Liquid-Side Mass Transfer Coefficients in Packed Towers

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The physical absorption of gas by water in a tower packed with Raschig rings has been investigated. The liquid-side mass transfer coefficient which was separated by dividing the capacity coefficient by the wetted surface area is discussed from the standpoints of the two-film and penetration theories. A new and simpler dimensionless group is presented which correlates about 90% of the data reported, including the author's own, within an accuracy of ±20%.

The relation of the liquid-side mass transfer coefficient to packed-tower performance and physical properties of the system is important from the standpoint of packed-tower designs and interesting from the viewpoint of the mechanism of mass transfer.

Many investigators (*7, 15, 20, 22*) have been studying the relation since Sherwood's (*16*) experimental equation was reported, but up to the present time the results seem to be far from satisfactory.

In this paper physical gas absorption by water in a tower packed with Raschig rings is described, and k_L is discussed. The relation of k_L, which covers about 90% of the data reported, including the authors', within a 20% accuracy, was obtained.

EXPERIMENTS AND RESULTS

The absorption of pure carbon dioxide by water was studied. To ascertain the exponent of the Schmidt number, the absorption of pure hydrogen was also studied. Because the degree of purity of the gas was more than 99%, the gas-side resistance was regarded as negligible in comparison with the liquid-side resistance. Figure 1 is the flow sheet of the equipment. The packed tower consists of a single glass cylinder about 80 cm. (2.6 ft.) long, with an inside diameter of 6.0 cm. (2.4 in.), packed to a height of 30 cm. (1 ft.) with 6- (¼-in.), 8- (⅓-in.) and 10-mm. (2/5-in.) ceramic Raschig rings. The wet packing method was used to pack Raschig rings. Tap water was introduced from the head tank into the tower through the thermostat.

The distributor was constructed as follows: Seven glass tubes with an inside diameter of 4 mm. (⅙ in.) were arranged in a position of an equilateral triangle. The lower end of each distributor tube was set as closely as possible on the top of the packing to avoid upper end effects.

The mass velocity of the liquid was ascertained by the measurement of the quantity of water flowing from the tower bottom per unit time. The termperatures at the top and bottom of the tower were almost the same and were maintained at 25 ± 1°C.

To measure the end effect samples were taken from the lower tower end and the funnel which was placed immediately below the support plate of the packing.

For carbon dioxide a sample of 50 cc. was introduced into 0.2N barium hydroxide solution and then back titrated with 0.1N hydrogen chloride solution. The end effect for 6-, 8-, and 10-mm. Raschig rings was equivalent to a packing height of 3.6, 3.5, and 3.1 cm. respectively.

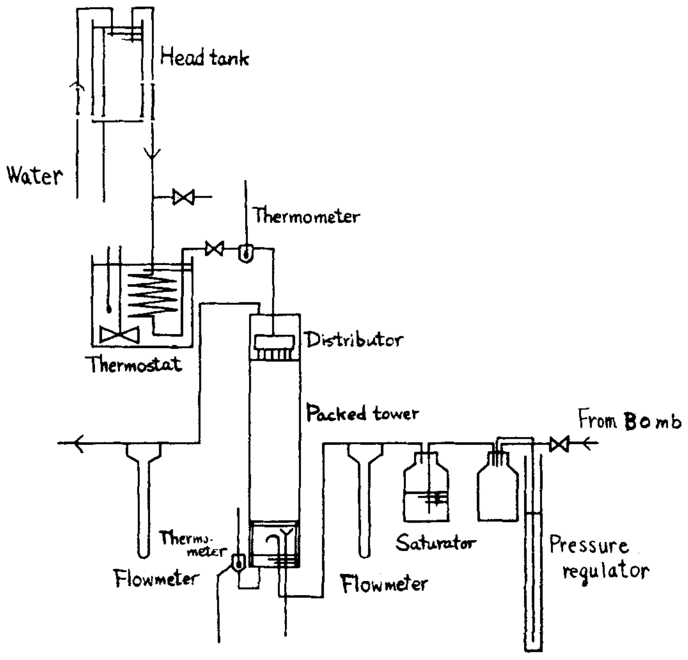

Fig. 1. Schematic diagram of the absorption system.

For hydrogen a sample of 500 cc. was withdrawn by Swanson and Hulett's method (*21*) and analyzed by the explosion method.

