redistributors.

laboratory column, which achieves six theoretical stages. It can be seen that up to a variation factor of 15% only slight changes can be detected in the concentrations at the top and bottom. This can also be seen in Figure 13, which shows the increase in theoretical stages with an increasing variation factor for the experimental column with constant distillate concentration. This means that the described influence of liquid-phase flow variation in laboratory or pilot-plant conditions may only be determined if mass transfer takes place at a very low or very high concentration level.

Influence of Liquid Redistributors on the Mass-Transfer Efficiency in Absorption and Desorption Columns

The simulation of an absorption or desorption column with unequal liquid variation over the column cross section can be explained as follows on the basis of the McCabe−Thiele diagram of Figure 14 and the flowchart of Figure 15.

At the top of the column the liquid flow L_0 is unequally distributed over, for instance, three column sections. The first section receives the liquid volume $L_1 = (1 − δ)L_0/3$, the second section $L_2 = L_0/3$, and the third section $L_3 = (1 + δ)L_0/3$, all with the concentration x_T. Because the gas flow, which enters the bottom of the packing with concentration y_B, is ideally mixed, the material and component balances result in different concentrations for the liquid and gas flows leaving the packing. The liquid leaving the column is mixed to concentration x_B in the sump, and the gas leaving the top of the column is mixed to concentration y_T.

For the description of the influence of liquid redistributors in absorption columns, the purification of a flow of exhaust air of acetone with the aid of water as
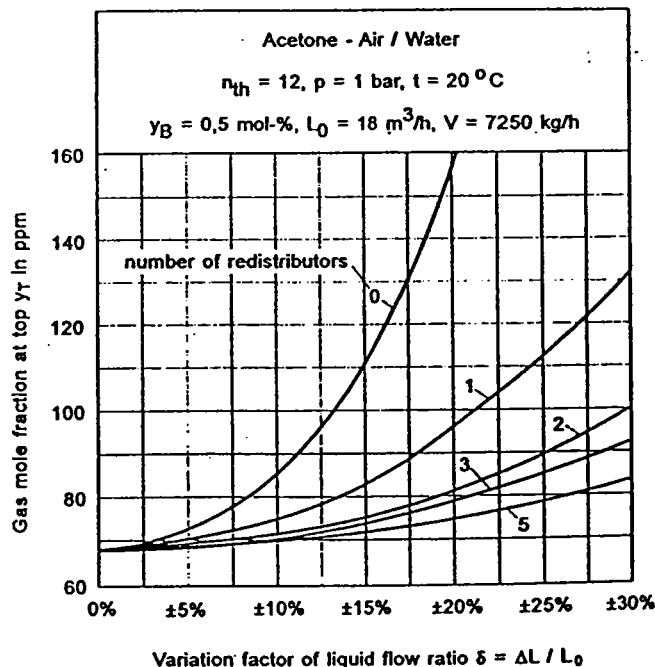

Figure 17. Off-gas concentration as a function of the variation factor $δ = ΔL/L_0$ with various liquid redistributors for an absorption system.

