IJCPE
Iraqi Journal of Chemical and Petroleum Engineering

Iraqi Journal of Chemical and Petroleum Engineering
Vol.12 No.1 (March 2011)
ISSN: 1997-4884

University of Baghdad
College of Engineering

ESTIMATION OF MASS TRANSFER COEFFICIENTS IN A PACKED DISTILLATION COLUMN USING BATCH MODE

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ABSTRACT

This research adopts the estimation of mass transfer coefficient in batch packed bed distillation column as function of physical properties, liquid to vapour molar rates ratio (L / V), relative volatility (α), ratio of vapour and liquid diffusivities (D_V / D_L), ratio of vapour and liquid densities (ρ_V / ρ_L), ratio of vapour and liquid viscosities (μ_V/ μ_L).
The experiments are done using binary systems, (Ethanol Water), (Methanol Water), (Methanol Ethanol), (Benzene Hexane), (Benzene Toluene). Statistical program (multiple regression analysis) is used for estimating the overall mass transfer coefficient of vapour and liquid phases (K_{OV} and K_{OL}) in a correlation which represented the data fairly well.

$$K_{OV} = 3.3 * 10^{-10}\, \alpha^{-0.7} (D_V / D_L)^{0.65} (L / V)^{3.5} (\rho_V / \rho_L)^{1.25} (\mu_V / \mu_L)^{-5.0}$$

$$K_{OL} = 2.8 * 10^{-6}\, \alpha^{-0.95} (D_V / D_L)^{0.03} (L / V)^{1.15} (\rho_V / \rho_L)^{0.077} (\mu_V / \mu_L)^{-0.9}$$

In this research a method where the resistances to mass transfer in both phases are accounted for separately through the use of HTU-NTU model for each phase

$$Z=HTU_{OV}.NTU_{OV} \quad \text{and} \quad Z=HTU_{OL}.NTU_{OL}$$

Results show that both overall vapour and liquid mass transfer coefficient are increased with liquid to vapour molar rates ratio, vapour to liquid diffusivities ratio and vapor to liquid densities ratio, but decreased with increasing the relative volatility and vapour to liquid viscosities ratio.

Key words: mass transfer coefficient; distillation; packed column.

INTRODUCTION

Batch distillation process is an important separation technique especially used in the fine specialty: pharmaceutical, biochemical and food industries. Because the demand and the uncertainty in specifications for high-value-added and low volume specialty chemicals have increased recently, the use of batch distillation is becoming increasingly popular. Instead of using many continuous columns in series, multiple products can be obtained from a single batch distillation column during a single batch run. Moreover, batch distillation processes can easily handle variations both in the product specifications and in the feed composition. This flexibility of batch distillation processes provides the ability to cope with a market characterized by short product life times and strict specification requirements. Batch operation should be considered when the quantity to be distilled is small; when it is produced at irregular intervals; when a range of products has to be produced; or when the feed composition is likely to vary considerably.

A batch column is like the top half of a continuous column – it has a rectifying section only. A portion is returned to the column as reflux and the remainder is removed as distillate or top product [1].

Interfacial mass transfer that is characterized by the partial volumetric mass transfer coefficients K_La and K_Va is a common physical background of absorption and distillation. The knowledge of both coefficients is hence essential in the design of packed absorption and distillation columns.

For every packing type correlations are used to describe the dependence of K_La and K_Va on physical properties and conditions prevalent in an experimental apparatus. Such correlations are based only on the absorption mass transfer data, for it is not possible to determine K_La and K_Va separately by distillation experiments. But as the physical properties (especially temperature) in absorption and distillation column significantly differ, the correlations suitable for both processes should fit their influence accurately in a sufficiently wide range. Up to now this requirement has not been fulfilled, so the use of the “absorption correlations” in the distillation column design is risky. In order to create a common mass transfer data correlation, the knowledge of K_La and K_Va not only under absorption, but also under distillation conditions is necessary [2].