As stated above, the gas used was of more than 99% purity, and therefore the gas-side resistance could be assumed to be negligible. The liquid-side capacity coefficient can consequently be computed from

$$k_L a = \{L/(\rho Z)\}\{\ln (C_s - C_1)/(C_s - C_2)\} \quad (1)$$

The saturated concentration C_s at 25°C. was taken from the International Critical Tables*.

The results for carbon dioxide are shown in Figures 2, 3, and 4. In these cases it is known by previous reports (*11, 16, 24*) that the gas velocity has no relation to the liquid-side capacity coefficient under its loading point, and here it was about 50 ∼ 150 kg./(sq. m.)(hr.) [11 ∼ 31 lb./(sq. ft.)(hr.)].

To test the dumped packing method the tower was repacked with 6-mm. Raschig ring, and the capacity coefficient was measured. The reproducibility was good (Figure 2).

The capacity coefficient increases linearly with the increase of liquid velocity; that is,

$$k_L a = c' L^{m''} \quad (2)$$

where for 6-, 8-, and 10-mm. Raschig rings c' and m'' were almost the same; c' = 0.047, and m'' = 0.72. These results agree with those of previous investigators (*16, 25*).

The results for hydrogen will be described later.

DERIVATION OF THE EQUATION OF k_L

It is obvious that k_L and a depend on packed-tower performances and physical

*Tabular material has been deposited as document 5874 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for $2.50 for photoprints or $1.75 for 35-mm. microfilm.

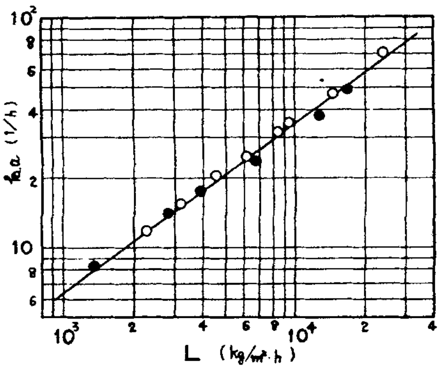

Fig. 2. Relation of k_La vs. L in carbon dioxide absorption by water; ● 6-mm. ceramic Raschig ring repacked.

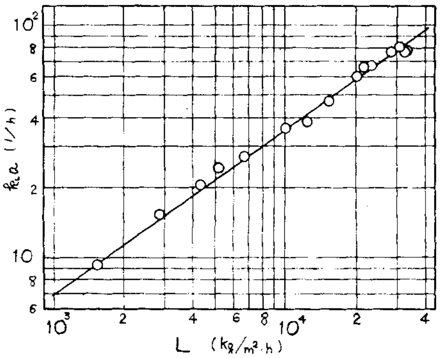

Fig. 3. Relation of k_La vs. L in carbon dioxide absorption by water, 8-mm. ceramic Raschig ring.

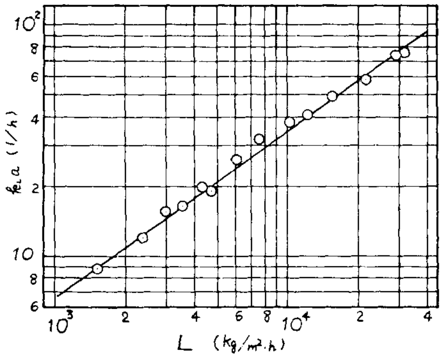

Fig. 4. Relation of k_La vs. L in carbon dioxide absorption by water, 10-mm. ceramic Raschig ring.

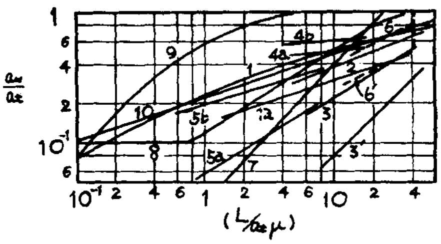

Fig. 5. Relation of a_w/a_t vs. $L/a_t\mu$ by various investigators: 1. Fujita and Sakuma (*3*), 1a. Niwa and Hashimoto (*12*), 2. Sherwood and Pigford (*18*), *3* and *3'* Shulman and DeGouff (*19*), *4a*, *4b*, *5a*, *5b* Weisman and Bonilla (*26*), *6* and *6'* Mayo, Hunter and Nash (*10*), 7. Pratt (*14*), 8. Grimley (*4*), 9. van Krevelen, *et. al.* (*22*), and 10. Hikita and Kataoka (*6*)

properties of fluids in different ways, because the mass transfer coefficient depends on the diffusion coefficient and film thickness or contact time, but the contact area does not.

