CHAPTER 2

LITERATURE SURVEY

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Effective interfacial areas and mass transfer coefficients in packed columns for cases of absorption without chemical reaction and absorption with chemical reaction have been investigated by many investigators. The relevant literature has been reviewed critically in this chapter. The other design parameters like liquid distribution, capacity limits, holdup, pressure drop. etc. studied by different investigators have also been briefly reviewed. Further the generalised correlations available in the literature (4) for predicting the values of holdup (static holdup 'h_{st}', operating hold up 'h_o'), interfacial areas (wetted surface area 'a_w', effective interfacial area for physical absorption 'a_p', and effective interfacial area for chemical absorption 'a_c') and the values of mass transfer coefficients (k_La, k_L and k_G) have also been briefly reviewed.

2.1.0 EFFECTIVE INTERFACIAL AREAS IN COLUMNS :

2.1.1 Wetted surface area (a_w) :

The values of wetted surface area (a_w) have been obtained by different investigators using different experimental techniques. Shulman and Degouff (5) measured the rates of vaporization of napthalene in dry bed as well as in water irrigated bed under otherwise identical conditions and accordingly estimated the values of a_w for 0.025m napthalene Raschig rings in a column of 0.25m internal diameter (i.d.). Shulman and coworkers (7) also utilised the same technique to obtain the values of a_w for 0.013m and 0.038m napthalene rings.

Grimley (8), using a electrical resistance method, estimated the values of a_w for 0.0095m stoneware Raschig rings in a 0.056m i.d. column for liquid rates ranging from 1 to 8 kg/m^2s.

Mayo et al. (9) utilised coloured technique to obtain a_w values wherein paper Raschig rings were wetted by passing coloured water over a packed bed of paper R.R. The range of variables covered by these investigators included paper R.R. of 0.013m packed in 0.075m i.d. column for liquid rates ranging from 4.2 to 13 kg/m^2s. Fujita and Sacuma (10) also measured the values of a_w of Raschig rings of different sizes 0.013m, 0.025m, and 0.038m in a column of 0.025m i.d., by the technique identical to Mayo et al. Apart these investigators also studied the effect of packed bed height on a_w values and concluded that the effect of packed bed height on wetted surface area was insignificant.

Hikita and Kataoka (11) investigated the effect of viscosity and surface tension of solvents/absorption media for paper Raschig rings of sizes 0.015m, 0.025m and 0.035m. The range of variables covered by these investigators included variation in viscosity from 0.9 to 3.8 mNs/m^2 and variation in surface tension from 30 to 65 mN/m.

Onda et al. (12) also obtained values of a_w for 0.008m ceramic Raschig rings for a variety of solvents with viscosities ranging from 1 to 10 mNs/m^2 and surface tensions ranging from 46 to 76 mN/m.

Thus, survey of existing literature on wetted surface area for napthalene, ceramic and paper packings indicates that the parameteric ranges covered by different investigations are the following :-

ρ_L (kg/m^3)	997.00	-	1150.00
μ_L (mNs/m^2)	0.89	-	10.80
σ (mN/m)	34.00	-	76.00
d_p (m)	0.008	-	0.038
a_t (m^2/m^3)	115.00	-	588.00
L (kg/m^{2}s)	0.14	-	13.50
a_w (m^2/m^3)	24.00	-	270.00

2.1.2 Effective interfacial area during physical absorption (a_p) :

The effective interfacial area for mass transfer during absorption without chemical reaction cannot be directly measured. However, the values of a_p can be estimated by dividing the values of volumetric gas/liquid side mass transfer coefficients by the appropriate values of k_G or k_L obtained under otherwise identical conditions.

Shulman and Coworkers (13) thus reported the values of effective interfacial area for the system ammonia-water using

Fellinger's data of K_Ga for the same system. A column packed with ceramic Raschig rings of different sizes 0.025m, 0.038m and 0.05m, with liquid rates ranging from 0.6-10kg/m^2s at various gas rates was utilised by these investigators for obtaining the values of a_p. It was observed that below loading point, the values of effective interfacial area were largely independent of gas rates.

Yoshida and Koyanagi (14) using systems CO_2-water and CO_2-methanol also reported the values of a_p in a column of 0.125m i.d., packed with 0.015m and 0.025m ceramic Raschig rings.

Hikita and coworkers (15) investigated the effect of viscosity, surface tension and density of solvents on the values of a_p for CO_2 absorption in various solvents viz. water, aqueous methanol solutions, cane sugar solutions etc. in a column of 0.125m i.d. packed with 0.025m and 0.015m ceramic Raschig rings. These investigators varied physical properties of the systems widely, with viscosities ranging from 0.5 mNs/m^2 to 3.7 mNs/m^2, surface tensions from 22 to 73.5 mN/m and densities ranging from 780 to 1150 kg/m^3.

Yoshida and Koyanagi (16,17) also obtained the values of a_p for absorption of methanol vapour in water using 0.12m i.d. column packed with 0.015m and 0.025m ceramic Raschig rings to the heights of 0.1 and 0.2m.

From literature survey, it is thus, observed that the parametric ranges covered by different investigators for the values of a_p in columns packed with ceramic Raschig rings are the following

ρ_L (kg/m^3)	787.00	-	1140.00
μ_L (mNs/m^2)	0.50	-	3.70
σ (mN/m)	22.00	-	74.00
d_p (m)	0.015	-	0.05
a_t (m^2/m^3)	102.00	-	330.00
L (kg/m^{2}s)	0.27	-	100.00
a_p (m^2/m^3)	14.00	-	120.00

2.1.3 Effective interfacial area during chemical absorption (a_c) :

Sharma and Danckwerts (18) have reviewed how gas absorption accompanied by chemical reaction could be used conveniently to measure the values of effective interfacial area in packed columns. The relevant literature on a_c data obtained by different investigators using these chemical techniques has been summarized in the following pages.

