

Mass and Heat Transfer in a Falling Liquid Film of Wetted Wall Tower

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Part I Mass Transfer

1. Introduction

Many investigations have been made, theoretically and experimentally, about the mass transfer rate in a falling liquid film, in the case of gas absorption or gas desorption with a wetted wall tower. Most of the results of experimental works deviated considerably from the theoretical values and different interpretations were given about these results by each investigator. Previously the authors explained the flow of a liquid film, measuring its thickness. From this it is surmised that the flow state has an important effect on the mass transfer rate in the liquid film. The present paper deals with the experiment of carbon dioxide gas absorption into water and the absorption rate is discussed from the above-mentioned point of view.

2. Previous Investigations

Hatta³⁾ and Pigford¹⁰⁾ analysed the absorption rate of stationary gas into a falling liquid film. They showed that the absorption rate which is expressed in term of $(H)_{LM}/l$ becomes a function of the following parameter ζ_M only, when the true molecular diffusion occurs in a perfect laminar liquid film that accords to the theory of Nusselt⁹⁾.

$$\zeta_M = \frac{4}{9} \eta^2 \frac{D_L l}{B^2 u_{LS}}, \quad (1)$$

where

$$\eta = \frac{u_{LS}}{u_{Lm}} \quad (2)$$

In a perfect laminar flow

$$\eta = \frac{3}{2} \quad (3)$$

and

$$B = \left(\frac{3\mu L^2}{4g\rho L^2} \right)^{1/3} Re_L^{1/3} \quad (4)$$

Substituting these values in Eq. (1) gives

$$\zeta_M = 2.92 \frac{lg^{1/3} D_L \mu L^{5/3}}{Re_L^{4/3} \mu L^{5/3}} \quad (5)$$

In Fig. 1 which shows the relation between $(H)_{LM}/l$ and $1/\zeta_M$, the curve P represents the theoretical result obtained by Pigford and the curve H by Hatta, and these two curves are very near to each other. With increase of ζ_M , $(H)_{LM}$ becomes independent of l as shown by the straight line A and with decrease of ζ_M , $(H)_{LM}$ becomes approximately proportional to \sqrt{l} as shown by the straight line B.

When the liquid film flows in a fully turbulent state or the gas flows counter-currently to the film, the analysis for it has not been conducted.

A number of experimental results of mass transfer rate have been published and some of them are shown in Fig. 1. Hikita et al.⁵⁾, treating the absorption of carbon

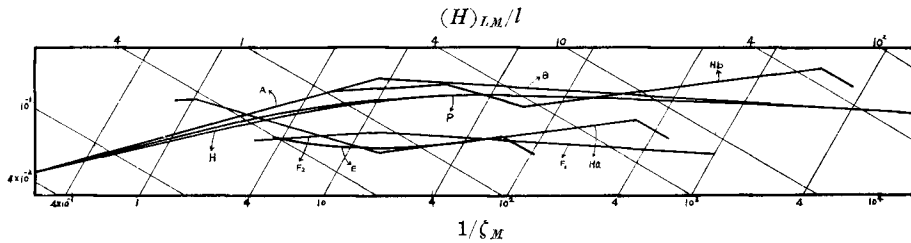


Fig. 1. Previous investigations of gas absorption in falling liquid film.

dioxide gas by water, gave the curves Ha and Hb for the the two experiments in which the value of l was different. It is, therefore, seen in their investigation that the observed value of $(H)_{LM}/l$ is not a function of ζ_M only. The straight lines F_1 and F_2 were obtained by Fujita et al.²⁾ in the similar experiment. Their values of $(H)_{LM}/l$ can be expressed as a function of ζ_M only, but are much smaller than the theoretical values. Emmert et al.¹⁾ investigated the absorption of carbon dioxide gas and oxygen gas by water and the desorption of these absorbed gases from absorbent, and drew the experimental curve E which was also far down from the theoretical line P or H. They attributed the discrepancy of theoretical and observed values to the presence of turbulence in the falling liquid film, but they did not give the quantitative considerations.

In these experimental studies all the observed values of absorption rate are 1.5 to 3.5 times as large as the theoretical values, which are computed on the basis of the true molecular diffusion in a perfect laminar liquid film. But according to the

study of liquid hold-up in a wetted wall tower accomplished formerly by the authors⁷⁾, the falling liquid film at low value of Re_L is not in a perfect laminar state except at very small value of Re_L , but it has a pseudo-laminar flow in which ripples appear on the surface of film. Under such conditions the true molecular diffusion may not be effected, and so the theory of Hatta or Pigford cannot be expected to be applied.

