Parameters Determination for Column Design in Gas Absorption Systems

JUAN CARLOS BELTRÁN-PRIETO*, KAREL KOLOMAZNÍK
Faculty of Applied Informatics,
Tomas Bata University in Zlín,
Nám. T. G. Masaryka 5555, 760 01 Zlín,
CZECH REPUBLIC
*Email: prieto@utb.cz

Abstract: - Separation processes by means of mass transfer play a key role in chemical industry. Very often, a number of different components are present in streams from chemical reactors and some of them may be separated from the main stream for environmental reasons, for purification purposes, for commercialization, to be used in other chemical manufacturing process or even reused in the same process, to improve industrial performance, or for sale as a final product. Gas adsorption is one example of this separation process, where it is normally desired to separate a dilute component from a gas stream. In the present paper we studied packed columns by graphical and algebraic methods and determine the tower height using volumetric mass transfer coefficients, we analyze a gas stream that contains a defined amount of a gas, which concentration decreases after being in contact to a packed absorption tower, and model this process by mass balance taking into consideration volumetric mass transfer coefficients, operating line and equilibrium line of the process. The model allows us finally to understand the variation of height column with column diameter, which is particularly useful for the design of absorption columns as it requires the determination of these parameters.

Key-Words: - gas absorption, gas-liquid interface, volumetric mass transfer coefficient, graphical methods

1 Introduction

Gas absorption is an operation of mass transfer that consists in the separation of components of a gaseous mixture by contact with a suitable fluid. Mass transfer separation operations involve the contact of two immiscible phases, sometimes is intermittent (i.e. staged columns) and other continuous, (i.e. packed columns). There exist several means to achieve the mass transfer between fluid phases (i.e. by diffusion or by convective mass transfer). There are cases for example. In which a solute diffuses through a vapor phase and later is absorbed in an immiscible fluid phase, like in the absorption of ammonia from air using water.

During the process, both phases remain in contact and a concentration gradient is generated in each phase, which allows the mass transfer process. The determination of concentration profile and mass transfer rates is normally of particular interest during the analysis of this process, where equilibrium relations play an important role. For this, is important to keep in mind concepts related to equilibrium relation in fluid systems, Henry's law, and equilibrium distribution coefficients.

Absorption is used in vapor recovery from gas streams, gas treatments in refineries, and also for the purpose of waste treatment from industrial sources. During this process, a fluid that exerts a differential solvent effect on the components of the gas is used. Normally, once absorption process is completed, it is desired to regenerate and recycle the absorbent and recovery of the absorbed compounds. For this purpose, a stripping process is subsequently carried out. During these two operations, mass or molar flow rates do not remain constant.

When only one of the components is being transferred between phases it is convenient to differentiate the flow rate and solute free phase for purpose of modeling the total flow rate, the mass balance around the lower end of the column, graphical determination of number of theoretical trays among other calculations [1].

Understanding of gas adsorption process is important in fluid stream analysis, study of hydrodynamic features of a packed column, determination of fluids mass balance [2], calculation of parameters such as mass transfer coefficients, design of plate absorption towers, drag and flooding flows calculation, study of physic-chemical properties and composition of fluids, component recovery contained in fluid streams, industrial design, process simulation, evaluation of the influence of parameters (i.e. diameter of column, packing height, mass transfer coefficient, molar

flow rates, number of trays) [3], process design, countercurrent two-phase flow studies [4], determination of operation line and equilibrium line, optimum stream ratio (inert liquid and gas), effective packing surface area, influence of packing type, evaluation of column efficiency for fractionators, absorbers and strippers [5].

Additionally, they have been successfully used in the removal of air pollutants, odor elimination, in the food industry during the treatment of waste and reuse in the processes, stripping of contaminants from flow stream, absorption in the solvay process where sodium bicarbonate ($NaHCO_3$), and this can be used in effervescent drinks such as fruit juices and baking powder, is produced; absorption in production processes of nitric acid, used as a descaling agent (products that prevent or eliminate the deposit of salts that are formed on the walls) as well in the agriculture and food industry and recovery of organic compounds from aqueous streams.