Absorption and distillation have the same physical background –interfacial mass transfer between gas and liquid phase which is characterized by mass transfer coefficient in liquid- K_La and vapour/gas- K_Va side. An effort to calculate the height equivalent to a theoretical plate (HETP) using absorption mass transfer coefficients recalculated to the conditions in distillation column has mostly led to large deviations from the experimental data (often higher than 50%). The discrepancy can be ascribed partially to large extrapolation in temperature used in the data transfer from absorption to distillation. However, the measurements of mass transfer coefficient directly in distillation columns are rare. In distillation namely, it is not possible to measure individual mass transfer coefficients by using a proper distillation system in which the overall resistance to the interfacial mass transfer is mainly due to one of the phases and to study behavior of the individual coefficients separately as it is possible in absorption [3].

Kayihan, et.al [4], studied simultaneous heat and mass transfer distillation theoretically in a wetted wall column using methanol water binary system for one atmosphere distillation. Confirmation of the liquid phase saturation was made through a comparison of the experimentally measured liquid temperatures with the calculated bubble point temperature. The authors deduced that all of the resistance to mass transfer was in the vapour phase. It was concluded that interphase diffusion is responsible for mass transfer in distillation and that there is no additional evaporation within the liquid phase. Honorat, et.al [5] examined the simultaneous heat and mass transfer process in binery distillation by combining enthalpy and material balances around the liquid and vapour phases which makes it impossible to obtain the mass transfer coefficient from distillation experiments by measuring compositions and temperatures as functions of column height. Their experiments carried out in a packed bed column at total reflux for the toluene-trichloroethylene system. Krishnamurthy et.al [6] used a non-equilibrium stage model of countercurrent separation processes to model a packed distillation column and a packed absorber. A key feature of the model is that the component material and energy balance relations for each phase are solved simultaneously with the mass and energy transfer rate equations and interface equilibrium equations. Computations of quantities such as HETP and HTU are completely avoided. The terminal stream composition, flow rates, and temperature profiles over the packed depth predicted by the model are compared with results of field tests. They refer to that resistances to mass transfer and heat transfer in both phases are accounted for separately through the use of rate equations for each phase. Lee et al [7], simulated a multi-component packed distillation [(methanol(1)/ethanol(2)/water(3)] using a rate-based model in which the so called MERQ (material balance, energy balance, rate equilibrium) equations in any segment of the packing are solved with an equation tearing technique. The simulation results are compared with the experimental results obtained from a 0.2 m diameter pilot-scale packed column using the published correlations to evaluate their applicability. Lee et.al,[8] obtained concentration profile of the acetone-methanol-2-propopanol system experimentally using a batch distillation column packed with 1 cm pall rings which compared with theoretical profiles. They performed experiments under 1 atm, total reflux condition and used matrix model for calculating theoretical concentration profiles. They estimated mass transfer coefficient and the height of transfer unit by empirical equation of Onda et al [9] and Billet [10]. Their calculated profiles were in reasonable agreement with the experimental profiles.

The objective of this research is determining the overall mass transfer coefficient based on the vapour and liquid phase from temperature composition experimental data using HTU-NTU model in a batch packed distillation column . Effect of verifying physical properties (Relative volatility (α), Ratio of vapour and liquid densities (ρ_V/ρ_L), Ratio of vapour and liquid viscosities (μ_V / μ_L), Ratio of vapour to liquid diffusivities (D_V / D_L), Vapour flow rates (V) and Liquid flow rates (L) under various reflux ratio values. Binary system is used for all the experiments (ethanol-water, methanol-water, ethanol-methanol, benzene-hexane and benzene-toluene. Multiple regression model (Statistical program) used for estimating overall mass transfer coefficient of vapour and liquid phases.

EXPERIMENTAL

A column of 0.5-m diameter and 1.5 m high was used for the distillation column as shown in Fig. (1). The column was filled with 10 mm of glass Raschig rings. The unit comprised of:

1. Reboiler system (Electrically

Heated):

Total working capacity (vessel+heater): 28 litters.

1.1- Spherical vessel: working capacity (13 litters).

