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Substance and heat transfer in multistage systems

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Abstract:- In this paper simultaneously substance and heat transfer in the multistage system was investigated. Substance transfer between liquid and gas phases in a distillation packed column was considered. Substance and heat transfer rate were examined and substance transfer coefficients were determined. Efficiency of substance transfer were examined by height of transfer units and height of the equilibrium stages. Based on derived mathematical model the total height of transfer units and individual height of transfer units for the liquid and gas phases were determined. The examination was performed by computer simulation and experiments.

Keywords: Material and heat transfer, height of transfer unit, height of equilibrium plate, substance transfer coefficient, binary system, distillation column.

1 Introduction

Gas absorption is operation in which one or more soluble components of a gas mixture are dissolved in a liquid. The reverse operation called stripping or desorption is employed to transfer one or more volatile components from a liquid mixture into gas.

There are three type broad steps in the separation system design. Data on the vapor-liquid equilibrium relations for the system are needed to determine to quantity of liquid necessary to absorb the required amount of the soluble components from the gas, or the quantity necessary to strip the desired amount of the volatile components from the liquid. Data of the liquid and vapor capacity of equipment of the type being considered for use are needed to determine the necessary cross sectional area and size of the equipment. Equilibrium data and material balances are used in combination with fundamental relations perculiar to the absorption-stripping process to calculate the number of equilibrium stages required for the separation desired.

When the composition of a fluid mixture varies from point to another, each component has a tendency to flow in the direction that will reduce the local differences in concentration. If the bulk fluid is either stationary or in laminar flow in a direction normal to the concentration gradient, the material transfer reducing the concentration difference occurs by a process of molecular diffusion. This mechanism, characterized by random movement of individual molecules, contrasts with the bulk transport by eddies which occurs in a turbulent fluid.

The theory of molecular diffusion has been the subject of extensive investigation because of its close relationship to the kinetic theory of gases. Detailed reviews are available have provided a valuable critical comparison of the various correlations which have been presented for the prediction of diffusivities in gases and liquids, including electrolytes and nonelectrolytes under a variety of conditions.

Different theoretical approaches to the description of diffusion in liquids have been made, depending upon whether the systems are electrolyte or nonelectrolitic. The available prediction procedures must therefore be divided into those suitable for nonelectrolytes and those suitable for electrolytes. Most studies have been denoted to the estimation of diffusivities in very dilute solution, although some progress has been made towards allowance for the substantial variations that occur with increasing concentration of the diffusing solute.

Material transfer phenomena where diffusion occurs through the liquid and gas interface has been studied in distillation columns. A different approaches to the modelling of the packed column, particularly applicable to binary and multicomponent distillation, were developed and applied in full-scale tests in the papers[1]-[10]. They put forward the concept of substance transfer section and defined vaporization efficiencies to compensate the deviation of each section from an ideal stage.

Ruckenstein [7] solved the transport equations in the liquid phase without using the two-film model. This was a simultaneous treatment of heat and mass transfer, but confined to the liquid phase. Dutkai-Rukenstein [8] abandoned the idea of thermal distillation and modelled the packed column using only overall mass transfer coefficients. The mass transfer coefficients and mass transfer efficiency were investigated by Savkovic-Stevanovic et.al.[9].

Many types of mass transfer operations are carried out in either continuous or stage wise columns. These columns are usually packed with Raschig rings, Pall rings, Berl saddles, Lessing rings, or other types of packing, to promote intimate contact between the two phases. Continuous contact is therefore maintained between the two countercurrent streams throughout the equipment, necessitating a differential type of treatment.

The preferred method of design involves determination of the number of transfer unit -NTU necessary to achieve the desired separation. Evaluation of the NTU requires preliminary construction of the equilibrium curve and the operating lines of the separation process.

Industrial separation processes involving acetic acid continue to be of considerable interest. Mass transfer between the liquid and vapor phase of the systems with association in a distillation column was studied [9]-[10].

In this paper the substance transfer coefficients and height of transfer units are determined, using effective diffusion coefficients for acetic acid -xylene. Method for substance transfer coefficients determination was derived by fitting operation curve and vapor-liquid equilibrium curve to the experimental data.