To separate k_L from k_La it is necessary to evaluate a, but at present a is a very obscure value. For this reason it was assumed that a is proportional to a_w. If it is possible within a reasonable error to correlate k_L separated by a_w instead of by a, it will be more convenient. The replacement of a by a_w means that $a = ka_w$. This assumption has been used throughout the paper to separate k_L from k_La and has proved its usefulness within reasonable error, at least for Raschig rings and water. Many investigators have presented the formulas for a_w (Figure 5). Among these formulas Fujita's (*3*) was selected because its form is more reasonable, and its validity for water has been confirmed by Hikita (*6*). Fujita's formula is

$$a_w/a_t = 1 - 1.02e^{-0.278(L/a+\mu)^{0.4}} \qquad (3)$$

The total surface area for Raschig ring was computed from $a_t = 4.7/D_p$ derived from Figure 6.

Derivation of the formula from the standpoint of two-film theory

Basic concept of k_L

From the dimensional analysis and the analogy of the heat transfer the following equation was proposed for the gas absorption in packed towers:

$$Nu' = \frac{k_L[L']}{D_L} = c(Re)^m(Sc)^n \qquad (4)$$

For this dimension of length previous investigators have proposed various lengths, for example, the tower diameter (*7*) and the diameter of a sphere having the same surface area as a packing (*20*), and have eliminated L' by Galilei's number (*15*, *22*). The correlations with various variables however are not always good.

This dimensionless group was interpreted as follows. According to the two-film theory the mass transfer coefficient for the liquid film, is equal to D_L/x; consequently k_L/D_L should be a reciprocal of the effective film thickness. If the dimension of length in the modified Nusselt number is chosen as a length related to x, the dimensionless group $(k_L[L']/D_L)$ will be more reasonable.

From this principle, operating holdup divided by wetted surface area was first chosen as L', and the modified Nusselt number became

$$Nu' = k_Lh/(D_La_w) \qquad (5)$$

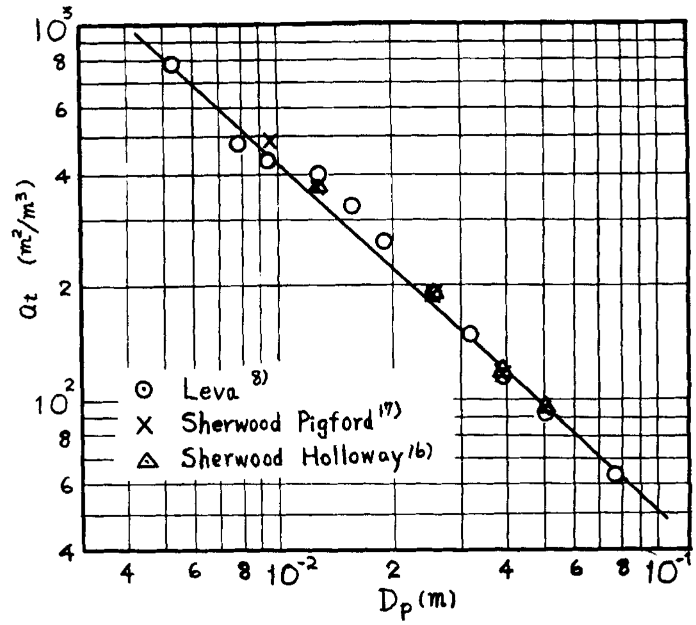

Fig. 6. Estimation of a_t vs. D_p by various investigators.

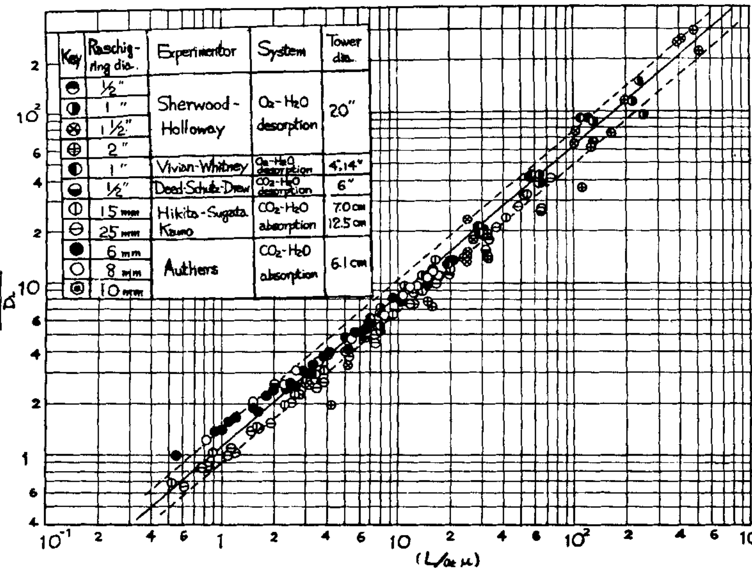

Fig. 7. Relation of $k_L(h/a_w)/D_L$ vs. $L/(a_t\mu)$; two broken lines indicate the region of ±20% error.