Danckwerts and coworkers (19) investigated absorption of pure CO_2 into sodium carbonate-bicarbonate buffer solutions in a 0.1m i.d. column packed with 0.013m ceramic Raschig rings and obtained values of interfacial area during chemical absorption by use of Danckwerts plots.

Vassilatos et al. (20) reported the values of interfacial area for CO_2 absorption into aqueous ammonia solutions using a column of 0.12m i.d. packed with 0.013m ceramic Raschig rings.

Richards et al. (21) studied the absorption of pure CO_2 into sodium carbonate-bicarbonate buffer solutions, containing varying amounts of arsenous oxide catalyst, in a 0.1m i.d. column packed with 0.013m ceramic Raschig rings and reported the values of a_c using the Danckwerts plot.

Danckwerts and Gilliham (22) measured the values of a_c in a 0.46m i.d. column packed with 0.038m ceramic Raschig ring for the system, pure CO_2 absorption into potassium carbonate-bicarbonate buffer solutions, using sodium hypochlorite as catalyst.

Gehlawat and Sharma (23) investigated the effect of viscosity of absorption media on the values of a_c by absorbing isobutylene into aqueous solution of sulphuric acid. (μ_L range from 8 to 9 mNs/m^2) in a column packed with 0.0095m ceramic Raschig rings.

Vidwans and Sharma (24) employing conservative values of specific rates of absorption obtained interfacial area in columns of 0.044m, 0.1m and 0.2m i.d. packed with 0.0095m ceramic Raschig rings, by absorbing lean CO_2 and 1.8 M NaOH solutions. Some data on a_c for columns packed with 0.006m glass Raschig rings, 0.013m ceramic Intalox saddles and 0.025m ceramic Raschig rings were also reported by these investigators using the same system. Further, they also obtained interfacial area values for the system CO_2 absorption into 1.7 N monoethanolamine solutions in a column packed with 0.0095m ceramic Raschig rings.

Jhaveri and Sharma (25,26) using columns packed with 0.0095m ceramic Raschig rings measured the values of a_c by absorbing oxygen into cuprous chloride solutions. The ionic strength of these solutions varied from 3.5 to 9.2 k ion/m^3 and the viscosity varied from 0.9 to 2.5 mNs/m^2. The kinetics of this reaction was second order with respect to the reactant and first order with respect to the solute. These investigators also measured the values of interfacial area by absorbing oxygen form air into sodium dithionite solutions. The reaction of O_2 with sodium dithionite followed first order kinetics with respect to dithionite when the dithionite concentration was below 0.08 M and second order kinetics when it was above 0.08 M.

Danckwerts and Rizvi (27) obtained the values of rates absorption of Oxygen in sodium sulphite solutions containing Cobalt salts as catalyst, in 0.45m i.d. column packed with 0.038m size packings made of different materials. The rates of absorption of O_2 were independent of k_L. The values of a_c obtained by these investigators for ceramic Raschig rings were the highest in comparison to the other packings.

Onda et al. (28) also utilised the sulphite oxidation technique for obtaining the values of a_c in a column of 0.15m i.d. packed with 0.015m ceramic Raschig rings using air as well as pure O_2. They observed that absorption of O_2 in sulphite solutions followed reaction kinetics with varying order and with an increase in partial pressure of O_2 (from 0.2 to 1.0 atm), the value of order of reaction decreased from two to one. These investigators also interpreted the data on O_2 absorption from air in sulphite solutions by DeWaal and Beek (29) satisfactorily using above mentioned conclusions.

Puranik (30) reported values of a_c for the absorption of CO_2 into 1 to 2 N NaOH solutions in a column packed with 0.013m ceramic Raschig rings.

Sahay and Sharma (31) also reported the values of a_c for two systems ; CO_2 + NaOH solutions and O_2 + sodium dithionite solution under different sets of conditions using different packing materials like plastics, stainless steel etc.

The sulphite oxidation method was also utilised by Andrieu and Claudel (32), Linek et al. (33) and Andrieu (34) for finding out the values of a_c in columns packed with packings made of different materials like pyrex glass, ceramic, plastics, pyrex rings coated with silicone etc. The influence of wettability of different packing materials on the values of a_c was investigated in detail by Linek, Krivsky and Hudec (35) also using the sulphite oxidation technique. These investigators utilised ceramic, polyethylene, polypropylene and hydrophylised plastic Raschig rings as well as Pall rings of 0.015m size in columns of 0.143m and 0.29m i.d.

Alper (36) determined the effect of packing length to diameter ratio (l/d_p) on the values of a_c for the system CO_2 absorption into potassium carbonate-bicarbonate buffer using Danckwerts plot, for columns packed with 0.016m diameter polyethylene rings, and observed that as ratio (l/d_p) decreases, the value of a_c increases.

Rizzuti et al. (37,38) measured interfacial area for absorption of CO_2 into potassium carbonate-bicarbonate buffer containing potassium arsenite catalyst, in a column packed with 0.01m glass Raschig rings, using Danckwerts plot. These investigators studied the effect of kinematic viscosity on a_c by varying ν from 1.035 mm^2/s to 1.55 mm^2/s which was achieved by adding sugar to the buffer solution.