3. Apparatus and Procedure

Fig. 2 is a schematic diagram of the apparatus used. The test section of the tower was made of a glass tube, 4.76 cm inner diameter and 250 cm long. The feed water were at room temperature or heated. The gas used was pure carbon dioxide gas in commerce. The gas was stagnant or it flowed countercurrently to the liquid film. The fresh gas was blown continuously into the system from the gas bomb in order to keep the pressure of the system slightly higher than the barometric pressure. The vaporization of water from the film into the gas phase was prevented by passing the gas through the steam saturator.

The composition of gas in the system changed from 92 to 100% in dry basis on account of the impurity of the commercial gas and the stripping of other gases contained in water. The temperature of the system was regulated at 8.5, 14, 25, 35

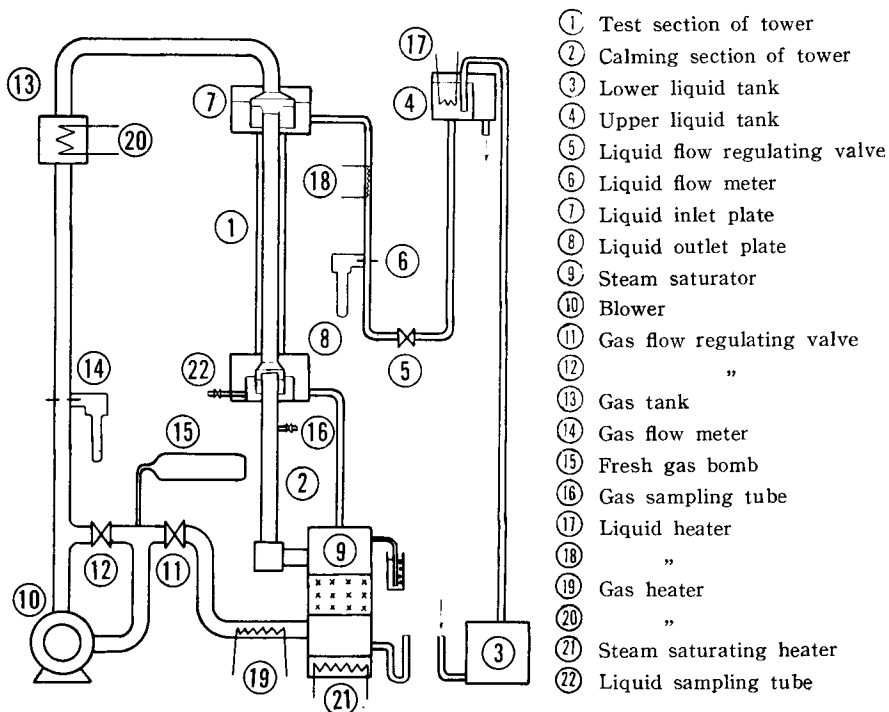


Fig. 2. Apparatus

or 50°C. Re_L varied from 55 to 11,000, and maximal Re_G was 27,800, when the linear gas velocity was 465 cm/sec. The absorption rate was determined by measuring the concentration of the exit liquid. A certain volume of the liquid sample was added to plenty of baryta water and the excess of baryta was titrated with hydrochloric acid by means of the conductometric titration method.

4. Results and Discussions

(1) In the case of stagnant gas

In Fig. 3 the observed values of $(H)_{LM}/l$ are plotted against Re_L at each temperature. The curve P gives the theoretical value by Pigford at respective temperature. Above 25°C the exit liquid was almost saturated with the gas when the liquid flow rate was low, and therefore the data at low value of Re_L became inaccurate.