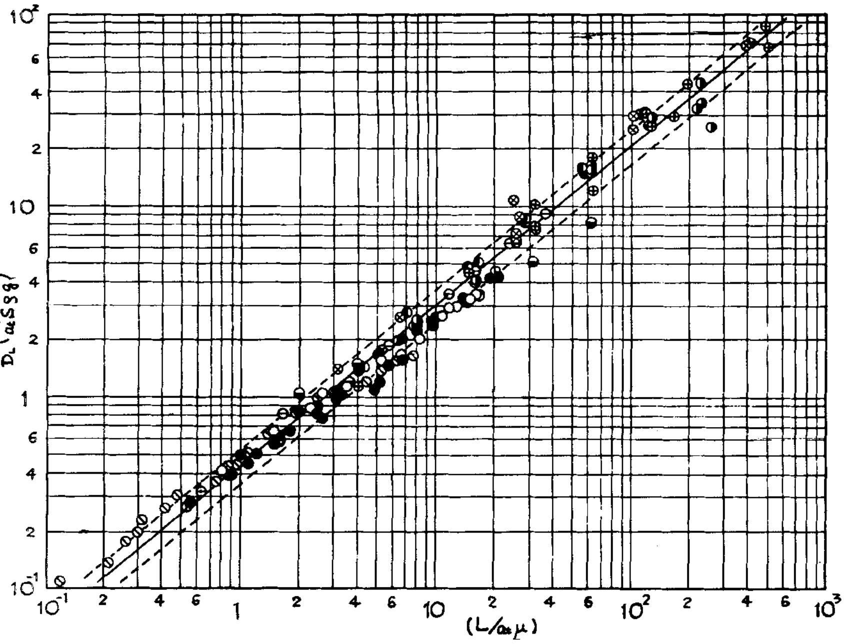

Fig. 8. Relation of $k_L(\mu V/\rho g a_t S)^{1/3}/D_L$ vs. $L/(a_t\mu)$. Keys are the same as those in Figure 7. Two broken lines indicate the region of ±20% error.

The operating holdup was calculated from the experimental equation of Ôtake (*13*) and a_w was calculated from Fujita's formula (*3*) as described above. Ôtake's equation is

$$h = 1.295(D_pL/\mu)^{0.676} \cdot (D_p^3 g\rho^2/\mu^2)^{-0.44}(a_tD_p) \qquad (6)$$

This equation is only partially correct, and moreover it is very inconvenient to calculate h and a_w from their experimental equations at every point.

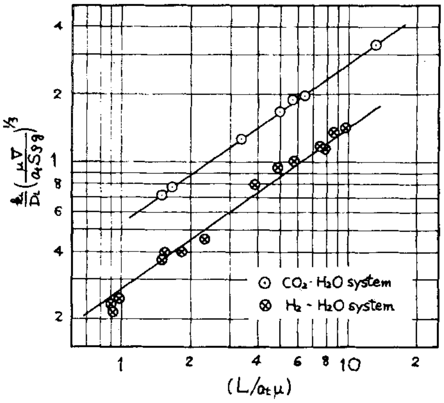

Fig. 9. Relation of $k_L(\mu V/\rho g a_t S)^{1/3}/D_L$ vs. $L/(a_t\mu)$ in the absorption system of carbon dioxide–water and hydrogen–water at 25°C.; tower diameter is 6 cm. and packed with 6-mm. Raschig ring to the height of 30 cm.

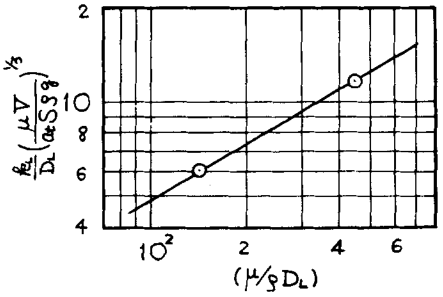

Fig. 10. Relation of $k_L(\mu V/\rho g a_t S)^{1/3}/D_L$, vs. $\mu/(\rho D_L)$ for the data in Figure 9 at constant $N_{Re} = 3$.