Recently Rizzuti and coworkers (39) reported the values of a_c for columns packed with stainless steel and glass Raschig rings by absorbing CO_2 into potassium carbonate-bicarbonate buffer solutions using Danckwerts plots. They observed that, under otherwise identical conditions, the values of a_c were higher for S.S. packings in comparison with glass packings.

2.2.0 MASS TRANSFER COEFFFICIENTS FOR GAS ABSORPTION :

2.2.1 Volumetric liquid side mass transfer coefficients (k_La) :

The volumetric liquid side mass transfer coefficient (k_La) appears to be the most extensively and elaborately studied design parameter. In recent years, the values of 'k_La' have been obtained under different conditions. The effect of wettability of packing materials, surface tensions and viscosities of the absorption media on the values of k_La have been investigated only during the past decade. The relevant literature on k_La data obtained by different investigators has been summarized in the following pages.

Hikita et al. (40) absorbed CO_2 into water in columns of 0.07m i.d. and 0.125m i.d. packed with 0.015 m and 0.025m ceramic Raschig rings respectively, and reported the values of k_La for liquid rates varying from 0.1 to 16.6 kg/m^{2}s.

Danckwerts and coworkers (19) using 0.1m i.d. column packed with 0.013m ceramic Raschig rings reported k_La values for CO_2 absorption into water and 1.05 M Na_2SO_4 solutions at the same volumetric flow rates.

Richards et al. (21) reported k_La values for the absorption of CO_2 into 1.78 m NaCl solution, with liquid rates ranging from 0.77 to 3.8 kg/m^{2}s in a 0.1m i.d. column packed with 0.013m ceramic Raschig rings.

Danckwerts and Gilliham (22) reported k_La values for CO_2 absorption into 0.53 M Na_2SO_4 solutions in a 0.45m i.d. column packed with 0.038m ceramic Raschig rings.

Onda and coworkers (28) have reported k_La values for physical absorption of O_2 into water containing sulphite solutions using a 0.15m i.d. column packed with 0.015m ceramic Raschig rings.

Sahay (41) investigated the effect of surface tension of absorption media on k_La values by absorbing CO_2 into water containing varying amounts of isopropanol, such that, the surface tension varied from 72 mN/m for pure water to 46 mN/m for water containing 8% isopropanol by volume. Columns packed with Raschig rings, Pall rings and Intalox Saddles made of different materials like ceramic, polypropylene, polyvinylchloride, and stainless steel were used and the values of k_La were obtained under widely varied conditions.

Joosten and Danckwerts (42) reported the values of k_La for CO_2 absorption in aqueous solutions of $NaNO_3$ containing sugar, the liquid viscosities ranged from 1.4 to 2.0 mNs/m^2.

Mangers and Ponter (43) obtained k_La values for absorption of CO_2 into water using a 0.1m i.d. column packed with 0.01m size glass Raschig rings as well as PTFE Raschig rings. The k_La values for glass rings were substantially higher than those obtained for PTFE rings under otherwise identical operating conditions.

Mangers and Ponter (44) also investigated the effect of viscosity of absorption media on the values of k_La by absorbing pure CO_2 into aqueous glycerol solutions containing upto 70% by weight of glycerol. With an increase in liquid viscosity from 0.9 to 26 mNs/m^2, the values of k_La decreased substantially.

Onda and coworkers (45,46) reported k_La data for the systems CO_2-water and H_2-water in a column of 0.06m i.d. packed with 0.006m and 0.008m ceramic Raschig rings.

The survey of existing literature on volumetric liquid side mass transfer coefficients indicates that the parametric ranges covered by different investigations are the following :

ρ_L	(kg/m^3)	984.00	-	1240.00
μ_L	(mNs/m^2)	0.77	-	16.80
σ	(mN/m)	46.40	-	76.80
D_L	(m^2/s)	2.7×10^{-10}	-	4.8×10^{-9}
d_p	(m)	0.006	-	0.038
a_t	(m^2/m^3)	130.00	-	785.00
L	(kg/m^2s)	0.37	-	18.80
k_La	(s^{-1})	1.58×10^{-3}	-	35.14×10^{-3}

2.2.2 Overall gas side mass transfer coefficients (K_Ga) during physical absorption :

Houston and Walker (47) obtained the values of K_Ga by absorbing highly soluble gases like ammonia and vapours of methanol, ethanol and acetone into water in a column of 0.3m i.d. packed with 0.025m carbon Raschig rings. For all the systems investigated by these investigators, the resistance to mass transfer in the liquid side was negligible.

Zabban and Dodge (48) investigated the effect of total pressure on the values of K_Ga by absorbing methanol/acetone vapours into water in a column of 0.15m / 0.3m i.d. packed with 0.013m Raschig rings/0.025m Berl Saddles, the operating pressure range being 1 to 14 atm a. Results indicated that gas side coefficient (k_G) were inversely proportional to the total pressure at constant gas flow rate.

Hutchings et al. (49) also determined K_Ga for absorption of acetone into water in a column of 0.15m i.d. packed with 0.013m/0.019m/0.032m ceramic Raschig rings.

Dyer and Dodge (50) using a 0.03m i.d. column packed with carbon rings of 0.025m absorbed ammonia into water and studied the effect of gas/liquid flow rates, degree of humidification and column repacking on the values of K_Ga.

Molstad et al. (51) also obtained the values of K_Ga for varying gas and liquid rates for the case of absorption of ammonia into water using a variety of packings such as, drip point grid packings, triple spiral tiles, partition rings, Berl saddles and Raschig rings.