2 Simultaneous material and heat transfer in the staged gas-liquid system

Let assume, in a distillation packed column concentration gradient is changed only in z direction elements Δz₁, Δz₂, Δz₃, …., Δzₙ counting from the top to the bottom (Fig.1). Phase equilibrium between vapor and liquid phases for component i, at the same point is:

$$f_i^V = E_i f_i^L \qquad (1)$$

where E_i vaporization efficiency at the point. Analogously, for any section Δz_j will be:

$$f_{ji}^V = E_{ji} f_{ji}^L \qquad (2)$$

where E_{ji} so called vaporization efficiency for any element Δz_{ji} . For equilibrium conditions when vapor and liquid phases pressure and temperatures are equal then,

$$y_{ji} = E_{ji} x_{ji} \gamma_{ji} (f_{ji}^V / f_{ji}^L) \qquad (3)$$

If the both vapor and liquid phases can be considered as ideal solution, then can be denoted with $K_{ji} = f_{ji}^V / f_{ji}^L$.

Material balance for steady state conditions:

Material balance for the vapor phase of the element Δz_{ji} is:

$$G_{j+1} - G_j + \int_{z_j}^{z_{j=1}} \sum_{i-1}^{M} N_i dz = 0 \qquad (4)$$

and for the liquid phase,

$$L_{j-1} - L_j - \int_{z_j}^{z_{j=1}} \sum_{i-1}^{M} N_i dz = 0 \qquad (5)$$

Energy balance for steady state conditions:

Energy balance for the vapor phase of the element Δz_{ji} is,

$$G_{j+1} H_{j+1} - G_j H_j + \int_{z_j}^{z_{j=1}} \sum_{i-1}^{M} N_i dz - \int_{z_j}^{z_{j=1}} Q_G dz = 0 \qquad (6)$$

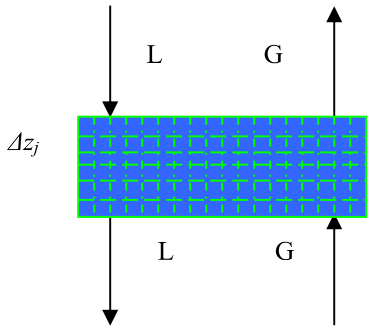


Fig.1 Substance transfer in the gas-liquid system in packed bed

Energy balance for the liquid phase of the element Δz_{ji} is,

$$L_{j-1} h_{j-1} - L_j h_j - \int_{z_j}^{z_{j=1}} \sum_{i-1}^{M} N_i dz + \int_{z_j}^{z_{j+1}} Q_G dz = 0 \qquad (7)$$

Energy balance for the vapor phase which including vapor phase, interphase and vapor film and reaction heat of the element Δz_{ji} is:

$$G_{j+1} H_{j+1} - G_j H_j + \int_{z_j}^{z_{j=1}} \sum_{i-1}^{M} N_i H_i dz - \int_{z}^{z_{j-1}} Q_G dz = \frac{d(U_{Gj} H_j)}{dt} \qquad (8)$$

Energy balance for the liquid phase which including liquid phase, liquid film, interphase and vapor film and reaction heat of the element Δz_j is,

$$L_{j-1} h_{j-1} - L_j h_j + \int_{z_j}^{z_{j=1}} \sum_{i-1}^{M} N_i h_i dz + \int_{z}^{z_{j-1}} Q_G dz = \frac{d(U_{Lj} h_j)}{dt} \qquad (9)$$

3 Substance transfer rate

Substance transfer driving force between gas and liquid phase can be defined as:

$$m_{i,G}^t = -m_{i,L}^t \qquad (10)$$

$$\frac{dc_{i,L}}{dt} = k_L a (c_{i,L} - c_{i,L/G}) (=) k_L a (c_{i,L} - c_{i,L}^*) \qquad (11)$$

$$\frac{dc_{i,G}}{dt} = k_G a(c_{i,G} - c_{i,G/L})(=)k_G a(c_{i,G} - c^*_{i,G}) \qquad (12)$$

where $c_{i,L/G}$ and $c_{i,G/L}$ mean concentration in the phases borders and can be approximated with concentration which in equilibrium with the existing average concentration of the liquid phase.