a solvent was calculated. A flowchart of an absorption/stripping column with redistributors is shown in Figure 16. The acetone/air mixture, which flows into the absorber with a concentration of 0.5 mol % of acetone, was to be reduced to below 70 ppm in the off-gas flow. A value of 1.5 times the minimum solvent rate was selected as the quantity of solvent. The simulation calculation resulted in the achievement of the specified conditions if 12 theoretical stages are provided. With a
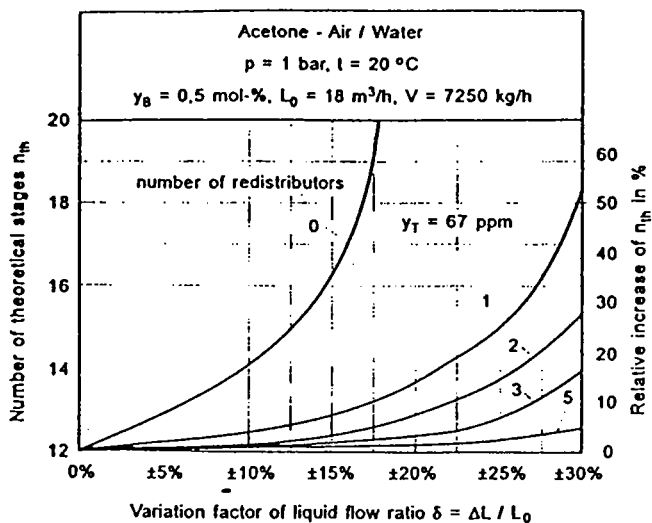
Figure 18. Number of theoretical stages as a function of the variation factor $\delta = \Delta L/L_0$ with various liquid redistributors for an absorption system.
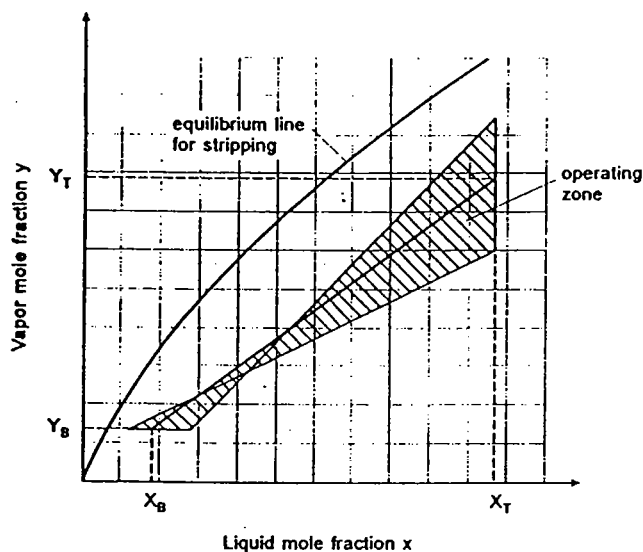

Figure 19. McCabe–Thiele diagram with L/V variations for a stripping column.

50 mm pall-ring made of metal, the HETP value was calculated to 1.1 m so that a bed height of 13 m is necessary.

The results of simulation are shown in Figure 17. The evaluation shows that up to a variation factor of 10% and without liquid redistribution 85 ppm can still be achieved in the off-gas flow. Only if three redistributors are used can an increase in the concentration over 70 ppm in the flow of off-gas be prevented by a variation factor of 10%. With a variation factor of 30%, the exit concentration in the off-gas flow rises to above 90 ppm even if three redistributors are used.

If the evaluation takes place under the aspect of the required off-gas concentration, Figure 18 follows. It can be seen that without liquid redistribution and a variation factor of 10%, 17% more theoretical stages are necessary than was determined by the simulation with a variation factor of 0%. With three redistributors the value is reduced to 1%. With three redistributors and 30% variation factor, 17% more theoretical stages are needed.

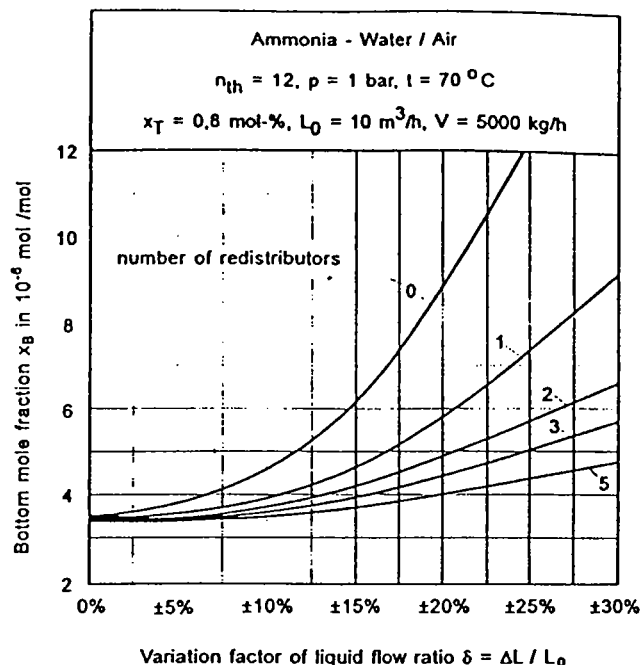Desorption columns react similarly to uneven irrigation of the packed bed by the liquid phase, *s* (Figure