Fellinger (52) has also reported extensive data of K_Ga for the absorption of ammonia in water using columns packed with Raschig rings/Berl saddles of sizes ranging from 0.013m to 0.05m.

2.2.3 Volumetric gas side mass transfer coefficients (k_Ga) :

(I) k_Ga by Instantaneous reaction technique :

The values of k_Ga obtained by carrying out an instantaneous reaction in the liquid phase have been reported exclusively by Sharma and coworkers. Vidwans and Sharma (24,53) absorbed Cl_2/SO_2 from air-Cl_2/SO_2 mixtures into aqueous NaOH and reported the values of k_Ga using a column of 0.044m i.d./0.1m i.d./0.2m i.d. packed with ceramic Raschig rings/Intalox saddles of different sizes.

Sahay (41) also obtained k_Ga values by absorbing SO_2/Cl_2 from air-SO_2 mixture/air-Cl_2 mixture into aqueous NaOH solutions in a column of 0.2m i.d. packed with 0.025m packings of different types like Raschig rings/Intalox saddles/Pall rings and made of different materials like ceramic, polypropylene and PVC.

(II) k_Ga by Vaporization technique :

Surosky and Dodge (54) vaporized water and various organic solvents like benzene, methanol and ethyl - n butyrate into air streams in a column of 0.2m i.d. packed with 0.025m size. Carbon Raschig rings and investigated the effect of diffusivity of gases on the value of k_Ga.

(III) k_Ga from HTU data :

Lynch and Wilke (55) determined the values of $(HTU)_G$ for the vaporization of water into different gases like, air, Helium and Freon-12 in a column of 0.3m i.d. packed with ceramic Raschig rings. These investigators also studied the effect of fluid properties on the gas side mass transfer coefficient.

2.2.4 True liquid side mass transfer coefficient (k_L) :

Danckwerts plots could be used conveniently to obtain the value of k_L and 'a' separately. The relevant information has been summarized in the following paragraphs.

Danckwerts and Gilliham (22) absorbed CO_2 into buffer solutions containing 0.6 M K_2CO_3 and 0.2 M $KHCO_3$ with sodium hypochlorite as catalyst and obtained the values of k_L in a column packed with 0.038m Raschig rings.

Alper (36) also absorbed CO_2 into buffer solutions containing 1M K_2CO_3 + 1 M $KHCO_3$ with arsenite as catalyst and reported the values of k_L in a column packed with polyethylene Raschig rings of size 0.016m.

Rizzuti and coworkers (37,38) absorbed CO_2 into potassium carbonate-bicarbonate buffer solutions containing arsenite as catalyst and obtained the values of k_L. These investigators also varied the kinematic viscosity of the absorption media by adding sugar to the liquid and investigated the effect of viscosity on the values of k_L.

Rizzuti and Brucato (38,56) also investigated the effect of viscosity of absorption media on the values of k_L by absorbing CO_2 into potassium carbonate - bicarbonate - arsenite buffer solutions also containing sugar in different proportions.

2.2.5 Overall gas side mass transfer coefficients (K_Ga) during chemical absorption :

When gas absorption is accompanied by a chemical reaction the capacity per unit volume of solvent/absorption media for absorbing gas increases substantially. The factor β (ratio of $k_L'a$

to k_La) appears to be an important parameter in this respect. If the experimental values of overall gas side mass transfer coefficient during chemical absorption (K_Ga) are known, the observed values of $k_L'a$ could be estimated conveniently. The pertinent information available in the literature about the values of K_Ga has been reviewed here.

Vidwans (53), Tepe and Dodge (57), also Blum and coworkers (58,59) studied absorption of CO_2 into aqueous solution of 1 to 3 N NaOH in columns packed with ceramic/carbon Raschig rings. These investigators concluded that the reaction between CO_2 and NaOH solution was liquid film controlled and the values of K_Ga were independent of the gas flow rates. Blum and coworkers (58,59) also obtained the values of K_Ga for the system CO_2-NaOH solution when concentration of reactive species in solution was very low.

Leva (60,61) carried out an extensive investigation on a pilot plant scale for the system CO_2-NaOH solutions using a column of 0.2m i.d. and 3.0m height packed with different packings like metal/ceramic Raschig rings, Berl saddles and Intalox saddles, and obtained the values of K_Ga under different sets of conditions.

Teller and Ford (62) also carried out an extensive investigations on a pilot plant scale for the system CO_2-monoethanol amine using a column of 0.2m i.d. and 2.0m height packed with different packings such as steel Raschig rings, ceramic Berl saddles and polyethylene Tellerettes and obtained the values of K_Ga under different sets of conditions. These investigators observed that the values of K_Ga for Tellerttes were higher than the values of K_Ga for Raschig rings and Berl saddles.

Absorption of CO_2 in ammonia solutions was investigated by Vassilatos et al. (63) while absorption of CO_2 in aqueous solutions of NaOH/KOH was investigated by Yoshida and Miura (64). These investigators utilised a column of 0.12m i.d. packed with 0.013m/0.025m stoneware/porcelain Raschig rings and reported K_Ga under different sets of conditions. Investigators Yoshida and Miura (64) also obtained the values of K_Ga for the system CO_2 + NaOH solutions when concentration of the reactive species in solution was very low.

Cryder and Maloney (65) obtained the values of K_Ga for the system : CO_2 absorption into diethanolamine solutions, using a packed column of 0.2m i.d. packed with 0.019m stoneware Raschig rings. These investigators concluded that the reaction between CO_2 and diethanolamine solutions followed third order kinetics first order with respect to CO_2 and second order with respect to diethanolamine solutions.