The relationship between the overall and individual substance transfer coefficients is given by the following relation:

$$\frac{1}{k_{OG}} = \frac{1}{k_G} + \frac{m'}{k_L} \qquad (13)$$

$$\frac{1}{k_{OL}} = \frac{1}{k_L} + \frac{1}{m'k_G} \qquad (14)$$

Substance rate between vapor and liquid phase can be defined as follow:

$$dN_i = Gdy_i = Ldx_i \qquad (15)$$

$$dN_i = k_G a(y_i - y_{iG/L})Sdz = k_L a(x_{iL/G} - x_i)Sdz \qquad (16)$$

Since, interphase $y_{G/L}$ and $x_{L/G}$ uncertain for given location it is more convention to use over all component transfer coefficient as following:

$$dN = k_{OG}a(y - y^*)Sdz = k_L a(x^* - x)Sdz \qquad (17)$$

and combining eqs.(15)-(16) is obtained:

$$\int_0^z dz = G\int_{y1}^{y2} \frac{dy}{k_{OG}aS(y_i - y_i^*)} = L\int_{x1}^{x2} \frac{dx}{k_{OL}aS(x_i^* - x_i)} \qquad (18)$$

By integration between top and bottom of the column is determine total column height Z.

$$Z = \frac{G}{k_{OG}aS}\int_{y_B}^{y_D} \frac{dy}{y_i - y_i^*} = \frac{L}{k_{OL}aS}\int_{x_B}^{x_D} \frac{dx}{x_i^* - x_i} \qquad (19)$$

Integrals in eq.(19) defines NTU –Number of Transfer Unit and Z/NTU is equal HTU-Height of Transfer Unit.

Relationships between overall and individual height of transfer unit are given by eqs.(20)-(21).

$$(HTU)_{OG} = (HTU)_G + \frac{m'G}{L}(HTU)_L \qquad (20)$$

$$(HTU)_{OL} = (HTU)_L + \frac{L}{m'G}(HTU)_G \qquad (21)$$

where *m'G/L* and *L/m'G, (m'=K)* are absorption and desorbtion factors.

4 Equilibrium stage method and transfer unit method comparison

The equilibrium stage method for characterization of the difficulty of separation provides an adequate measure of separability for distinct substance transfer elements in a counter current columns in a highly efficient plate column.

On the other hand, the transfer unit method is preferred when vapor and liquid are in continuous contact, in packed columns and plate columns which have an efficiency will below 1. The equilibrium stage method is used often in practice, because graphical determination of the number of equilibrium steps on the y-x diagram is much easer than integration with respect to the driving force.

The equilibrium curve and the operating line can almost always be linear in narrow concentration ranges. The following relationship is then obtained

$$(NTU)_{OG} = n_t \frac{\ln \frac{m'}{(L/G)}}{\frac{m'}{(L/G)} - 1} \qquad (22)$$

In the special case when $m'/(L/G) \rightarrow 1$ $(NTU)_{OG} \rightarrow n_t$. The height equivalent of theoretical plate -HETP can be defined.

$$HETP = Z/n_t. \qquad (23)$$

5 Columns with packing

Packed columns are often used for distillation and gas absorption. In this paper only distillation will be considered.

In the gas liquid contact in a packed bed column, the liquid flows down the column over the packing surface and the gas or vapor, counter–currently, up the column. In some gas-absorption columns co-current flow is used. The performance of a packed is very dependent on the maintenance of good liquid and gas distribution throughout the packed bed, and this is an important consideration in packed column design.

The choice between a plate or packed column for a particular application can only be made with complete assurance by costing each design. However, this will not always be worthwhile, necessary, and the choice can usually be made, on the basis of experience by considering main advantages and disadvantages of each type.

Plate columns can be designed to handle a wider range of liquid and gas flow rates than packed columns. The packed columns are not suitable for very low liquid rates. The efficiency of plate can be predicted with more certainty than the equivalent term for packing HTU or HETP. Plate columns can be designed with more assurance than packed columns. There is always some doubt that good liquid distribution can be maintained throughout a packed column under all operating conditions, particularly in large columns. It easier to make provision for the withdrawal of side streams from plate columns.