Name	Packing height	Tower diameter, in.
Sherwood-Holloway	6 ~ 49 in.	20
Vivian-Whitney	2 ~ 8 ft.	4 and 14
Hikita *et al.*	30 cm. (1 ft.)	2.8 and 5 (7 and 12.5 cm.)
The authors	30 cm. (1 ft.)	2.4 (6 cm.)

Name	Liquid velocity	Gas velocity
Sherwood-Holloway	250 ~ 32,000 lb./(sq. ft.)(hr.)	36 ~ 1,320 lb./(sq. ft.(hr.)
Vivian-Whitney	970 ~ 16,000 lb./(sq. ft.)(hr.)	60 ~ 120 lb./(sq. ft.)(hr.)
Hikita *et al.*	380 ~ 70,000 kg./(sq. m.)(hr.)	20 ~ 80 kg./(sq. m.)(hr.)
	78 ~ 14,300 lb./(sq. ft.)(hr.)	4 ~ 16 lb./(sq. ft.)(hr.)
The authors	1,500 ~ 35,000 kg./(sq. m.)(hr.)	50 ~ 150 kg./(sq. m.)(hr.)
	310 ~ 7,200 lb./(sq. ft.)(hr.)	11 ~ 31 lb./(sq. ft.)(hr.)

To avoid such inconvenience, the depth of the thin water layer flowing over packings was considered. Generally, the depth of thin layer flowing down over a plain plate inclining θ' toward the horizontal plane is derived theoretically as

$$D = [3\mu V/(B\rho g \sin\theta')]^{1/3}$$

The breadth of the plate may be considered as a_wSZ (sin θ'')/Z, because $a_wSZ \sin\theta''$ is the perpendicular projection of the total wetted surface area in the tower.

The following formula is gained:

$$D = [3\mu V/a_w\rho g S (\sin\theta')(\sin\theta'')]^{1/3}$$

Putting a_t instead of a_w and omitting $(\sin\theta')(\sin\theta'')$, one gains the following modified Nusselt number:

$$Nu' = k_L[\mu V/(a_t\rho g S)]^{1/3}/D_L \qquad (7)$$

where D_L for carbon dioxide is calculated by the formula of Wilke and Chang (*27*). When one uses a_t instead of a_w and omits $(\sin\theta')(\sin\theta'')$, a hypothetical depth of viscous flow over packings is taken in the same way as if $L/a_t\mu$ were used as the Reynolds number of a liquid. If a_w can be replaced by a_t to correlate k_L within reasonable error, it will be much more convenient, and this paper will have proved its effectiveness. By this replacement one could correlate k_L in the same equation, as described later.

Dependence of Equations (5) *and* (7) *on Reynolds number*

The modified Nusselt number in Equation (5), $k_Lh/(D_La_w)$ vs. $L/(a_t\mu)$, is plotted in Figure 7 for the authors' experimental data as well as those of Sherwood-Holloway (*16*), Vivian-Whitney (*24*) and Hikita *et al.* (*5*). The ranges of the conditions are as follows:

The agreement of the data in the ±20% error region however is rather poor; moreover it is most inconvenient to calculate h and a_w from experimental Equations (3) and (6), as mentioned above.

From Equation (7) $k_L[\mu V/(\rho g a_t S)]^{1/3}/D_L$ vs. $L/(a_t\mu)$ is plotted in Figure 8 with the same data used as in Figure 7. In Figure 8 the agreement of the data is fair, and the values are easy to calculate. The straight line in Figure 8 represents

$$k_L[\mu V/(\rho g a_t S)]^{1/3}/D_L = 0.44[L/(a_t\mu)]^{0.82} \qquad (8)$$

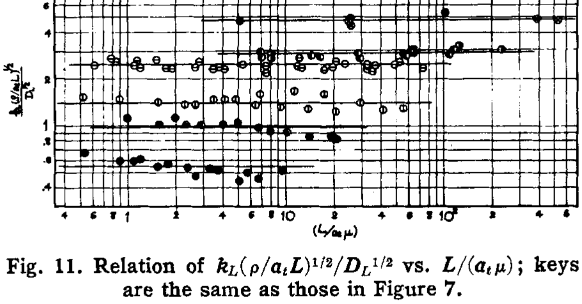

Fig. 11. Relation of $k_L(\rho/a_tL)^{1/2}/D_L^{1/2}$ vs. $L/(a_t\mu)$; keys are the same as those in Figure 7.