Onda and coworkers (66) investigated the absorption of CO_2 into aqueous NaOH solutions, particularly in the region of very low concentration of reactive species in the absorption media (the range being 0.05 to 1 N NaOH solution) and obtained the values of K_Ga using a column 0.12m i.d. packed with 0.015m ceramic Raschig rings.

Yoshida and Miura (67) also investigated the absorption of CO_2 into aqueous solution of KOH particularly in the region of low concentration of reactive species in the solution and obtained values of K_Ga using a column of 0.12m i.d. packed with 0.025m and 0.013m Raschig rings.

2.3.0 OTHER DESIGN PARAMETERS :

2.3.1 Liquid distribution in packed columns :

The distribution of liquid flowing through a randomly packed bed has been extensively studied by numerous investigators (68-76). Scott (68), Tour and Lerman (69) proposed a stochastic model based on random walk hypothesis for the distribution of liquid. Cihla and Schmidt (70) proposed a diffusion model for the liquid distribution in packed columns which in corporated a radial spread factor. The diffusion model proposed by Cihla and Schmidt (70) and other investigators (71,72,73) assumes formation of a liquid film on the packing and liquid flows over packing as a moving fluid. LeGoff and Lespinasse (74) further indicated that the flow of liquids through a packed bed follows preferred paths through packings. Some paths being stagnant/semistagnant and other being dynamic/moving paths. Further, the effect of packing size, wettability and liquid surface tension have been investigated by Wijffels et al. (75) and also by Bemer and Zuiderweg (76). The relevant information available in these references has been critically analysed elsewhere in Chapter (6) : Mathematical modelling of interfacial areas and mass transfer coefficients.

2.3.2 Capacity limits and pressure drop :

The operating range of a packed column is defined by the upper and lower capacity limits. Determination of the upper capacity limit (flooding velocity) is of prime importance, because from its numerical value, the column diameter can be estimated. The first generalised correlation for flooding velocities was given by Sherwood et al. (77) which was modified by Lobo et al. (78) by introducing a term 'F' packing factor to characterize a particular packing size and shape. Leva (79) improved the above mentioned correlation by introducing the parameter of constant pressure drop. Eckert (80,81) finally modified the same correlation by incorporating an appropriate index to the μ_L term and changing the packing factor (F) of Lobo based on flooding, to averaged figures based on operating ranges of packings. The modified correlation proposed by Eckert (81) could be expressed as a log-log plot of

$$\frac{G^2 F \ \mu_L^{0.1}}{\rho_G(\rho_L - \rho_G)} \quad \text{versus} \quad \frac{L}{G}\sqrt{\left(\frac{\rho_G}{\rho_L - \rho_G}\right)} \qquad (2.1)$$

Recently, Billet and Schultes (82) have also developed a generalised correlation for predicting the capacity limits in packed column within ± 10 % error.

The pressure drop of gas phase below the loading point can be predicted by the correlations proposed by Leva (79), Bemer and

Kalis (83) and Takahashi et al. (84). In the correlation proposed by Takahashi et al., the total pressure drop is expressed as the sum of dry and wet pressure drop. As could be seen from Equations (2.1.1 and 2.1.2) mentioned in Table (2.1) for estimating the values of ΔP_{dry} and ΔP_{wet}, one requires the knowledge of hydrodynamic conditions, packing characteristics and the value of liquid holdup.

2.3.3 Liquid holdup in packed columns :

Numerous investigators (85-93) have developed correlations to predict the liquid holdup in packed columns. The relevant important correlations for estimating the values of operating holdup (h_O), static holdup (h_{st}) are presented in Table (2.1)

Based on the different correlations reported in Table - (2.1) the following observations can be made :

(i) With an increase in the liquid mass flow rate (L) and its viscosity (μ_L) and surface tension (σ) and with a decrease in density of liquid (ρ_L), the operating holdup increases.

(ii) The operating holdup can be correlated in terms of dimensionless numbers such as Film number (Fi), Froude number (Fr) Weber numbers (We), Reynold numbers (Re) and Gallileo number (Ga). With an increase in the values of Fi, Fr and Re, and with a decrease in the values of We and Ga, the operating holdup increases.

Table - 2.1

Correlations for pressure drop and hold up

Correlations for Δ P

Takahashi (84) :

$(\Delta P\ dry/Z) = 4(f/d_p)\ (\mu_G/\varepsilon)\ (\rho_G/Z)$ (2.1.1)

$(\Delta P\ wet/Z) = K\ h_t^3\ (U_G/\varepsilon - h_t)^2$ (2.1.2)

Correlations for operating holdup (h_o)

Otake and Okada (87) :

$h_o = 1.295\ (d_p L/\mu_L)^{0.676}\ (d_p^3 g\ \rho_L^2/\mu_L^2)^{-0.44}$ (2.1.3)

Mohunta and Laddha (88) :

$h_o = 16.13\ (\mu_L\ L^3\ N/\rho_L^4 g^2)^{0.25}\ (N\ d_{pe}^3)^{-0.5}$ (2.1.4)

where N is the packing density and d_{pe} is the diameter of a sphere having the same volume as a piece of packing.

Buchanan (90) :

$h_o = 9.25\ (Fi)^{0.48} + 0.845\ (Fr)^{0.36}$. (2.1.5)

where Fi is the film number defined as $Fi = (L\,\mu_L/\rho_L^2 g d_p^2)$

Gelbe (92) :

$h_o = 1.59\ (d_i/d_p)^{-0.55}\ (We/Fr)^{0.143}\ (Ga)^{-0.3}\ (Re)^n$ (2.1.6)

where d_i is the hydraulic diameter of the smallest inner area of the ring. When Re > 1 the value of index n = 0.454 otherwise n = 0.333.