Additionally, it has been used in absorption and desorption of CO_2 [6], control of CO_2 emissions [7]. It has been reported that the increment in removal efficiency is proportional to the liquid flow rate and solvent concentration. Additionally, gas mass transfer rate increases with an increase in the liquid flow rate [8]. Comparative studies between membrane contactors and absorption towers for post combustion CO_2 capture [9].

Accordingly, simulations have been performed using an absorber model with the corresponding experimental validation in a membrane contactor setup. Using membrane absorbers often require gas treatment. Membranes acts as non-selective barrier between the gas and the solvent and requires the pore size to be larger than the mean free path of diffusing gas. Selection of membrane material depends on chemical solvent properties.

2 Description of Gas-Liquid Systems

The study of the interaction between gas and liquids can be analyzed from two different perspectives, namely as a separation process or as a reaction system. In the case of separation process, the main purpose is to remove an undesirable component from a gas stream and is commonly referred to as gas absorption. The transport of solute specie occurs from a bulk gas to a bulk liquid with measurable solubility and difference of chemical potential of solute in the two phases.

This difference is expressed by a difference in concentration of the substance in the two phases. An interface is present at any point in the gas absorption equipment in which gas contacts the liquid. There are several models that describe the mass transfer from a substance in the gas phase to the liquid phase without reaction.

The two film model developed by Whitman considers that the concentrations at any position in the column do not change with time, the presence of a film at the interface on both sides, a rate of diffusion across the gas-phase film which is equal to the rate of diffusion across the liquid-phase film, negligible resistance to mass transfer across the interface, and the presence of a stagnant gas film on one side of the interface and a stagnant liquid film on the other. [10]

In case reaction occurs, the systems aims to yield a desirable product, as occurs in the manufacture of nitric acid, phenol, nylon. The analysis of this system requires understanding of kinetics and reaction models, which are necessary to include together with gas-liquid mass transfer equations.

3 Mass Transfer Column Design

In several fields of chemical industry it is important to separate compounds that are in the same phase either for environmental reasons, process performance or quality reasons. The separation using immiscible phases as in gas absorption and desorption in dilute systems is one example of this mass transfer operation. It is possible to separate SO_2 using atmospheric pressures in an amine of low vapor pressure, so that small amount of amine is transferred between the fluids, additionally the use of low pressure helps the mass transfer of SO_2 and avoids dissolution of air in the amine.

For this purpose, is necessary the proper design of columns or towers in which a gas enters to the system and flows countercurrent to the liquid. It is often assumed that it is not necessary to remove heat from the system because there is no chemical reaction taking place in the system, therefore mass transfer is the main process involved.

Different types of devices such as grids, sieves, bubble caps or porous solids of different shapes, namely spheres, coils, mesh, cylinders, etc., made of a variety of construction materials such as polymers, ceramics, metals are often placed randomly dumped or structured inside the column to increase the interfacial area between the fluids and improve mass transfer.

The main purpose of the inclusion of this material in the column is to provide a large interfacial area between the phases and to promote

turbulence within the fluids. In the case of turbulent fluids, there is a high transfer rate per unit area of both momentum and mass and as the pressure drop increases, there is also a rise in the rate of transfer of momentum and mass transfer at the same time. Packed column have less liquid holdup and lower pressure drops.

They are often used in cases where compression cost of the gas is significant due to high values of pressure drop through the tower. On the other hand, manual cleaning in packed towers is often difficult, additionally; the weight can be also a limitation, particularly in cases where there the packing is crushed at the bottom. To avoid this intermediate supports are often used but this can increase considerably the price. [11]

Other example of absorption process is the separation of benzene from a gas stream using a heavy oil stream, or the separation of H_2S or CO from a gas stream using monoethanolamine or diethanolamine in aqueous solution. In order to improve the contact between the liquid and the vapor and make more efficient the transport process during absorption or distillation stages, plate towers are generally used. The contact between these phases is achieved either in countercurrent flow or with cross flow of liquid against vapor flow upward.

There exist several configurations depending on the type of flow. In the case of countercurrent flow, the main configurations are dual flow with round holes, turbogrid and ripple trays. However, there are some disadvantages associated to this type of configuration like low turndown ratio (the liquid drains completely off the tray at lower vapor rates) and so, these arrays have not been commonly used.