Dependence of Equation (7) *on the Schmidt number*

The exponent of the Schmidt number in Equation (4) has been discussed frequently by many investigators. Some of them (*11, 16, 20, 25*) suggest $n = 1/2$, but others (*7, 22*) support the value $n = 1/3$. To decide which exponent is right, the absorption of hydrogen by water was carried out in the same apparatus as described above with 6-mm. Raschig rings.

D_L for hydrogen, a correction of the value used in Perry's Hand Book, was selected; that is, $D_L = 6.30 \times 10^{-5}$ sq. cm./sec. at 25°C. $N_{Sc} = 143$. The experimental data are plotted in Figure 9; the ordinate $k_L[\mu V/(\rho g a_t S)]^{1/3}/D_L$, and the abscissa is $L/(a_t\mu)$.

The data for carbon dioxide and hydrogen in Figure 9 were calculated from the value at the tower end, because for hydrogen it was very difficult at a low Reynolds number to take the sample of 500 cc. from the funnel placed immediately below the support plate.

From Figure 9 the values of $k_L[\mu V/(\rho g a_t S)]^{1/3}/D_L$ vs. Schmidt number at $N_{Re} = 3$ were plotted in Figure 10. (As the two lines in Figure 9 are almost parallel, the value of the Reynolds number is indifferent.) The slope of the line in Figure 10 is 0.58; it is clear that the exponent of $N_{Sc} = 1/2$ is preferable.

Results

The correlation of k_L for the Reynolds and Schmidt numbers becomes

$$k_L[\mu V/(\rho g a_t S)]^{1/3}/D_L = 0.021[L/(a_t\mu)]^{0.82} \cdot [\mu/(\rho D_L)]^{1/2} \qquad (9)$$

Dividing both sides of Equation (9) by $(Re)^{1/3}$, one obtains

$$k_L[\mu^2/(\rho^2 g)]^{1/3}/D_L = 0.021[L/(a_t\mu)]^{0.49} [\mu/(\rho D_L)]^{1/2} \qquad (10)$$

The left-hand side of Equation (10) is the modified Sherwood number named by Krevelen (*22*).

By dividing both sides of Equation (10) by the Schmidt number, one obtains

$$k_L[\rho/(\mu g)]^{1/3} = 0.021[L/(a_t\mu)]^{0.49} \cdot [\mu/(\rho D_L)]^{-1/2} \qquad (11)$$

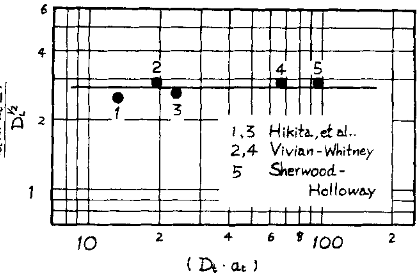

Fig. 13. Relation of $k_L(\rho/a_tL)^{1/2}/D_L^{1/2}$ vs. (D_ta_t) at constant (Galilei) (D_p = 1 in.)

The exponent of the Reynolds number in Equations (10) and (11) is in accord with that of wetted-wall (*1, 9*) and liquid-rod (*23*) experiments.

Derivation of the formula from the standpoint of the penetration theory

Basic concept of k_L

In the packed tower, where the two fluids flow countercurrently, the model of the two-film theory can hardly be understood, even though the interface of the fluids is modified as an effective one.

The penetration theory or its modification, the surface renewal theory, seems to be preferable to the two-film theory. By Higbie's penetration theory k_L can be described as

$$k_L = 2[D_L/(\pi\theta)]^{1/2} \qquad (12)$$

Using an idea similar to that described in the two-film theory one can consider the following dimensionless formula:

$$k_L[T]^{1/2}/D_L^{1/2} = [\text{Dimensionless}] \qquad (13)$$

If T is taken to have a closer connection with θ in Equation (13), a more reasonable dimensionless group can be obtained. However since the phenomena in the packed tower is very complex, a simplified model can hardly be presented; therefore the dimensional analysis has been used.