1.2- Heat exchanger: electrical immersion type:

Rating at 240 volt, 50 HZ, single phase 6KW maximum.

2. Reflux flow meter (RI.1A): variable area type range (0-7.5) litters/min.

3. Product distillate flow meter (RI.1B.): variable area type range (0-7.5) litters/min.

4. Product distillate flow meter (RI.1C.): graduate tube calibrated in milliliters.
5. Cooling water flow meter (FI.2.): variable area type, range (0-15) litters/min.
6. Water pressure indicator: Bourdan type, range (0-4) bar range.
7. Column temperature indicator (TI.1,2,3) , locally mounted, mercury in glass thermometer range (20-120oC).
8. Reboiler temperature indicator (TI.4) remote reading filled system, dial thermometer, range (0-50oC)
9. Cooling water temperature indicators (TI.5, TI.6) remote reading dial thermometer, range (0-50oC).

10. Electrical Heated Version:
HC.4 Heater control: thyristor phase angle.
Power controller
I.4: ammeter moving iron type.
Range (0-40) amps
E.4: Voltammeter moving iron
Type (0-240 V).

The reboiler vessel is charged with chemical mixture (0.5 mole fraction) through the charge-port and valve V.6 and evaporated in the thermosyphone reboiler. The temperature of the boiling liquid is obtained from the temperature indicator TI.4.

Vapours from the reboiler enter at the base of the distillation column which is packed with glass Raschig rings. Liquid and vapour temperatures across each packed section are given by temperature indicators TI.1 to TI.3 inclusive. At first, the column was set to operate at total reflux condition,

Allowing a period of fifteen minutes for the equipment to maintain thermal equilibrium with the surroundings. Vapors leaving the top of the column are totally condensed to liquid distillate in the overhead condenser and pass to
the reflux control flow meters RI.1A and RI.1B. RI.1A indicates the quantity of distillate returned to the column as reflux, while RI.1B indicates the quantity of distillate removed as product. The desired reflux ratio is given by the ratio of the rotameter (RI.1A and RI.1B) scale reading which present the reflux ratio. A graduated cylinder RI.1C positioned in the product distillate line below the product cooler enables the volumetric flow rate to be measured. Temperature of the reboiler (TI.4) and top temperature (TI.1) are read experimentally and from temperature/equilibrium data, compositions are calculated. Overall material balance was applied for measuring distillate mole fraction.

The Differential Height Technique

For a given packed distillation column, overall liquid mass transfer coefficients (K_{OV}, K_{OL}) are calculated according to the HTU-NTU model.

$$Z=HTU.NTU \qquad (1)$$

According to Eq. (1) the overall vapour mass transfer coefficient obtained from HTU_{OV} (overall height of mass transfer units) and number of overall vapour phase transfer units (NTU_{OV}) which are defined by the concentrations of the boiler and top of the column(at packing height 1.5 m), dependent on the separation task and physicalproperties.

$$Z=HTU_{OV}.NTU_{OV} \qquad (2)$$

$$Z=HTU_{OL}.NTU_{OL} \qquad (3)$$

$$Z=\frac{V/S}{K_{OV}A}\int\frac{dY}{Y-Y^*} \qquad (4)$$

$$Y^*=\frac{\alpha.X}{1+(\alpha-1)X} \qquad (5)$$

The mount of distillate and distillation component is computed from material balance on the column:

$$S_O-S_1=D \qquad (6)$$

$$S_OX_{SO}-S_1X_{S1}=DX_D \qquad (7)$$

S_1: amount of mixture in the reboiler at any run, which is computed from Rayliegh equation:

$$\ln\frac{S_O}{S_1}=\frac{1}{\alpha-1}\ln\left[\frac{X_{S1}(1-X_{SO})}{X_{SO}(1-X_{S1})}\right]+\ln\left[\frac{(1-X_{SO})}{(1-X_{S1})}\right] \qquad (8)$$

Temperature - composition curve [11] is used for measuring mole fractions. The same procedure is applied for overall liquid phase mass transfer coefficient calculation:

$$Z=\frac{L/S}{K_{OL}A}\int\frac{dX}{X^*-X} \qquad (9)$$

$$X^*=\frac{Y}{\alpha+(1-\alpha)Y} \qquad (10)$$

Set of experiments are carried out under various reflux ratio values which calculated from minimum reflux ratio, the last is measured from vapour liquid equilibrium curve, then the reflux ratio used for the experiments as (1.1, 1.3 and 1.5) of the minimum reflux ratio. The physical properties are estimated from the references [1, 12].