On cross flow set up, the main configurations are sieve tray, valve tray, and bubble cap tray. The spacing of each tray is determined after consideration of vapor velocity, vessel diameter and accessibility of trays to maintenance. In sieve tray, for example, there are holes of diameter equal to 3-12 mm through which gas bubbles pass through the flowing fluid, which is prevented from flowing down through the holes due to the kinetic energy of the gas. In this system, the level of the fluid is maintained with an overflow weir while the gas ascends through the holes at enough rate to keep most of the liquid from weeping through. Around 5-15% of the total area corresponds to active cross section. These measurements depend on pressure drop values, mass transfer efficiency, weeping and entrainment. Some factors such as vapor rate, density, weir length and tray spacing are important for the determination and calculation of tower diameter.

When valve openings are used, there exist liftable caps that cover the valve trays and through which the vapor flows horizontally into the liquid, providing better mixing in comparison to sieve trays. Additionally, in most cases, the cost associated to the design and construction of valve trays is usually less than sieve trays due to the larger holes and thicker plates which need less support [1]. When tray equipment is used, there occurs a stepwise change in concentration, on the other hand, when packed towers are used, the variation in concentration occurs gradually. Previously, packed towers were generally used in small equipment, for low pressure drop and in cases when it was necessary the consideration of corrosive conditions, however this has changed and currently both type of equipment are competitive and generally used for different purposes and scales. When it is desired to disperse the gas on the tray and maintain a minimum liquid level, bubblecap trays are generally used. However, they are normally associated with considerable costs, hydraulic gradient problems and therefore are not commonly installed.

They are commonly used in low liquid flow rate situations as it occurs in crude vacuum towers. Other problems associated to this type of array are the variation in vapor flow across the cross section and efficiency degradation.

The design of absorption columns requires determination of column height, column diameter to handle liquid and gas flow rates and proper selection of column internal features (packing support, fluid distributor) [12]. When equilibrium data is available, it is possible to determine the number of equilibrium stages for a particular fluids separation process. However, this is not always possible in trays and the theoretical stage is a highly variable quantity. In the case of tower with diameters higher than 3 m, concentration gradients along the path of liquid flow are formed and the amount of mass transfer is higher than the calculated considering the average terminal compositions. Flow rates, viscosities, relative volatilities, surface tension, geometrical configuration of the tray or packing and other parameters associated to Reynolds and Schmidt dimensional numbers affect the efficiency of trays and packing.

It is difficult to perform an experimental measurement of interfacial area between the liquid and the gas. Moreover, it is also complicated the determination of film and overall coefficients k_x' , k_y', K_x' and K_y'. Generally, the measurements performed yield a volumetric mass-transfer coefficient which contains the interfacial area and mass transfer coefficient.

Experimental measurements in a packed tower yield a volumetric mass transfer coefficient that combines the interfacial area and mass transfer coefficient [13]. We aim to study this type of system and determine the tower height using $k_y^{'}a$, we analyze a gas stream that contains a defined amount of a gas, which is reduced after passing through a packed absorption tower.

3.1 General Considerations in Gas Absorption Systems

In the analysis of gas absorption systems, the main information required is the molar rate at which the gas stream enters to the column, including the mole fraction of component present in that stream, which is often small to satisfy the assumptions with regard to the mass transfer coefficient, the exit gas concentration of the component or in all cases the percent of fraction that is desired to achieve, additionally, it is also possible to have information about the molar rate of the liquid, the inlet fraction of the component in the liquid and the type and size of packing. Equilibrium data is often provided in thermodynamic tables from literature references or from experimental data. It is often assumed that the carrier gas is insoluble in the liquid and the liquid has negligible vapor pressure.

With this information is feasible to calculate design parameters such as the packing height or the tower diameter required to achieve the desired separation. It is important to consider that there could be cases where there is a very large difference in the molar rates at which components are transferred, particularly when the mass transfer process deviates significantly from equimolecular counter diffusion. Therefore in gas absorption process only the soluble component is transferred.