It may be considered reasonable that the time during which the elementary surface of the liquid is exposed to the gas depends upon the operating conditions, the physical properties of the liquid, and the characteristics of the tower and packings; it can be assumed as

$$\theta = f(\rho, \mu, g, D_t, a_t, L) \qquad (14)$$

By dimensional analysis various combinations of dimensionless groups were obtained, and from the many results reported the next relation was selected.

$$(1/\theta)[\rho/(a_tL)] = c'[L/(a_t\mu)]^{m'} \cdot [\rho^2 g/(\mu^2 a_t^3)]^{n'}(D_ta_t)^{p'} \qquad (15)$$

The first term of the right-hand side of Equation (15) is the Reynolds number based on the total area of packings in unit volume, and the second term is Galilei's number. This latter number was introduced in Equation (15) by the consideration of the dependence of gravity acceleration. The third term refers to dimensions of tower diameter and packing piece.

From Equations (13) and (15) the following is obtained:

$$k_L[\rho/(a_tL)]^{1/2}/D_L^{1/2} = c[L/(a_t\mu)]^m[\rho^2 g/(\mu^2 a_t^3)]^n \cdot (D_ta_t)^p \qquad (16)$$

The constants and the exponents of Equation (16) will now be determined from the experimental data and the results reported by others.

Dependence of Equation (16) *on the Reynolds number*

The exponent of the Reynolds number in Equation (16) was determined by the authors' experimental data and those of Sherwood-Holloway (*16*), Deed-Schutz-Drew (*2*), Vivian-Whitney (*24*), and Hikita *et al.* (*5*) given in Figure 11. The range of the Reynolds number is from 0.5 to 500, and it is found that the exponent $m = 0$. In Figure 11 some of the experimental data are omitted to avoid confusion. (All data of each experiment are plotted in Figure 14.

Dependence of Equation (16) *on Galilei's number*

To determine the exponent of Galilei's number the values of $k_L[\rho/(a_tL)]^{1/2}/D_L^{1/2}$ at constant Reynolds number were plotted against each Galilei's number (Figure 12). From the slope of the plotted line the exponent of Galilei's number = 0.38. The range of Galilei's number is from 2.5×10^4 to 1.1×10^7.

Dependence of Equation (16) *on the tower diameter*

From the data of Sherwood-Holloway (*16*) (tower diameter 20 in.), Vivian-Whitney (*24*) (tower diameter 14 and 4 in.), and Hikita *et al.* (*5*) (tower diameter 12.5 and 7 cm.) the values of $k_L[\rho/(a_tL)]^{1/2}/D_L^{1/2}$ for 1-in. Raschig ring

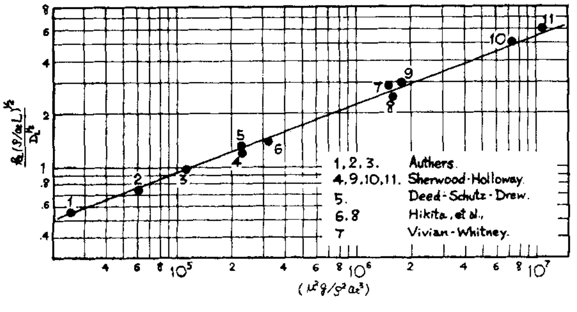

Fig. 12. Relation of $k_L(\rho/a_tL)^{1/2}/D_L^{1/2}$ vs. (Galilei) for the data in Figure 11 at constant Reynolds.

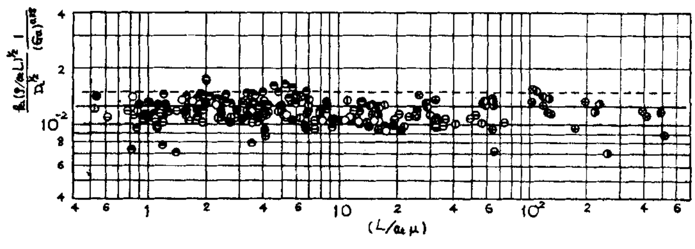

Fig. 14. Relation of $k_L(\rho/a_tL)^{1/2}/(D_L^{1/2}Ga^{0.38})$ vs. $L/(a_t\mu)$ for the data reported which is thought to be purely physical; keys are the same as those in Figure 7.

are plotted against the values of $(D_t a_t)$ in Figure 13. From this figure it is clear that the exponent of $(D_t a_t)$, p, equals 0.

Results

All results are given in Figure 14, where almost all the data of each experiment reported are plotted; over 90% of the results lie within a ±20% error.