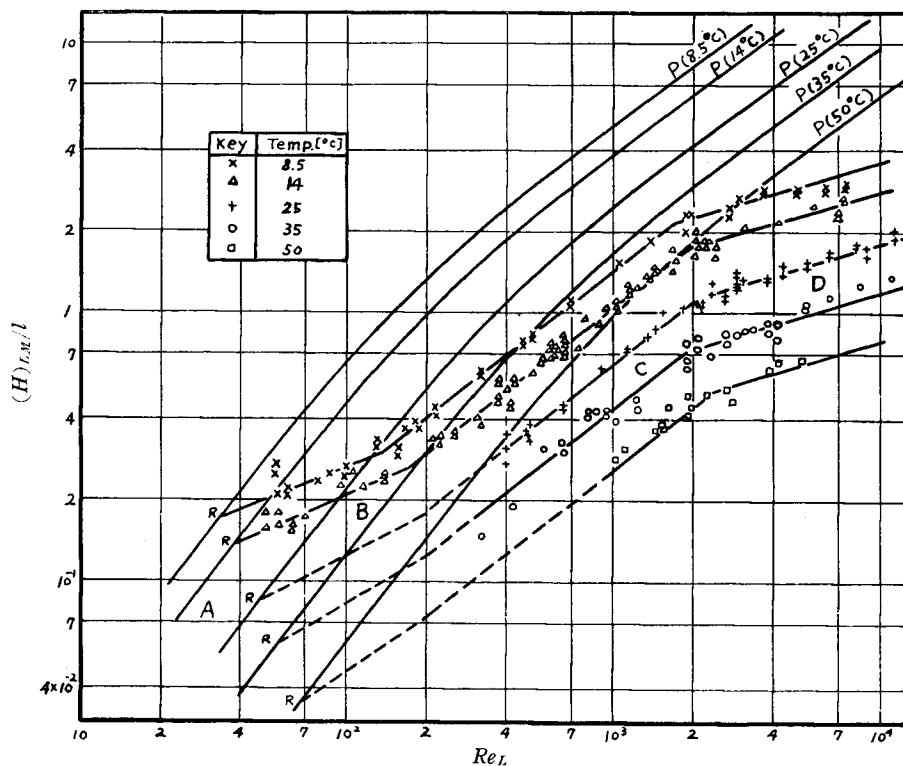


Fig. 3. Observed values of $(H)_{LM}/l$.

The observed values are fairly smaller than the theoretical at every temperatures as in the previous investigations. The plotted points at 8.5 and 14°C are divided into three parts B, C and D and above 25°C into two parts C and D, according to the range of Re_L . The part B corresponds to the region below 150 of Re_L , the part

C to the region between 150 and 2,000, the part D to the region above 2,000, and the plotted points seem to lie on a straight line in each part. When the point of intersection of the curve P and the straight line B is denoted by R, the value of Re_L of the point R is 34 at 8.5°C and 38 at 14°C. These values are equal to the values of Re_{LC1} , which are the critical Re_L between the perfect laminar flow region and pseudo-laminar one. From this fact it can be said that the discrepancy between the theoretical and the observed values of $(H)_{LM}/l$ occurs together with the transition of the flow from perfect laminar to pseudo-laminar and the parts B and C correspond to the pseudo-laminar region. The theory may hold good in the perfect laminar region, although the data were not obtained in this region. The part that agrees with the theory is denoted by A. The part D corresponds to the fully turbulent region. It may also be true above 25°C. Such characteristic of the variation of the absorption rate is also recognized in the study of absorption of hydrogen gas by Hikita et al.⁴⁾

It is certain that the decrease of $(H)_{LM}/l$, that is the increase of the absorption rate, is caused by the generation of turbulence in the liquid film as was interpreted by Emmert et al. With the transition of the flow from perfect laminar to pseudo-laminar the true molecular diffusion is present no longer, but the turbulent diffusion reveals itself gradually. Under such conditions the actual diffusion coefficient is not D_L , but \bar{D}_L , whose value can be calculated from the curve P or H in Fig. 1, assuming that the theory of Nusselt can be applied in spite of the presence of turbulence. The value of \bar{D}_L thus calculated is divided into D_L and D_L' as the equation

$$\bar{D}_L = D_L + D_L', \tag{6}$$

where D_L' is a coefficient of eddy diffusion dependent on the state of flow only.

As the factors which had great influence upon a pseudo-laminar flow were L' , μ_L , ρ_L , σ_L and g , D_L' appears to be affected also by the same factors. The dimensional analysis of these factors gives three dimensionless groups $(g\rho_L^3/\sigma_L^3)^{1/4}D_L'$, μ_L/ρ_LD_L' , and L'/μ_L . L'/μ_L is equivalent to Re_L , and it is more reasonable to substitute $(Re_L - Re_{LC1})$ for L'/μ_L . Fig. 4 shows a plot of $(g\rho_L^3/\sigma_L^3)^{1/4}D_L'$ against $(Re_L - Re_{LC1})$. Where $(Re_L - Re_{LC1})$ is less than 150, that is, Re_L less than 200, the plotted points