Correlations for static hold up (h_{st}) .

Gelbe (92) :

$h_{st} = 1.65 \times 10^{-4}\ (a_t d_p)^2\ \log\ (X/X_{crit})$ (2.1.7)

where X is a force ratio defined as $X = [We.Fr/Re^2\ (a_t d_p)^6]$ when $We/Fr < 10$ else $X = 0.01\ [We^3/FrRe^2\ (a_t d_p)^6]$ and the critical value of $X_{crit} = 1.4 \times 10^{-13}$.

(iii) The static holdup also appears to be dependent on ρ_L, μ_L and σ . With an increase in μ_L and σ, and with a decrease in ρ_L, the static hold up increases.

Liquid holdup in packed columns has been analysed in detail by Gelbe (92). Further, he has distinguished between adherent holdup and static holdup. According to his analysis, the adherent holdup becomes equal to the highest values of static holdup when liquid rate approaches to zero. Also, when liquid flow rates are very high, the values of static holdup approaches to zero and operating holdup becomes equal to total holdup. Investigations by Bennet and Goodridge (93), Schubert et al. (94) and Kushalkar and Pangarkar (95) who have utilised tracer techniques to obtain the values of static and operating holdup, also suggest that the actual static holdup is likely to be less than the adherent holdup (so called static holdup) reported by Shulman et al. (85). In view of the above analysis, the correlations proposed by Gelbe for predicting the values of h_o and h_{st} appear to be more appropriate and sound than other correlations.

2.4.0 CORRELATIONS FOR INTERFACIAL AREAS IN PACKED COLUMNS

2.4.1 Correlations for wetted surface area (a_w) :

The following empirical correlations have been proposed in terms of dimensional quanlitities for predicting the values (a_w/a_t):

Fujita et al. (11).

$$a_w/a_t = 1 - 1.02\ e^{-0.278\ (L/a_t\ \ \mu_L)^{0.4}} \qquad (2.2)$$

Hikita et al. (12).

$$a_w/a_t = 0.464\ L\ (\sigma/20)^{(1.42\ d_p^{-0.7})} \qquad (2.3)$$

VanKrevelen et al. (96).

$$a_w/a_t = 1 - e^{(-5000\ L/\rho_L\ )} \qquad (2.4)$$

where L = liquid mass flow rate, ρ_L = liquid density, d_p = packing size, μ_L = liquid viscosity and σ = liquid surface tension.

The generalised correlations in terms of dimensional numbers/groups for predicting the values of a_w/a_t have been proposed by Onda et al. (97), Kolev (98) and Gnielinski et al. (99). The Onda's generalised correlation incorporating the dimensionless numbers - Reynold's number (Re), Weber number (We), Froude number (Fr) and the term (σ / σ_c) as a measure of the wetting characteristics of the packing is listed below.

$$\frac{a_w}{a_t} = 1 - \exp\ [-1.45(Re)^{0.1}\ (We)^{0.2}\ (Fr)^{-0.05}\ (\ \sigma_c/\sigma\ )^{0.75}\ ] .. (2.5)$$

This equation could be utilised conveniently for predicting the values of (a_w/a_t) within ± 20% error.

2.4.2 Correlations for effective interfacial area during physical absorption (a_p) :

Weismannn and Bonilla (100) were the first to estimate the values of effective interfacial area during physical absorption (a_p)

by dividing the values of $k_G a$ (obtained from humidification experiments) by the values of k_G (obtained from evaporation of water) in a column packed with 0.025m R.R. and have proposed the following empirical correlation.

$$(a_p/a_t) = 0.54\, G^{0.31}\, L^{0.07} \tag{2.6}$$

Mada et al. (101) have proposed the following correlation in terms of dimensionless numbers, We and Fr for the data of Shulman et al. (7) and Yoshida et al. (14)

$$(a_p/a_t) = C\,(Fr)^{-0.5}\,(We)^{0.666} \tag{2.7}$$

where C = 0.071 and 0.055 for Raschig ring and Berl saddles respectively.

Generalised correlations in terms of dimensionless numbers for predicting the values of a_p have proposed by Semmelbauer (102), Jackson and Marchello (103) and also Zech and Mersmann (104). Using theoretical considerations based on hydrodynamics of rivulets, Zech and Mersmannn have proposed the following generalised correlation :

$$(a_p/a_t) = 0.0155\,(Re)^{0.5}\,(We/Fr)^{0.45}\,(a_t d_p)^{-0.5} \tag{2.8}$$

Calculating the values of various dimensionless numbers based on hydraulic diameter of packing, Billet and Schultes (105) have recently proposed the following generalised correlation for predicting the values of a_p.

$$(a_p/a_t) = 1.5\,(Re)^{-0.2}\,(We)^{0.75}\,(Fr)^{-0.45}\,(a_t d_h)^{-0.5} \tag{2.9}$$

where d_h = hydraulic diameter of packing.