RESULTS AND DISCUSSION

Overall mass transfer coefficient for vapour and liquid phases are evaluated from HTU-NTU model as function of physical properties for vapour and liquid phases (viscosities and densities), transport properties (relative volatility and diffusivity) and vapour and liquid molar rates under various reflux ratio values.

It was concluded that interphase diffusion is responsible for mass transfer in distillation and that there is no additional evaporation within the liquid phase caused by heat transfer from the vapour phase as proposed by some previous investigators. Honart and Orville[5] confirmed the same in a distillation packed column, i.e., the zero resistance to mass transfer in the liquid phase.

An assumption that all of the resistance to mass transfer is in the vapour phase has been used in many application of simultaneous heat and mass transfer model for multicomponent distillation. For example, Pelkonen et.al [13] used the model for the evaluation of the vapour phase mass transfer coefficient in a wetted wall column in the benzene-toluene-ethylbenzene ternary system and assumed a total resistance to mass transfer in the vapour phase. In the present work, resistances to mass transfer in both phases are accounted for separately through the use of HTU-NTU model for each phase. The mathematical expression for mass transfer in distillation column using binary system can be expressed as:

$$\begin{pmatrix} K_{ov} \\ K_{oL} \end{pmatrix} = f\left[\alpha, D_{AB}, L, V, \mu_V, \mu_L, \rho_V, \rho_L\right] \quad (11)$$

$$K_{ov} = C_1 \alpha^{a1} \left(\frac{D_V}{D_L}\right)^{a2} \left(\frac{L}{V}\right)^{a3} \left(\frac{\rho_V}{\rho_L - \rho_V}\right)^{a4} \left(\frac{\mu_V}{\mu_L}\right)^{a5} \quad (12)$$

$$K_{oL} = C_2 \alpha^{a1} \left(\frac{D_V}{D_L}\right)^{a2} \left(\frac{L}{V}\right)^{a3} \left(\frac{\rho_V}{\rho_L - \rho_V}\right)^{a4} \left(\frac{\mu_V}{\mu_L}\right)^{a5} \quad (13)$$

Where: α Relative Volatility.

D_V and D_L: Vapour and Liquid Diffusivity (m^2/sec).

$\left[\frac{L}{V}\right]$: Liquid and Vapour molar rates ratio.

$\frac{\rho_V}{\rho_L - \rho_V}$: Liquid and Vapour densities.

$\frac{\mu_V}{\mu_L}$: Liquid and Vapour viscosities.

C_1, C_2: Constants which have the unit of mass transfer coefficient.

a_1-a_5: Indices.

Estimation of mass transfer coefficient is done using multiple regression model (statistics program), which gives the correlation factors for both overall liquid and vapour phase transfer coefficients:

$$K_{OV} = 3.3 * 10^{-10}\ \alpha^{-0.7}\ (D_V / D_L)^{0.65}\ (L / V)^{3.5}\ (Pv / \rho L)^{1.25}\ (\mu V / \mu L)^{-5.0}$$

$$K_{OL} = 2.8 * 10^{-6}\ \alpha\text{-}^{0.95}\ (D_V / D_L)^{0.03}\ (L / V)^{1.15}\ (\rho_V / \rho_L)^{0.077}\ (\mu_V / \mu_L)^{-0.9}$$

The experimental data and the detailed analysis may be found elsewhere [14].

Effect of Liquid Molar Rate (L) on K_{OV} and K_{OL}:

Most of the distillation data used in the correlation were reported under total reflux conditions, thus a separation of the liquid and vapour rates effect could not be studied from the previous available data.