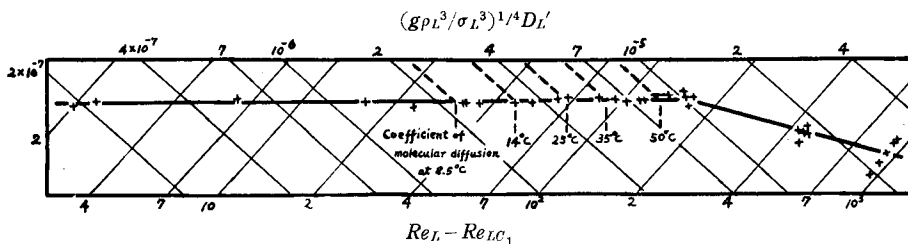


Fig. 4. Coefficient of eddy diffusion in perfect laminar and pseudo-laminar liquid film.

at all temperatures lie on one straight line represented by the following equation.

$$(g\rho_L^3/\sigma_L^3)^{1/4}D_L' = 1.015 \times 10^{-7} (Re_L - Re_{LC1})^{0.91} \quad (7)$$

This part corresponds to the part B in the previous figure. Where Re_L is greater than 200, the plotted points begin to scatter with increase of Re_L , because the degree of deviation from the theory of Nusselt becomes greater. This part corresponds to the part C. In Fig. 4 the value of ordinate equivalent to D_L at each temperatures is marked. Comparing this with the observed values it is known that D_L' is 1.2 to 3.5 times and 3 to 11 times as large as D_L when Re_L is about 200 and 1,000 respectively.

In the part D also, \bar{D}_L or D_L' can be calculated, and its calculation must be done on the basis of the velocity profile in a fully turbulent liquid film. If the exponential law concerning the velocity profile of the flow between parallel planes by Kármán and Prandtl is applied here, η in Eq. (2) takes 1.14. From the investigation of liquid hold-up by the authors,

$$B = 0.0140(\mu_L/\rho_L)^{0.68} Re_L^{0.578} \quad (8)$$

Substituting these values for η and B and \bar{D}_L for D_L in Eq. (1) gives

$$\zeta_M = 145 \bar{D}_L l (\mu_L/\rho_L)^{1.68} Re_L^{-1.58} \quad (9)$$

Using Eq. (9) and the curve H in Fig. 1, \bar{D}_L and D_L' are computed. The factors

that affect D_L' are L' , μ_L and ρ_L , and two dimensionless groups, $\mu_L/\rho_L D_L'$ and Re_L , are obtained. σ_L has no influence upon D_L' , as it had not upon B . In Fig. 5 $\rho_L D_L'/\mu_L$ is plotted against Re_L . The plotted points do not lie on one line, which means that $\mu_L/\rho_L D_L'$ is not a function of Re_L only. But it is impossible to choose any other factors than those above-mentioned, and the value of D_L' that is 6 to 12 times as large as D_L at 2,000 of Re_L and 50 to 65 times as large as D_L at 10,000 of Re_L seems to be too little as a usual value in a fully turbulent flow.

The authors would like to mention the following phenomenon as a reason for the abnormality of D_L' . A laminar sub-layer in which the true molecular diffusion occurs grows in a liquid film and the calculated D_L is a mean value taken over both the sub-layer and the turbulent core.

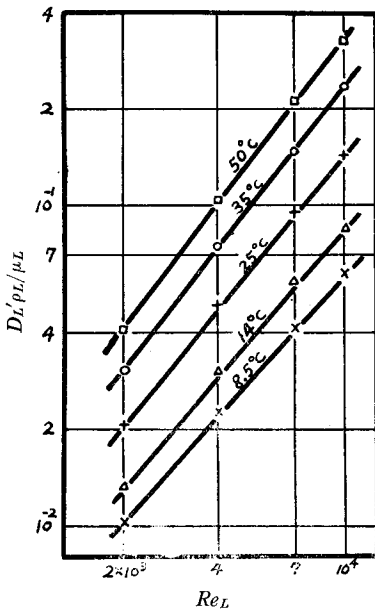


Fig. 5. Coefficient of eddy diffusion in turbulent liquid film.