Figure 20. Off-liquid concentration as a function of the variation factor $\delta = \Delta L/L_0$ with various liquid redistributors for a desorption system.
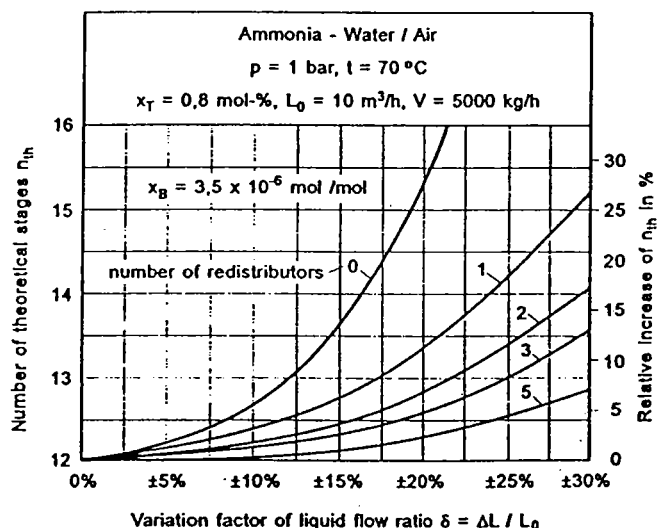

Figure 21. Number of theoretical stages as a function of the variation factor $\delta = \Delta L/L_0$ with various liquid redistributors for a desorption system.

19). The desorption of ammonia from water with humid air was calculated as an example. The wastewater flows to the column with an ammonia concentration of 0.8 mol % and was to be purified to below 4 ppm. A value of 1.6 times the minimum air flow rate was selected as the quantity of air. The simulation calculation resulted in 12 theoretical stages being sufficient to achieve the stated conditions. With a calculated HETP value of 0.6 m for a 50 mm pall-ring made of metal, a bed height of 7.2 m has to be installed.

It can be seen from Figures 20 and 21 that, despite a variation factor of 10% and without liquid redistribution, the liquid discharge concentration rises only to 4.6 ppm and that 5% more theoretical stages have to be installed to meet the required liquid discharge concentration. There is practically no increase in concentration in the liquid discharge with a variation factor of 10% when three redistributors are used. With one liquid

redistributor and a variation factor of 30%, the concentration in the liquid discharge rises only to about 9 ppm. To be able to meet the discharge concentration with one liquid redistributor, 27% more theoretical stages would have to be installed with a variation factor of 30%.

Conclusion

The relations described illustrate that uneven irrigation of a packed bed has a decisive influence on the behavior of mass-transfer columns. The distillation example calculated reacted more sensitively to liquid flow variation than the absorption and desorption examples. This is particularly the case if rectification columns are operating close to the minimum reflux ratio and high product purities have to be achieved. In this case the equilibrium curve approximates the operating line, and changes in the L/V material flow ratio have a disadvantageous effect on the mass-transfer efficiency of packings.

Numerous traditional packings have a greater maldistribution factor for the liquid phase than modern packings. This results in a larger variation of liquid flow in the traditional packings, which is why those should be filled to a lower height and why redistribution of the liquid should take place sooner in comparison to modern packings. On the basis of the experimental results by Stikkelman, an uneven irrigation of the packing is naturally given, i.e., always exists despite an even liquid distribution by a liquid distributor.[17]

The advantage of the simulation presented is in the fact that the considerations are possible for any multicomponent systems and feed conditions. The examples shown can be extended in the case of further conditions. For example, it was left unconsidered that the liquid phase can have less than an optimal initial distribution by the liquid distributor or redistributor. Any maldistribution of the liquid phase caused by a malfunction of liquid distributor also has a disadvantageous effect on the mass-transfer efficiency and can be included in the simulation algorithm presented. Maldistributions of the gas phase can also be taken into consideration by introducing a variation factor for the gas flow.

The most important value for such kinds of simulations is the knowledge of the liquid flow variation factor δ. Unfortunately, there is up to now no equation given that describes the liquid flow variation from influencing parameters such as the maldistribution factor M_f and type of packing. However, for example, in the case that a variation of liquid flow of 10−15% is assumed, the presented algorithm verifies if a mass-transfer system reacts sensitively or not to the liquid flow variation and if liquid redistributors are recommended. In this case, the evaluation can be used to determine the best possible packed heights, after which a liquid redistributor should be installed.

Nomenclature

L = molar liquid flow rate (mol/h)
L_i^{vol} = liquid volume rate of section i (m³/h)
L_{mean}^{vol} = mean liquid volume rate (m³/h)
M_f = maldistribution factor
n = number of sections
n_{th} = number of theoretical stages
p = pressure (bar)

r_D = reflux ratio
V = molar vapor flow rate (mol/h)
x = liquid mole fraction (mol/mol)
y = vapor mole fraction (mol/mol)
α = relative volatility
δ = variation factor

Indices

B = bottom
T = top
F = feed
D = distillate
E = entrance

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