This implies that there is no transfer of the insoluble component across the interface. [14]. Columns used in absorption process normally contain trays with a liquid layer flowing across the tray and are often modeled as equilibrium stages. Therefore, each stage of the process is an equilibrium stage; the vapor that exits a stage is in thermodynamic equilibrium with the liquid on that stage. Then, the total amount of solute on each stage corresponds to the sum of the solute in the inert fluid and the gas phase. From this information, it could be possible to determine the rate of change of the amount of solute and the solute mass balance around each particular stage. Moreover, in process design it is desirable to develop time dependent models and linear models describing the system.

4 Model Development

For the present system, we consider the case of gas absorption in column as depicted in Fig.1. The liquid stream absorbs a component containing in the gas feed stream, which enters to the systems at the bottom of the column. The gas leaving at the top of the column exits with less concentration of the component removed [15]. It is assumed that the liquid stream remains inert, without reacting or absorbing into the gas stream.

Accordingly, the major component of the gas stream also remains without reaction or diffusion into the liquid stream. We take into consideration numerical values of $k_y^{'}a = 0.0739\ kg\ mol/s \cdot m^3 \cdot molfrac$ and $k_x^{'}a = 0.169\ kg\ mol/s \cdot m^3 \cdot molfrac$, also we consider that the process occurs at 293 K and 1.013 x 10^5 Pa. The overall mass balance applied on a solute in a packed absorption tower which is diffusing through a stagnant gas and then into a stagnant fluid can be expressed as:

$$L^{'}\left(\frac{x_2}{1-x_2}\right) + V^{'}\left(\frac{y_1}{1-y_1}\right) = L^{'}\left(\frac{x_1}{1-x_1}\right) + V^{'}\left(\frac{y_2}{1-y_2}\right) \quad (1)$$

were y_1 and x_1 are the mol fraction of solute in the gas and liquid respectively [16].

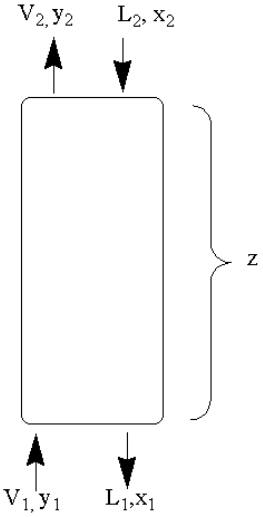

Fig.1: General description of the system under study. Mass balance in absorption column.

For numerical analysis, we consider the case of a 1 mol% gas entering the column ($y_1 = 0.01$) and 0.05 mol% as outlet. ($y_2 = 0.0005$). Additionally, we take into account an inert liquid flow ($L^{'}$) of 100 kg mol/h, while the inert gas flow (V_1) is 90 kg mol/h. We then proceed with following calculations: $V^{'} = (90 \cdot [1 - 0.01]) = 89.1\ kg\ mol\ inert\ air/h$,

$V_2 = V^{'}/(1 - 0.0005) = \left(89.1 kg\ mol\ inert \frac{air}{h}\right)/(1 - 0.0005) = 89.1446$. Additionally,

$$100\left(\frac{0}{1-0}\right) + 89.1\left(\frac{0.01}{1-0.01}\right) = 100\left(\frac{x_1}{1-x_1}\right) + 89.1\left(\frac{5\times10^{-4}}{1-5\times10^{-4}}\right) \quad (2)$$

Which leads to $x_1 = 0.008478$. We now proceed to perform a mass balance to calculate an intermediate point in the operation line,

$$L'\left(\frac{x}{1-x}\right) + V'\left(\frac{y_1}{1-y_1}\right) = L'\left(\frac{x_1}{1-x_1}\right) + V'\left(\frac{y}{1-y}\right) \quad (3)$$

Setting $y = 0.02$

$$100\left(\frac{x}{1-x}\right) + 89.1\left(\frac{0.01}{1-0.01}\right) = 100\left(\frac{0.008478}{1-0.008478}\right) + 89.1\left(\frac{0.02}{1-0.02}\right) \quad (4)$$

Which solution leads to x = 0.0124. Setting y = 0.03 we obtain

$$100\left(\frac{x}{1-x}\right) + 89.1\left(\frac{0.01}{1-0.01}\right) = 100\left(\frac{0.008478}{1-0.008478}\right) + 89.1\left(\frac{0.03}{1-0.03}\right) \quad (5)$$

Which solution leads to $x = 0.0264$. The previous points found are plotted in the operating line. The approximate slope at x_1, y_1 can be calculated by

$$\text{slope} \cong -\frac{k_x' a(1-y_1)}{k_y' a(1-x_1)} = -\frac{0.169(1-0.01)}{0.0739(1-0.008478)} = -2.28336 \quad (6)$$

If we plot this line, the values at M_1 are $y_{i1} = 0.00707$, $x_{i1} = 0.009758$. Data for equilibrium line is presented in Fig. 2 according to information provided in Table 1 [13].