2.4.3 Correlations for effective interfacial area during chemical absorption (a_c) :

Inspite of the fact that the effective interfacial area during chemical absorption (a_c) have been obtained by numerous investigators during the last two decades, it is rather surprising that hardly any attempt has been made to correlate such data in a systematic manner. Only Puranik and Vogelpohl (106) have presented a generalised correlation in terms of Re, We and (σ / σ_c) for predicting the values of a_c. This generalised correlation is as under :

$$(a_c/a_t) = 1.045\ (Re)^{0.041}\ (We)^{0.133}\ (\sigma / \sigma_c)^{-0.182} \qquad (2.10)$$

This correlation could be utilised conveniently to predict the values of (a_w/a_t). It could also be utilised to predict the values of (a_p/a_t) after subtracting the contribution due to the term (a_{st}/a_t). The equation (2.11) could be used for predicting the values of (a_{st}/a_t).

$$(a_{st}/a_t) = 0.229 - 0.091\ \ln\ (We/Fr). \qquad (2.11)$$

Linek et al. (107), Dharwadkar and Sawant (108) have also proposed some correlations for a_c. However, the applicability of these correlations is restricted to the limited data bank of these investigators.

2.5.0 CORRELATIONS FOR MASS TRANSFER COEFFICIENTS IN PACKED COLUMNS:

2.5.1 Correlations for volumetric liquid side mass transfer coefficient (k_La) :

Different correlations available in the literature for predicting the values of volumetric liquid side mass transfer coefficients (k_La) have been critically reviewed by Au-Yeung and Ponter (109). Some of the important correlations have been listed here.

Sherwood and Halloway (110) have suggested the under mentioned correlation for predicting the values of k_La :-

$$(k_La/D_L) = \alpha \; (L/\mu_L)^{1-n} \; (\mu_L/\rho_LD_L)^{0.5} \qquad (2.12)$$

The values of constants α and n for different packings have also been reported by these investigators.

The existing data on k_La could be correlated satisfactorily within ± 20% error by the following generalised correlation proposed by Norman (111).

$$(k_La/D_L) = 530 \; (L/\mu_L)^{0.75} \; (\mu_L/\rho_LD_L)^{0.5} \qquad (2.13)$$

The generalised correlation proposed by Mohunta et al. (112), specifically applicable to larger diameter columns and different packing sizes, is as under :-

$$k_La \; (a_t \, \mu_L/\rho_Lg)^{0.666} \; (\mu_L/\rho_Lg^2)^{0.111} = 0.0025 \; (\mu_LL^3a_t^{\,3}/\rho_L^{\,4}g)^{0.25} \; (\mu_L/\rho_LD_L)^{-0.5} \qquad \ldots(2.14)$$

In recent years Sahay and Sharma (31,41), also Mangers et al. (43,44) have obtained k_La data for different types and sizes of packings made of different materials like polypropylene, PVC, PTFE, etc. Hence, there is need to analyse this data along with previous data on k_La and to develop a generalised correlation which will take into account the wettabilities of packings.

2.5.2 Correlations for the true liquid side and gas side mass transfer coefficients (k_L, k_G) :

Numerous investigators (45,86,96,97,104,105,113 to 115) have developed correlations to predict the value of true liquid side and gas side mass transfer coefficients (k_L and k_G). The relevant important correlations for predicting the values of k_L and k_G are presented in Table (2.2).

Based on the different correlations reported in Table (2.2), the following observations can be made :-

(i) The values of k_L and k_G can be correlated in terms of dimensionless numbers such as Reynold's number (Re) and Schmidt number (Sc) and other parameters such as (a_td_p), etc.

(ii) With an increase in flow rates (L,G) and Diffusivity (D_L and D_G) and a decrease in viscosity (μ_L and μ_G), the values of k_L and k_G increase.

(iii) With a decrease in packing size and hence an increase in dry surface area (a_t) of packing, the values of k_L decreases, but that of k_G increases.

Table - 2.2

Correlations for k_L and k_G

Correlations for k_L :

Shulman and coworkers (86) :

$$(k_L d_p/D_L) = 25.1\ (d_p L/\mu_L)^{0.45}\ (\mu_L/\rho_L D_L)^{0.5} \qquad (2.2.1)$$

Van Krevelen and Hoftijzer (96) :

$$k_L(\mu_L^2/g\,\rho_L^2)^{0.33}\ D_L^{-1} = 0.015\ (L/a_w\,\mu_L)^{0.66}\ (\mu_L/\rho_L D_L)^{0.33} \qquad (2.2.2)$$

Onda et al. (97) :

$$k_L(\rho_L/g\,\mu_L)^{0.33} = 0.0051(L/a_w\,\mu_L)^{0.66}\ (\mu_L/\rho_L D_L)^{0.33}(a_t d_p)^{0.4} \qquad (2.2.3)$$

Zech and Mersmann (104) :

$$k_L = K^{-1/3}\ (6D/\pi d_p)^{0.5}\ (\rho_L g d_p^2/\sigma)^{-0.15}\ (Lgd_p/3\,\rho_L)^{0.166} \qquad (2.2.4)$$

where $K = 0.0155$ for Raschig rings and $K = 0.022$ for Berl saddles.

Billet and Schultes (105) :

$$k_L = C_L\ (g/\nu_L)^{0.166}\ (D_L/d_h)^{0.5}\ a_t^{-0.33}\ (L/\rho_L)^{0.33} \qquad (2.2.5)$$

where $C_L = 0.412$ for Raschig rings.