This sub-layer does not exist on the interface of liquid and gas as is usual with ordinary cases, but it exists on the wall of tower. If the shearing stress or the friction factor on the wall is represented by the Blasius equation and the velocity profile by the exponential law,

$$\delta/B = 257 Re_L^{-7/8} \tag{10}$$

Eq. (10) evaluates the values of δ/B at 0.33 and 0.08 when Re_L are 2,000 & 10,000 respectively, and the presence of the sub-layer cannot be neglected under the conditions of this experiment. If so, it is preferable to establish an empirical equation for $(H)_{LM}$, and the following is recommended.

$$(H)_{LM}/l = 14 Re_L^{0.3} (\mu_L^2/g\rho_L^2l^3)^{0.25} (\mu_L/\rho_L D_L)^{0.556} \tag{11}$$

According to Eq. (11) $(H)_{LM}$ is inversely proportional to $D_L^{0.556}$ and proportional to $l^{0.25}$. The exponent of D_L , 0.556, shows that there is a considerable effect of the laminar sub-layer. The exponent of l is gained indirectly by determining the exponents of μ_L , ρ_L and D_L . It seems that with increase of l the quantity of absorbed gas reaching the sub-layer increases and consequently $(H)_{LM}$ increases.

(2) In the case of countercurrent flow of gas

The influence of the countercurrent flow of gas upon absorption rate can be solved theoretically for a perfect laminar liquid film. Assuming that the perfect laminar flow remains under the countercurrent flow of gas,

$$u_{LM} = g\rho_L B^2/3\mu_L - \tau_s B/2\mu_L \tag{12}$$

$$u_{LS} = g\rho_L B^2/2\mu_L - \tau_s B/\mu_L \tag{13}$$

From these two equations, neglecting the infinitesimal of higher order,

$$\eta = \frac{3}{2} \left(1 - \frac{\tau_s}{2\rho_L B_0 g} \right) \tag{14}$$

As η is 3/2 when the gas is stagnant, it is found that η decreases with the gas flow. From the research of liquid hold-up, neglecting the infinitesimal of higher order,

$$\frac{B}{B_0} = 1 + \frac{\tau_s}{2\rho_L B_0 g} \tag{15}$$

From Eqs. (1), (14) and (15)

$$\begin{aligned} \zeta_M &\simeq \frac{8}{3} \frac{1}{Re_L} \frac{\rho_L D_L}{\mu_L} \frac{l}{B_0} \left(1 - \frac{\tau_s}{\rho_L B_0 g} \right) \\ &= \zeta_{M_0} \left(1 - \frac{\tau_s}{\rho_L B_0 g} \right), \end{aligned} \tag{16}$$

where ζ_{M_0} is the value of ζ_M when the gas is stagnant, represented by Eq. (5). ζ_M decreases with the gas flow at the constant values of Re_L and $\mu_L/\rho_L D_L$ as shown in

Eq. (16), and the theoretical value of $(H)_{LM}/l$, therefore, becomes greater than in the case of the stagnant gas. This theory can be applied as well in a pseudo-laminar region, if \bar{D}_L is used for D_L , and the same may be also recognized in a fully turbulent region. Fig. 6 shows the plot of $(H)_{LM}/(H)_{LM0}$ against Re_G in some

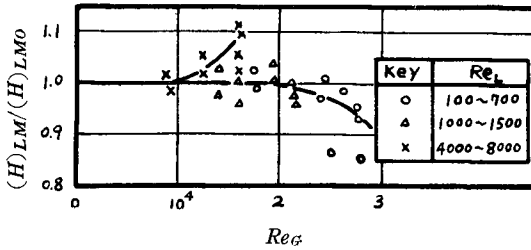


Fig. 6. Effect of gas flow.

experiments, where $(H)_{LM0}$ is the value of $(H)_{LM}$ when the gas is stagnant. The plotted points have different aspects according to the range of Re_L . In the range of low value of Re_L , $(H)_{LM}$ decreases above 18,000 of Re_G , contrary to the above-mentioned theory. But if the liquid film is disturbed and \bar{D}_L be-

comes greater than in the case of the stagnant gas due to the gas flow, ζ_M increases naturally in spite of the variations of B and η , and so $(H)_{LM}$ must decrease in accordance with the results obtained. In the range of 1,000 to 1,500 of Re_L , $(H)_{LM}$ varies no longer, because the liquid film is already disturbed and the effect of the gas flow on B and η cancels the effect on \bar{D}_L . In the fully turbulent flow \bar{D}_L has considerably large value and is not affected by the gas flow. The increase of δ with the increase of B results in the increase of mass transfer resistance. Then, $(H)_{LM}$ may increase as shown in the figure.