Table 1. Equilibrium data for the system under study.

equilibrium data		
x (mol fraction)	partial pressure (mm Hg)	Y (mol fraction)
0	0	0.000
0.021	12.000	0.016
0.026	15.000	0.020
0.031	18.200	0.024
0.041	24.900	0.033
0.050	31.700	0.042
0.074	50.000	0.066
0.096	69.600	0.092
0.137	114.000	0.150
0.175	166.000	0.218
0.210	227.000	0.299
0.241	298.000	0.392
0.297	470.000	0.618

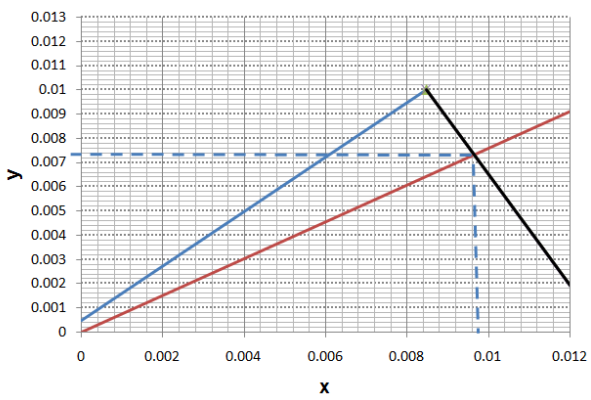

Fig. 2: Operation line (blue), equilibrium line (red) and interface composition (P_1M_1) (dashed line).

Following, we proceed to determine $(1-y)_{iM}$ after equation (7)

$$(1-y)_{iM} = \frac{(1-y_{i1})-(1-y_1)}{ln\left[\frac{(1-y_{i1})}{((1-y_1))}\right]} \quad (7)$$

$$(1-y)_{iM} = \frac{(1-0.00707)-(1-0.01)}{ln\left[\frac{(1-0.00707)}{((1-0.01))}\right]} = 0.9914 \quad (8)$$

$$(1-x)_{iM} = \frac{(1-x_{i1})-(1-x_1)}{ln[(1-x_{i1})/((1-x_1))]} \quad (9)$$

$$(1-x)_{iM} = \frac{(1-0.009758)-(1-0.008478)}{ln[(1-0.009758)/((1-0.008478))]} = 0.9908 \quad (10)$$

$$slope = -\frac{k_x' a(1-y)_{iM}}{k_y' a(1-x)_{iM}} = -\frac{0.169(0.9914)}{0.0739(0.9908)} = -2.28822 \quad (11)$$

Replotting $P_1M_1, y_{i1} = 0.007072, x_{i1} = 0.009757$ for the slope at point 2

$$slope = -\frac{k_x' a(1-y_2)_{iM}}{k_y' a(1-x_2)_{iM}} = -\frac{0.169(1-0.0005)}{0.0739(1-0)} = -2.28573 \quad (12)$$

The slope is essentially constant for P_1M_1 and P_2M_2 as a result, using an algorithm developed to find the intersection between the operation line and equilibrium line we found that $y_{i2} = 0.000125, x_{i2} = 0.00016$, which is represented graphically in Fig. 3.

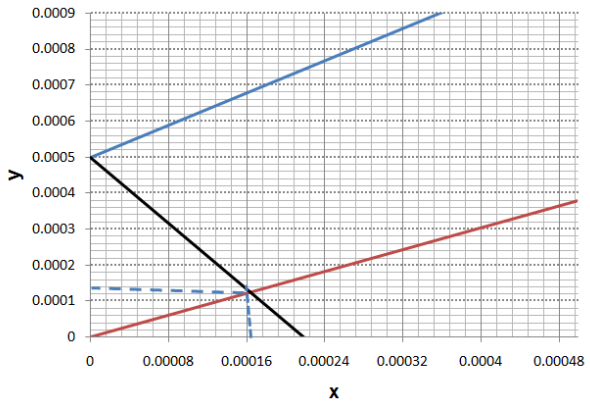

Fig. 3: Operation line (blue), equilibrium line (red) and interface composition (P_2M_2) (dashed line).