If the liquid causes fouling, or contains solids, it is easier to make provision for cleaning a plate column, man ways can be installed on the plates. With small diameter columns it may be cheaper to use packing and replace the packing when it becomes fouled. For corrosive liquids a packed column will usually be cheaper than the equivalent plate column. The liquid holdup is appreciably lower in a packed column than a plate column. This can be important when the inventory of toxic or flammable liquids needs to be kept as small as possible for safety reasons. The packed column are more suitable for handling foaming systems. The pressure drop per equilibrium stage can be lower for packing than plates, and packing should be considered for vacuum columns. Packing should always be considered for small diameter columns, say less than 0.6 m, where plates would be difficult to install, and expensive.

The design of a packed column will involve the following steps: select the type and size of packing, determine the column height required for the specified separation, determine the column diameter, capacity, to handle the liquid and vapor flow rates, select and design the column internal features packing support, liquid distributor, redistributors.

The principal requirements of a packing are that it should: provide a large surface area (a high interfacial area between the gas and liquid), have an open structure, low resistance to gas flow, promote uniform liquid distribution on the packing surface, promote uniform vapor gas flow across the column cross section.

With continuous contactors, stage wise contactors utilize intermitten contact between the phases. The stages often take the form of horizontal plates of varied design, arranged vertically above each other in the column.

The two phases enter a stage from opposite directions in countercurrent flow, mix together to facilitate transfer and then separate and leave the stage. When two phases leave in a state of equilibrium the stage is said to be an ideal or theoretical one. This concept has been extended to packed columns by defining the height, of packing, equivalent to a theoretical stage such that the streams leaving this section are in equilibrium.

The number of theoretical stages to which this packed column is equivalent is given by the broken-line stepwise construction between the operating and equilibrium curves.

6 Experimental section

The experimental setup has shown in Fig.1. The experimental data were obtained in a ordinary glass distillation column with a packing height of 1000 mm and a diameter of 33 mm. The average diameter of packing particle Rashig rings 0.4 x 0.4 cm. The bed void fraction was 0.60, and the specific surface of packing 0.01609 cm^2/cm^3.

The batch distillation unit is controlled by the acquisition block. Normatron was connected for reflux flow rate control over microprocessor. Sensor for pressure drop and temperature sensors Pt-100 were used.

The experiments were carried out by batchwise distillation operation and were semi-continuous in adiabatic conditions. The compositions of the distillate and the bottom product were determined by refractometer.

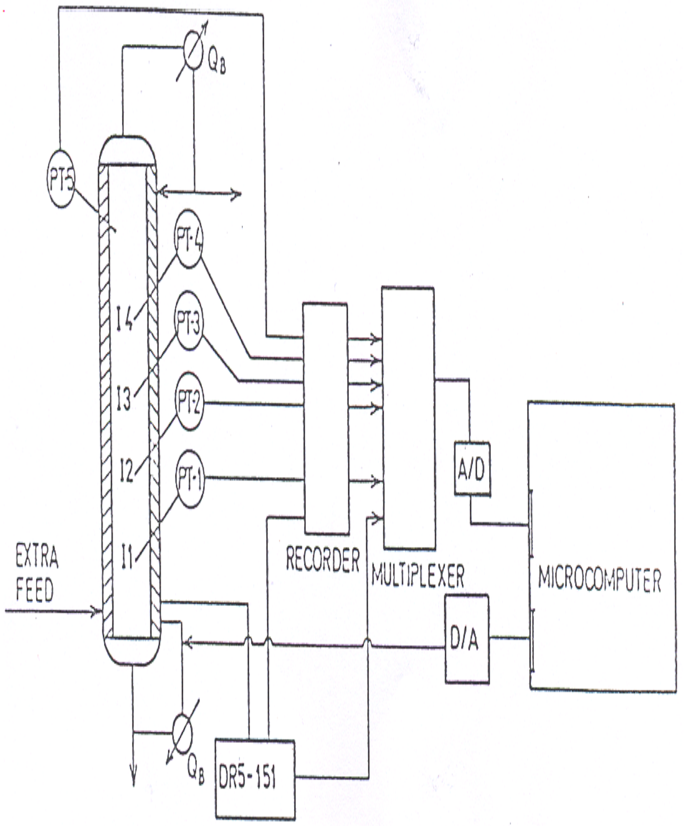


Fig.1(a) Scheme of the laboratory distillation column with temperature and pressure acquisition system

The distillation was carried out at the atmospheric pressure. The charge consisted of 90mol% acetic acid and 10mol% p-xylene. Holdup on each stage was 0.2 mole, for total condenser and reboiler 2 moles. The distillation was performed at different flow rates through the column. The hydrodynamic characteristics with Rashig rings were determined and shown in Fig.2. The optimal flow rate through the column was established at 0.56 m/s.