Nevertheless it is difficult to maintain a constant temperature of the system under the condition of the gas flow. The data obtained are rather incorrect and the quantitative interpretations cannot be made about them.

Part II Heat Transfer

1. Introduction

Heat transfer rate in a falling liquid film has been seldom studied, because there have been no appropriate methods of measuring the heat transfer rate in a thin film correctly.

The experiment of heating of a falling liquid film by hot gas in an air-water system is treated in this paper.

2. Basic Principle

The rate of heat transfer in a laminar liquid film by conduction only is expressed analogously to the theoretical rate of mass transfer, using λ_L for D_L . The observed value of heat transfer rate must be determined by measuring the temperature of the

surface of liquid film precisely in order to compare it with the theoretical value. But the measurement is too difficult. There is another method to calculate the rate only in the falling liquid film with the rate over the liquid phase and the gas phase on the basis of the double film concept. But the heat transferred through a liquid film is not equal to the heat through a gas film on account of vaporization of liquid or condensation of vapor contained in the gas, and the value of the over-all resistance to the transfer cannot be obtained from the value of the apparent temperature difference.

It is better to use the enthalpy of gas as a driving force of transfer as proposed by Merkel⁸⁾ and Hirsch⁹⁾. Assuming that the Lewis equation is applied and the humidity difference is used as a driving force of transfer of vapor in the gas phase,

$$\frac{\pi d L k_G'}{G_0} = \int_{i_{Gm_1}}^{i_{Gm_2}} \frac{di_{Gm}}{i_{Gm} - i_{GS}} \tag{17}$$

As i_{GS} is the enthalpy of gas saturated with the vapor at the temperature of the interface of gas and liquid, it is in a definite relation to t_{LS} as follows.

$$i_{GS} = f(t_{LS}) \tag{18}$$

At any cross section of the tower i_{Gm} is related to t_{Lm} with the following equations.

$$G_0(i_{Gm} - i_{Gm_1}) = L(t_{Gm_2} - t_{Lm}) \tag{19}_1$$

$$G_0(i_{Gm_2} - i_{Gm}) = L(t_{Lm} - t_{Lm_1}) \tag{19}_2$$

The relation between i_{LS} and i_{Lm} is given as follows.

$$i_{Gm} - i_{GS} = \frac{h_L}{k_G'} (t_{LS} - t_{Lm}) \tag{20}$$

Eliminating t_{LS} and t_{Lm_1} or t_{Lm_2} from Eqs. (18), (20) and (19)₁ or (19)₂, i_{GS} is gained as a function of i_{Gm} and the parameter h_L/k_G' . Substituting this i_{GS} in the right hand of Eq. (17) and integrating the equation, the resultant is a function of h_L/k_G' , the solution of which gives the value of h_L/k_G' . Practically the algebraic form of Eq. (18) is unknown and the graphical method must be used as shown in Fig. 7. In the figure the straight line O expresses Eq. (19)₁ or (19)₂, the curve E Eq. (18) and the tie line T Eq. (20). Lines T are drawn through some arbitrary points on the line O with the hypothetical value of h_L/k_G' and the points (t_{LS}, i_{GS}) are determined on the curve E. With the

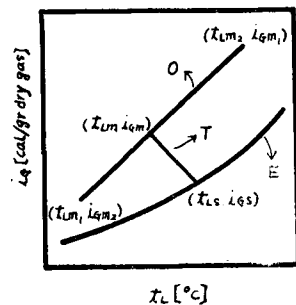


Fig. 7. Temperature-enthalpy diagram.

obtained values of i_{GS} and i_{Gm} the integration of the right hand of Eq. (17) is graphically accomplished. h_L/k_G' is determined when the integral value becomes equal to the value of the left hand of Eq. (17) by varying the slope of the lines T. It should be noted that this method is not applicable if h_L/k_G' is varied along the tower.

3. Apparatus and Procedure

Fig. 8 is a schematic diagram of the apparatus used. The test section of the tower was made of a glass tube, 4.85 cm inner diameter and 282 cm long, and was enclosed with electrical heating wires to prevent heat losses.