For Fig. 2, we can also used equation (13)

$$y_2 = slope(x_2 - x_1) + y_1 \quad (13)$$

considering the log mean driving force (14) [17]

$$(y - y_i)_M = \frac{(y_1 - y_{i1}) - (y_2 - y_{i2})}{ln[(y_1 - y_{i1})/((y_2 - y_{i2}))]} \quad (14)$$

$$(y - y_i)_M = \frac{(0.01 - 0.00707) - (0.0005 - 0.000125)}{ln[(0.01 - 0.00707)/((0.0005 - 0.000125))]} \quad (15)$$

$$(y - y_i)_M = 0.0013 \quad (16)$$

To calculate the total molar flow rate $\overline{V} = (V_1 + V_2)/2 = (90 + 89.14)/2 = 89.57 kg\ mol/h = 0.0248 kg\ mol/s$.

Applying $(\overline{V}/S)(y_1 - y_2) = k_y' az(y - y_i)_M$ leads to the next equation: $(0.0248/S)(0.01 - 0.0005) = 0.0739z(0.0013), (0.0002363/S) = 0.000124z$. The graphical representation of previous equation is depicted in Fig. 4, where we can observe the variation of column height with column diameter.

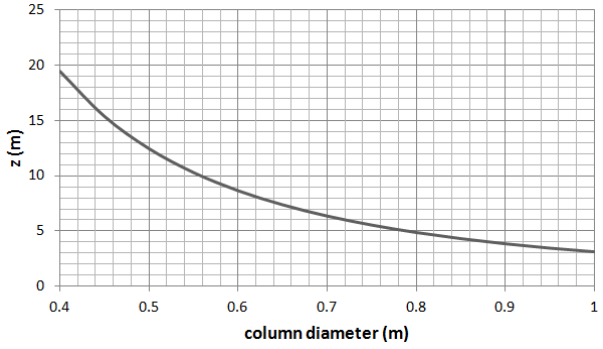

Fig. 4: Variation of height column (m) with column diameter (m).

After determination of the concentration changes in the column, the height of the column for a required outlet concentration of one of the components can be calculated [18]. For numerical purposes, we consider the case of tower diameter of 0.5m

$$([0.0002363 \cdot 4]/(\pi \cdot [0.5]^2)) = 0.000124z \quad (17)$$

$$0.0012 = 0.000124z \quad (18)$$

$$z = 9.74 \quad (19)$$

5 Conclusion

Absorption towers are used to retrieve different type of gases from the vapor phase into the liquid phase, but not necessarily only water. Packed towers are generally used for this purpose. It consists of a cylindrical column, or tower, with a gas inlet; entrance and a distributor at the top and outputs for gas and liquid. A tower is packed so it provides a large surface area to facilitate contact between liquid and gas. This allows continuous contact between the liquid and the gas in both the countercurrent flow and the parallel flow. The liquid is distributed and runs down through the packed bed, so that it exposes a large surface in contact with the gas.

A study was performed to evaluate interface composition and height column in an absorption process. This is important particularly for the sequence of chemical and physical operations, operating conditions, construction specifications during process design to ensure proper mass transfer process and achieve the desired recovery, yield or mass transfer rate during the absorption process. Additionally, the model allows us to understand the variation of height column with column diameter. The design of absorption columns requires determination of these parameters as a tool to handle liquid and gas flow rates and proper selection of column internal features such as packing support, or fluid distributor

Acknowledgements:
This work was supported by the Ministry of Education, Youth and Sports of the Czech Republic within the National Sustainability Programme project No. LO1303 (MSMT-7778/2014), the European Regional Development Fund under the project CEBIA-Tech No. CZ.1.05/2.1.00/03.0089 and also by the internal project No.

RVO/CEBIA/2019/003 "Vývoj recyklačních a zpracovatelských technologií".

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