From the results discussed above Equation (16) becomes

$$k_L[\rho/(a_t L)]^{1/2}/D_L^{1/2} = 0.013[\rho^2 g/(\mu^2 a_t^3)]^{0.38} \quad (17)$$

The left-hand side of Equation (16) is the result of the use of $\rho/(a_t L)$ as the dimension of time in Equation (13), but the meaning of $\rho/(a_t L)$ can be considered as $L/\rho = u$, and $a_t = 4.7/D_p$; then $\rho/(a_t L) \propto D_p/u$; that is, $\rho/(a_t L)$ is proportional to the apparent mean residence time over a packing.

RESULTS FROM THE TWO THEORIES

The exponent of Galilei's number in Equation (17) may be assumed to be ⅓; then Equation (17) becomes

$$k_L[\rho/(\mu g)]^{1/3} = 0.013[L/(a_t\mu)]^{1/2} \cdot [\mu/(D_L\rho)]^{-1/2} \quad (18)$$

The coincidence of Equation (11) and Equation (18) is most satisfactory, and the correlation of k_L from both theories becomes

$$k_L[\rho/(\mu g)]^{1/3} = c[L/(a_t\mu)]^{1/2} \cdot [\mu/(\rho D_L)]^{-1/2} \cdot c = 0.01 \sim 0.02 \quad (19)$$

The agreement of ±20% in Figure 14 corresponds to a value of c of 0.013.

The exponent of the Reynolds number in Equation (19) is in accord with those found in the experiments by wetted wall (*1, 9*) and by liquid rod (*23*), and this seems to have an interesting significance for the investigation of the mechanism of gas absorption in packed towers; moreover these coincidences indicate that the assumption made in this paper: $(a = ka_w)$ is not only convenient but also reasonable. Here the proportionally constant k is independent of L and G. If a were a function of G under its loading point, it is most natural that $k_L a$ would also be a function of G, but all the data reported prove that $k_L a$ is independent of G(*11, 16, 24*).

SUMMARY

1. The absorption of carbon dioxide and hydrogen by water in the tower packed with Raschig rings was investigated.

2. It was proved within an accuracy of ±20% that k_L is a function of the total surface area, as shown in Equations (11) and (18).

3. From the standpoint of the two-film theory a new modified Nusselt number is presented which takes the apparent mean depth of liquid as a dimension of length, and this number covers about 90% of the data reported, including the authors', within an accuracy of ±20%.

4. From the standpoint of the penetration theory a new and simpler dimensionless formula is presented in Equation (17) which was applicable within an accuracy of ±20% to almost all the data reported, as shown in Figure 14.

5. From both theories it is found that k_L depends on the Reynolds and Schmidt numbers, as shown in Equation (19).

The exponent of the Reynolds number is in accord with that found in the experiments by wetted wall and liquid rod; it is believed reasonable that a is proportional to a_w, where the proportional constant is independent of L and G within an accuracy of ±20%, at least for Raschig rings and water.

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NOTATION

a = effective surface area of packings, sq. m./cu. m.
a_t = total surface area of packings, sq. m./cu. m.
a_w = wetted surface area of packings, sq. m./cu. m.
B = breadth of a plate, m.
c, c' = a constant
C_1, C_2, C_s = concentration of liquid at the entrance, the exit of the tower, and at the saturation, respectively, kg./m³.
D = depth of thin layer of viscous flow, m.
D_L = diffusional coefficient of solute gas, sq. m./hr.
D_p = nominal diameter of a packing, m.
D_t = diameter of the tower, m.
G = mass flow rate of gas, kg./(sq. m.)(hr.)
g = gravity acceleration, m./hr.²
h = operating holdup.
k = proportional constant independent of gas and liquid velocity
k_L = liquid-side mass transfer coefficient, m./hr.
L = mass flow rate of liquid, kg./(sq. m.)(hr.)
L' = length, m.
m, m' = exponent of the Reynolds number
m'' = exponent of liquid velocity
n, n' = exponent of the Schmidt number
Nu' = modified Nusselt number, $k_L[L']/D_L$
p, p' = exponent of $(D_t a_t)$
S = sectional area of empty tower, sq. m.
T = time, hr.
u = mean linear velocity in the tower, m./hr.
V = volumetric flow rate of liquid, cu. m./hr.
x = effective film thickness, m.
Z = height of packing layer, m.

Greek Letters

μ = viscosity of liquid, kg./(m.)(hr.)
ρ = density of liquid, kg./cu. m.
θ = time during which an element of the liquid surface is exposed to the gas, hr.
θ', θ'' = inclination angle of a plate to the horizontal
π = ratio of circumference of a circle to its diameter

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