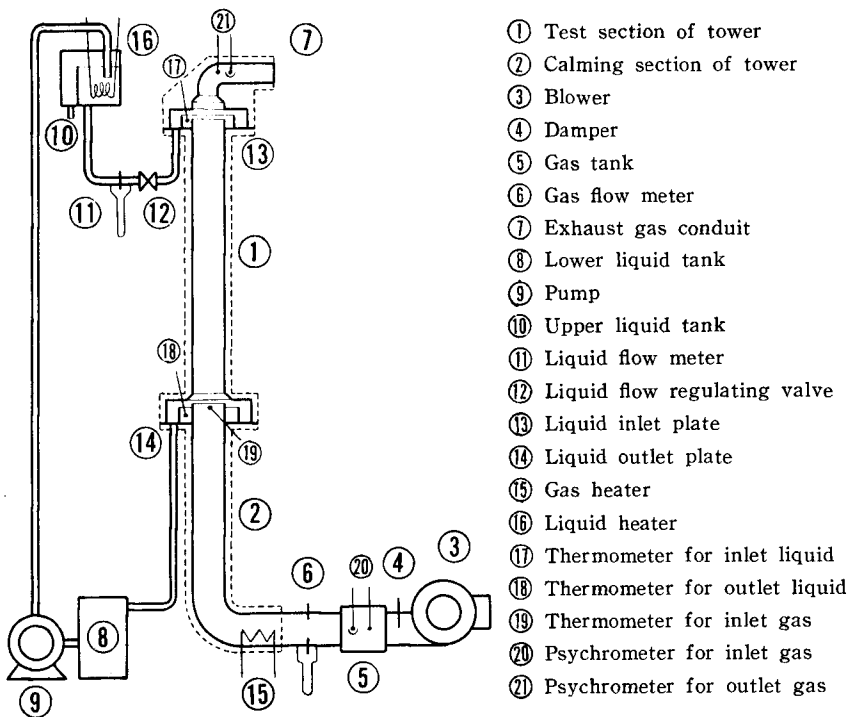


Fig. 8. Apparatus.

Air entered the bottom of the tower after heated to 88 to 100°C and exhausted after cooled and humidified. Water entered the top of the tower at 20 to 30°C and fell down on the wall, being heated by the countercurrent flow of air. The temperature of water was regulated so that the saturated vapor pressure at the temperature was always larger than the vapor pressure of air present in the tower. At any cross section of the tower, therefore, the vapor moved from water to air.

The feed rate of water was limited within 260 to 940 of Re_L . The temperature of air decreased 43 to 59°C and the temperature of water increased 1 to 7°C.

4. Results and Discussions

The heat transfer rate was calculated by means of the method described in Introduction, assuming that h_L/k_G' was not varied along the tower, where k_G' was gained from the empirical equation given in the study of mass and heat transfer in a gas stream of wetted wall tower by the authors. As the increase of the temperature of water was only 1 to 7°C, the error in the measurement of the temperature had a great influence upon the calculation and the obtained values of h_L became very rough. In order to compare the results with those of the gas absorption, the obtained h_L was transformed into $(H)_{LH}$ with the following equation.

$$\frac{(H)_{LH}}{l} = \frac{L's_L}{h_L l} = \frac{Re_L}{4} \frac{s_L \mu_L}{h_L l} \tag{21}$$

The effect of the gas flow could be neglected as the flow of liquid was pseudo-laminar and Re_G was 23,500 at highest.

As mentioned above, the theoretical heat transfer rate is a function of ζ_H only which is defined as

$$\zeta_H = \frac{\lambda_L l}{B^2 \mu_L s_L \rho_L} = 2.92 \frac{\lambda_L l}{Re_L^{4/3} s_L \rho_L} g^{1/3} \left(\frac{\rho_L}{\mu_L} \right)^{5/3}, \tag{22}$$

and the relation between $(H)_{LH}/l$ and ζ_H is the same as between $(H)_{LM}/l$ and ζ_M .

In Fig. 9 the observed values of $(H)_{LH}/l$ are plotted against Re_L . The plotted points scatter considerably, though the good data are chosen. The straight lines I and II in the figure give the theoretical values at the highest and lowest temperature of water in the experiment respectively, and as ζ_H is very small under the condition in the experiment they are expressed by the equation,

$$\frac{(H)_{LH}}{l} = \frac{1}{5.121 \zeta_H} = \frac{s_L \rho_L Re_L^{4/3}}{15 l \lambda_L g^{1/3}} \left(\frac{\mu_L}{\rho_L} \right)^{5/3} \tag{23}$$

In this equation $(H)_{LH}$ is independent of l , and, therefore, the observed values of $(H)_{LH}$ which are obtained on the assumption of its independence of l can be compared with the theoretical ones. In spite of the scattering of the points, the tendency of the data is generally in accordance with the theory which is on the basis of the molecular heat conduction in a liquid film. It means that the eddy diffusional process