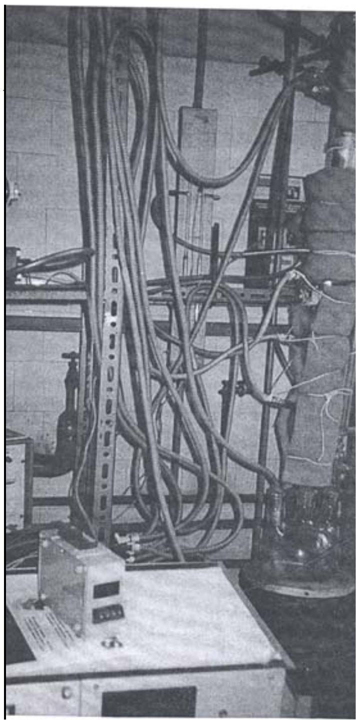
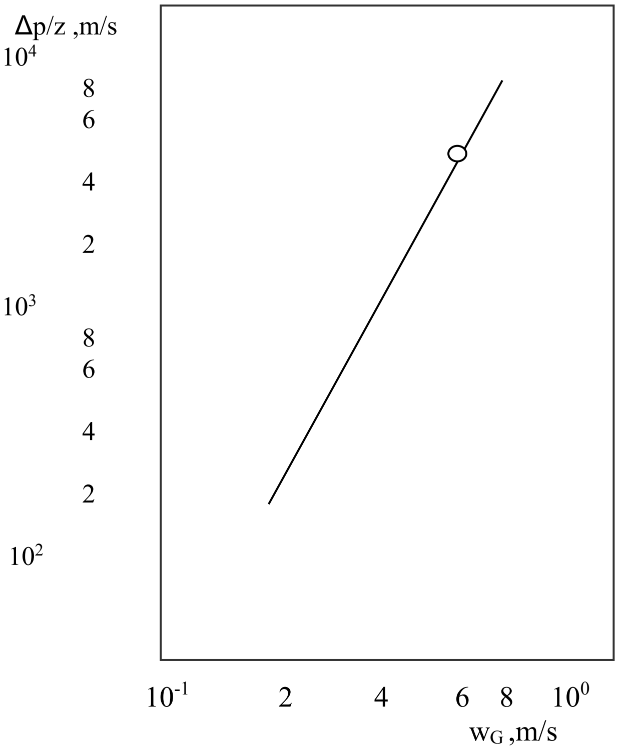


Fig.1 (b) Experimental set up



Fig

Fig. 3 shows dependence distillate quality of vapor velocity, through the column under infinite reflux ratio R=∞, and optimal hydrodynamic region.

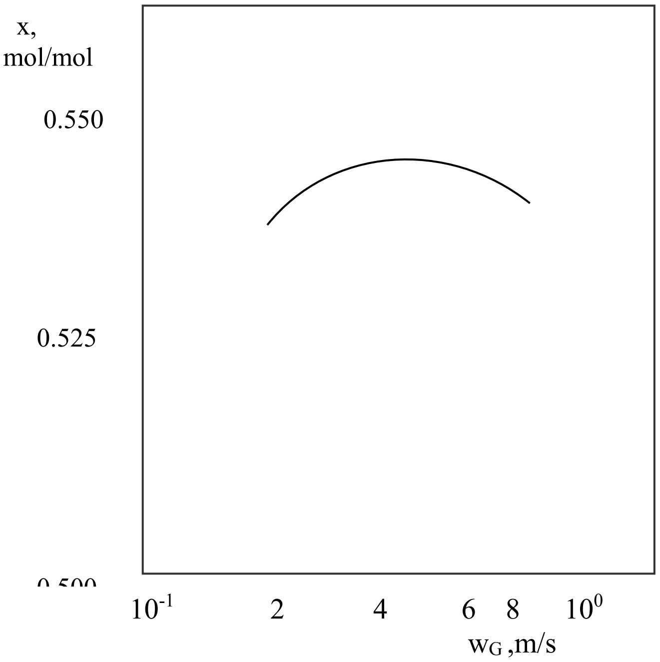


Fig.3 Top xylene composition versus gas velocity for the initial charge mixture 10%mol xylene and 90mol% acetic acid (R=∞)