Overall mass transfer coefficient based on vapour and liquid phase (K_{OV}) and (K_{OL})

increased with increasing liquid molar rate, since as the liquid molar rate increased, the liquid distribution in the bed is improved and mass transfer coefficient increased, because a good liquid distribution enhanced the contact of the gas and the liquid in the packed section. The effective transfer area should be proportional to the liquid rate since increased liquid velocity would provide a more effective renewal of the liquid film.

Effect of Vapour Molar Rate (V) on K_{OV} and K_{OL}:

Both (K_{OV}) and (K_{OL}) decreased with increasing the vapour rate (V). This may be due to that when increasing the vapour molar rate, the thickness of the concentration boundary layer on the vapour side at the vapour-liquid interface becomes thinner, this result in a lower mass transfer coefficient which is inversely proportional to the thickness of the concentration boundary layer.

Vapour and Liquid Viscosities Ratio Effect:

The overall vapour and liquid mass transfer coefficient (K_{OV}) and (K_{OL}) decreased with increasing the ratio of vapour to liquid viscosity , i.e when the vapour viscosities increased the coefficients decreased, this can be attributed both to the reduction in mass diffusivity. Also when liquid viscosity increased the coefficients decreased, this effect may be due to the maximum wettability obtained and ripples are formed when liquid viscosity increased.

Effect of Ratio of Vapour and Liquid Densities:

Overall mass transfer coefficient based on vapour a

The overall vapour and liquid mass transfer coefficient (K_{OV}) and (K_{OL}) increase with increasing densities ratio as expected.
These results tend to the increasing of vapour and liquid masses. As the mass of the vapour increased, it may cause ripples to form on the liquid film (especially at the end effects) and thus increase the mass transfer coefficient. Also when the mass of the liquid increased, the column holdup increased such that the empty spaces within the bed close up and the liquid flows downwards as a continuous phase. The gas phase then rose up through the liquid layer in the form of bubbles, these all tend to decrease the mass transfer coefficient.

Effect of Ratio of Vapour and Liquid Diffusivities:

Vapour and liquid mass transfer coefficient increased with increasing the ratio which tend to that increasing K_{OV} with increasing D_V and K_{OL} with decreasing $_{DL}$. The mass transfer between the vapour and liquid phases results from the combined contribution of molecular diffusion and a bulk transport of material through the interface. Both vapour and liquid diffusivities are proportional to the temperature, therefore its values increased together to give increasing index with mass transfer coefficient values.

CONCLUSIONS

The overall vapour and liquid mass transfer coefficients (K_{OV} and K_{OL}) increased with increasing the liquid to vapour molar rate ratio (L/V), the diffusivity ratio (D_V /D_L), the density ratio (ρ_V / ρ_L). However, the effect on K_{OV} is more pronounced as indicated by the powers on these parameters in the resulting final equations. But the coefficients decreased with the viscosity ratio and the effect on K_{OV} is larger than on $K_{OL.}$

Finally both K_{OV} and K_{OL} decreased with increasing the relative volatility (α) and at nearly the same order of magnitude.

NOMENCLATURE

D	Distillate flow rate k mole/s.
A	Specific area of packing (m^2/m^3)
a	Constants
K	Mass transfer coefficient kmole/m^2s.
N	Number of moles k mole
HTU	Height of transfer unit, m.
NTU	Number of transfer unit.
L	Liquid molar rate, kmole/hr.
V	Vapour molar rate, kmole/hr.
S	Moles of feed in the still, kmole.
Y	Mole fraction, vapour phase.
X	Mole fraction, Liquid phase.
T	Temperature (K).
D_V	Vapour diffusivity (m^2/s.)
D_L	Liquid diffusivity (m^2/s.)
Z	Column height (m)

Subscript:

b	Bottom
t	Top
ol	Overall liquid
ov	Overall vapour
s	still

Greek letters

μ	Kinematic viscosity, kg/m.s
ρ	Density , kg/m^3
α	Relative volatility.

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