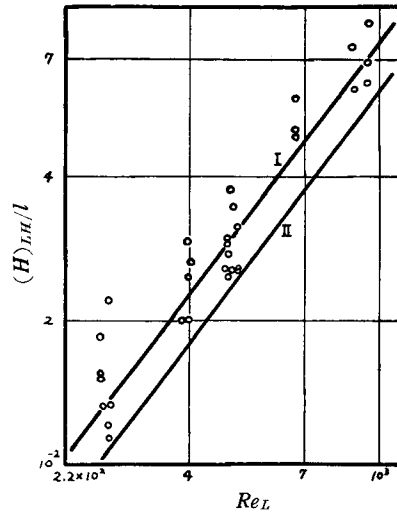


Fig. 9. Heat transfer rate in falling liquid film.

of heat does not exist. In the case of carbon dioxide gas absorption D_L' is 4×10 to 12×10 cm²/sec in a pseudo-laminar flow region as described above. If the analogy of eddy diffusion between heat and mass transfer can be postulated, $\lambda_L'/s_L\rho_L$ should be equal to D_L' and it is 1/40 to 1/10 times as large as $\lambda_L/s_L\rho_L$. Hence, in a liquid film the eddy diffusional process of heat transfer may be neglected, compared with the molecular diffusional process. This is the reason the observed values coincide with the theory.

Notations

- B : Thickness of falling liquid film (cm)
 B_0 : B without gas flow („)
 D_L : Coefficient of molecular diffusion (cm²/sec)
 \bar{D}_L : Coefficient of actual diffusion (cm²/sec)
 D_L' : Coefficient of eddy diffusion (cm²/sec)
 d : Diameter of tower (cm)
 G_0 : Mass flow rate of dry gas (gr/sec)
 g : Acceleration of gravity (cm/sec²)
 $(H)_{LH}$: Height per transfer unit of heat in falling liquid film (cm)
 $(H)_{LM}$: Height per transfer unit of mass in falling liquid film (cm)
 $(H)_{LMO}$: $(H)_{LM}$ without gas flow („)
 h_L : Film coefficient of heat transfer in falling liquid film (cal/cm² sec °C)
 i_{Gm} : Mean enthalpy of wet gas at any cross section of tower per unit mass of dry gas (cal/gr dry gas)
 i_{Gm_1} : i_{Gm} at inlet of tower („)
 i_{Gm_2} : i_{Gm} at outlet of tower („)
 i_{GS} : Enthalpy of wet gas on interface of gas and liquid per unit mass of dry gas (cal/gr dry gas)
 k_G' : Film coefficient of mass transfer in gas phase (gr/cm² sec)
 L : Mass flow rate of liquid (gr/sec)
 L' : Mass flow rate of liquid per unit wetted periphery (gr/cm sec)
 l : Length of wetted part of tower (cm)
 Re_G : Reynolds number of gas flow (—)
 Re_L : Reynolds number of liquid film (—)
 Re_{LC_1} : Re_L at critical point between perfect laminar and pseudo-laminar flow („)
 s_L : Specific heat of liquid (cal/gr °C)
 t_{Lm} : Mean temperature of liquid at any cross section of falling liquid film (°C)
 t_{Lm_1} : t_{Lm} at inlet of tower („)
 t_{Lm_2} : t_{Lm} at outlet of tower („)

t_{LS}	: Temperature of water at interface between gas and liquid ($^{\circ}\text{C}$)
u_{Lm}	: Mean velocity of falling liquid film (cm/sec)
u_{LS}	: Surface velocity of falling liquid film (cm/sec)
δ	: Thickness of laminar sub-layer (cm)
ζ_H	: Parameter in heat transfer (—)
ζ_M	: Parameter in mass transfer (—)
ζ_{M0}	: ζ_M without gas flow (—)
η	: Ratio of u_{LS} to u_{Lm} (—)
λ_L	: Molecular heat conductivity of liquid (cal/cm sec $^{\circ}\text{C}$)
λ_L'	: Eddy heat conductivity of liquid (cal/cm sec $^{\circ}\text{C}$)
μ_L	: Viscosity of liquid (poise)
π	: Ratio of circumference of circle to its diameter (—)
ρ_L	: Density of liquid (gr/cm ³)
σ_L	: Surface tension of liquid (dyne/cm)
τ_s	: Shearing stress acting on falling liquid film (dyne/cm ²)

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