The integrals in equation (19) are evaluated by numerical integration. Information for this procedure is obtained from the equilibrium curve-operating line plot on x,y coordinates

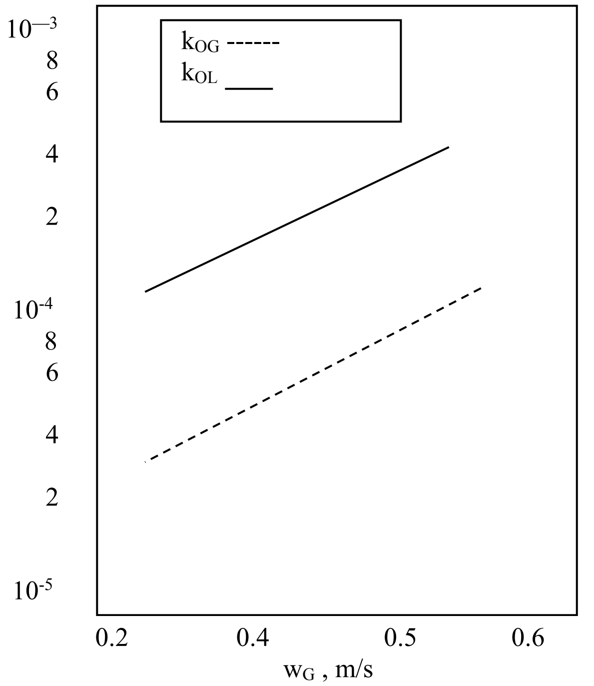
It is often inconvenient to determine interfacial compositions (y*, x*) corresponding to each point on the operating line, so overall NTU values are frequently determined in preference to the individual ones.

The evaluation could be performed by numerical integration. This would require equations for the equilibrium and operating curves. Numerical integration was realized using Simpon's rule. An algorithm was derived and program module was developed in Fortran programming language.

Fig.4 shows the substance transfer diffusion coefficients vs. vapor velocity.

7 Conclusion

The substance and heat transfer rate and height of transfer units are examined, using effective diffusion coefficients. The substance transfer diffusion coefficients were determined. The substance transfer coefficients determination method was derived by fitting working curve and vapor-liquid equilibrium curve to the experimental data.



Fi [illegible] etric substance transfer coefficients for acetic acid –xylene in the both liquid and gas phases vs. flow rate (R=4)

Notation

c-concentration, mol/ cm^3
E-vaporization efficiency
f-fugacity
G-gas flow rate, mol/s
g-gravitational acceleration, cm/s^2
H-enthalpy of the liquid phase, J/mol
h- enthalpy of the vapor phase, J/mol
HETP-Height of equivalent theoretical plate, cm
HTU- Height of transfer unit, cm
$(HTU)_L$, $(HTU)_G$- height of transfer unit over the liquid and gas phase, respectively, cm
K- equilibrium constant
k_L k_G, individual transfer coefficient for the liquid and the gas phase, respectively, mol/cm^3 s
k_{OG}, k_{OL}, overall substance transfer coefficient,mol/cm^3 s
L-liquid flow rate, mol/s
M-total number of component
m'- equilibrium phase ratio($= \alpha /[1+(\alpha-1)\overline{x}]^2$)
m^t -amount of substance transfer
N- total number of molecules
n-constant
n_t.-number of theoretical plate
NTU-number of transfer unit
Q-heat
R-reflux ratio
S-radial column surface, cm^2
T-temperature, K
U-hold up, mol
y- vapor composition
y^*- equilibrium composition
x-liquid phase composition
V-volume,cm^3
w-gas phase velocity, cm/s
z- column height, cm

Subscript
G-gas
L-liquid
OG-overall liquid phase
OL-overall gas phase

Greek Symbol
α -relative volatility

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