Correlations for k_G :

Onda et al. (97) :

$$k_G\ RT/a_t\ D_G = 5.23\ (Re_G)^{0.7}\ (Sc_G)^{0.33}\ (a_t d_p)^{-2.0} \qquad (2.2.6)$$

Tacker and Hougen (113) :

$$(k_G\ P_{BM}\ M_{avg}/G) = m\ (Re_G)^a\ (Sc_G)^{-0.66} \qquad (2.2.7)$$

where $m=1.07$ and $a = -0.41$ for Raschig rings and $Re_G = G\sqrt{a_t}/\mu_G$

Shulman and Delaney (115) :

$$(k_G\ M_{avg}\ P_T/G) = 1.195\ (Re_G)^{-0.36}\ (Sc_G)^{-0.66}\ (P_{BM}/P_T^{-0.66}) \qquad (2.2.8)$$

where $Re_G = d_p G/\mu_G\ (1-\varepsilon)$, P_T is the total pressure.

It is observed that in order to obtain the values of k_L and k_G from the corresponding values of volumetric liquid side and gas side mass transfer coefficients (k_La, k_Ga), the values of wetted surface area (a_w) have been mostly used by these investigators. Thus, the values of k_L and k_G predicted using these generalised correlations are also expected to involve considerable amount of error. Further, the data on k_L and k_G obtained by different investigators (22,36,37,56) using chemical techniques cannot be correlated satisfactorily by these correlations. Hence, there is a need to reobtain the values of k_L and k_G from the data bank of k_La and k_Ga by dividing these values by the appropriate values of 'a'. Hence, there is also a need to analyse both these data banks on k_L and k_G and to develop generalised correlations for predicting the values of k_L and k_G.

2.6.0 DISTILLATION IN PACKED COLUMNS :

Although packed columns are extensively used for gas absorption operations, their application in distillation operation has been rather restricted. That is why the experimental HETP data for distillation in packed columns available in the literature is also limited. The following is the relevant information available in the literature on "HETP" data :-

Bolles and Fair (116) compiled the HETP data on pilot plant scale and presented it in the form of a data bank (117) containing all the relevant information inclusive of physical properties of the different systems.

Billet (118) reported HETP data for the system ethylbenzene-styrene using a column of 0.5m i.d. packed with steel Raschig rings of different sizes (0.015m, 0.025m and 0.035m), also steel Pall rings of sizes (0.015m, 0.025m, 0.035m and 0.05m) with bed heights ranging from 1.5m/2.0m at a pressure of 0.066/0.13 atm a. Further, Billet also reported HETP data for the system ethanol-methanol using steel Pall rings of different sizes ranging from 0.015m to 0.5m in a column 0.5m i.d. packed upto a height of 2.0m.

Clump (119) reported HETP data for the system n heptane-toluene at atmospheric pressure using ceramic Raschig rings of 0.025m size packed in a 0.3m column upto a height of 2.0m.

Kirschbaum (120) reported HETP data for the system ethanol water using a column of 0.4m i.d. packed upto height of 1.0m with ceramic Raschig rings/Berl saddles of 0.025m and 0.035m size at atmospheric pressure. Further, he reported HETP values for the same system at atmospheric pressure using metal Pall rings of 0.025m size in a column of 0.3 m i.d. packed upto height of 2.0m.

Silvey (121) reported HETP distillation data for the system cyclohexane - n heptane at a pressure of 1.6 atm a using ceramic Raschig rings of 0.038m and 0.075m size packed in a column of 1.2m i.d. with packed height ranging from 4m to 10.5m.

2.7.0 SUMMARY :

(I) Considerable experimental information is available in the literature on interfacial areas and mass transfer coefficients.

This information could be utilised to prepare the Data Banks for : (i) Wetted surface area, (ii) Effective interfacial area for physical absorption, (iii) Effective interfacial area for chemical absorption, (iv) Volumetric liquid side mass transfer coefficient, (v) Overall gas side mass transfer coefficient, (vi) Volumetric gas side mass transfer coefficient and (vii) True liquid side mass transfer coefficient.

(II) Further, for designing a packed column , one requires knowledge of the following design parameters :- (i) Flooding velocity (capacity limit), (ii) Pressure drop, (iii) Holdup and (iv) Volumetric gas and liquid side mass transfer coeffcients. Further, for the case of gas absorption accompanied by a chemical reaction, one also requires the knowledge of true liquid side mass transfer coefficient (k_L) and effective interfacial area for mass transfer (a) separately. The information available in the literature on flooding velocity (capacity limit), pressure drop and liquid holdup etc. is adequate. These design parameters could also be predicted satisfactorily by the use of generalised correlations cited in this chapter.

(III) There exists considerble amount of discrepancy regarding the values of effective interfacial area and wetted surface area. In particular the value of interfacial area during physical absorption (a_p) and absorption with chemical reaction (a_c) are not at all comparable under otherwise similar conditions. Further, the values of wetted surface area have been mostly used by many investigators to obtain the values of the gas side and liquid side mass transfer coefficients (k_G and k_L respectively) from the corresponding values of volumetric mass transfer coefficients. Thus, the predicted values of k_L and k_G are expected to involve considerable amount of error.

(IV) The existing generalised correlations available in the literature for prediciting the values of a_w, a_c, a_p, k_La, k_L and k_G appear to have some drawbacks/limitations.

(V) There is need to reanalyse the data on k_La and to develop a generalised correlation which will take into account the wettabilty of packing materials (σ_c).

(VI) Very few of the existing generalised correlations available in the literature for predicting the values of interfacial areas and mass transfer coefficients are able to elucidate the mechanism of mass transfer during gas absorption in packed columns.

(VII) No correlation is available in the literature for predicting the values of $k_L'a$ - volumetric liquid side mass transfer coefficient when absorption is accompanied by a chemical reaction.

(VIII) Information available about the applicability of generalised correlations developed for absorption in packed columns to other similar operations like distillation is very limited.