EFFECT OF FLUID PROPERTIES ON MASS TRANSFER IN THE GAS PHASE

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Water was vaporized into air, helium, and Freon-12 in turbulent gas flow over a wet-bulb thermometer and through a 1-ft.-diam. tower packed with 1-in. Raschig rings. The wet- and dry-bulb measurements indicate that the gas-film mass transfer coefficient varies with the Schmidt number to the —½ power for flow perpendicular to single cylinders.

Heights of a transfer unit, H.T.U., were measured in the packed tower for flow of the gases countercurrent to water over a range of gas and liquid rates. When compared at equal-gas-flow Reynolds numbers at constant liquid rate, H.T.U. varied as the 0.9 power of the Schmidt group. When compared at equal values of ρu^2 (ρ = gas density, u = velocity), H.T.U. varied as the 0.47 power of the Schmidt group. With reference to the psychrometric study, the latter method of comparison of H.T.U.'s seems preferable and indicates that further study of criteria for dimensional similitude in packed columns may be needed.

According to the two-film theory of Whitman (*50*), the resistance to mass transfer between a gas and a liquid stream can be divided into two additive resistances which result from hypothetical films of stagnant gas and liquid on either side of the interface. The liquid-film resistance in a packed column has been evaluated by Sherwood and Holloway (*41*) for the commonly used packings, but as yet a satisfactory correlation of gas-film absorption coefficients has not been made.

This investigation proposed to clarify the role of the gas-film resistance by determining the effect of the fluid properties on the rate of mass transfer in a packed tower. To do this, a twofold plan was followed. The most direct method was the actual measurement of mass transfer coefficients in a tower packed with 1-in. Raschig rings. To eliminate any influence from the liquid properties, it was decided to vaporize a single pure liquid into inert gases. The gases chosen were air, helium, and Freon-12. These gases, with water as the liquid, gave a thirtyfold variation in density, a twofold variation in viscosity, an eightfold variation in diffusivity, and a four-and-one-half-fold variation in Schmidt number.

As the Schmidt-number effect was of particular interest, the second method of approach utilized wet- and dry-bulb-thermometer measurements to determine the psychrometric ratio. As the Prandtl number of the gases is nearly constant, this method will give the variation of the mass transfer coefficient with the Schmidt number in evaporation from single cylinders. Although it is not necessary that the same Schmidt-number effect hold for single cylinders and packed towers, the effect should be of similar magnitude and should confirm the results obtained by the more direct method.

THEORY

Approximate analytical solutions for the rate of mass transfer from an interface to a fluid moving in forced convection in a straight circular pipe have been developed by various authors (*5, 10, 34, 40*) through the use of the analogy between heat, mass, and momentum transfer. These solutions have indicated that the mass transfer coefficient, k_G for this case, is a function of the Reynolds number, $(du\rho/\mu)$, and the Schmidt number, $(\mu/\rho D)$. The Reynolds number ordinarily enters as a function replacing the friction factor. These solutions can be expressed generally by the equation

$$k_G M_m p_f/G = \phi'(du\rho/\mu), (\mu/\rho D) \qquad (1a)$$

where

ϕ' = some mathematical function
k_G = mass transfer coefficient, lb. moles/(hr.)(sq.ft.)
d = diameter of the tube, ft.
D = diffusion coefficient, sq. ft./sec.
u = velocity, ft./sec.
ρ = density, lb. mass/cu.ft.
μ = viscosity, lb. mass/(ft.)(sec.)
G = mass velocity of the gas, lb. mass/(hr.)(sq.ft.)
M_m = mean molecular weight, lb. mass/mole
p_f = film pressure factor (*51*); accounts for the presence and movement of the inert fluid

This equation can be expressed alternately in terms of the transfer unit defined by Chilton and Colburn (*8*):

$$a\text{H.T.U.}_G = \psi'(du\rho/\mu), (\mu/\rho D) \qquad (1b)$$

where

H.T.U.$_G$ = height of a transfer unit = $G/k_G a p_f M_m$, ft.
a = packing area/cu.ft. of packing, sq.ft./cu.ft.

These equations do not include the effect of the relative roughness on the mass transfer rate and consequently are limited to the particular pipe geometry for which the functions Φ' and Ψ' are experimentally evaluated.

Mass Transfer in Packed Columns. Equations (1*a*) and (1*b*) have been frequently used to satisfy a need for some theoretical framework for the correlation of packed-column data; however, this extension of the functional relationship developed for simple pipe flow has

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never been subjected to adequate experimental investigation to justify its general use. In an attempt to correlate the results of this work it was found that Equations (1*a*) and (1*b*) were not satisfactory, and an alternate method of correlation was therefore developed based on the following qualitative analysis.

In the study of mass transfer rates from single cylinders placed perpendicular to the flow stream (*29*), it has been found that the bulk of the mass transfer taking place occurs in the vicinity of the stagnation point and in the region of the turbulent wake. These same two sections of the cylinder are of primary concern in the loss of momentum of the gas stream as manifested by the pressure drop. Consequently, it might be expected that the two phenomena, transfer of momentum and transfer of mass, are to a large extent functions of the same properties of the flow field, particularly for gases in which the diffusivities for mass and momentum are of similar magnitude. Pressure-drop measurements made in flow past single cylinders (*15*) show that the drag coefficient becomes relatively constant at high values of the Reynolds number. Therefore, as a first approximation, the drag coefficient is independent of the Reynolds number and the pressure drop is dependent only upon the inertia of the gas stream, ρu_m^2. Since it is postulated above that the rate of mass transfer will depend on the same flow properties as does the pressure drop, it may follow that the mass transfer coefficient will also be a function of the inertia force and not the Reynolds number at high flow rates. With this modification, Equations (1*a*) and (1*b*) may be rewritten for application to single cylinders for conditions of high turbulence in the fluid:

$$k_G M_m p_f / G = \varphi(\rho u_m^2),\ (\mu/\rho D) \quad (2a)$$

$$a\text{H.T.U.}_G = \psi\ (\rho u_m^2),\ (\mu/\rho D) \quad (2b)$$

where u_m = mean velocity of the flowing fluid in feet per second. Pressure-drop measurements made in packed absorption towers (*37*) have also shown very little dependence on the Reynolds number over the usual operating range. A tower packed with Raschig rings being considered in a broad sense as a group of cylinders, the extension of Equations (2*a*) and (2*b*) to a packed tower would appear to follow.

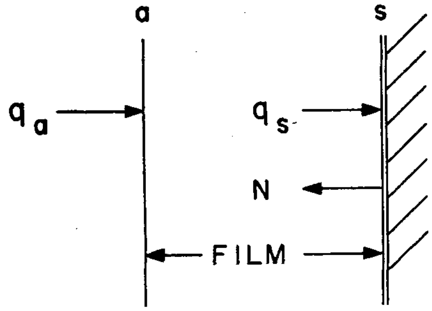

FIG. 1. SCHEMATIC REPRESENTATION OF HEAT AND MASS TRANSFER TAKING PLACE AT A WET-BULB THERMOMETER.

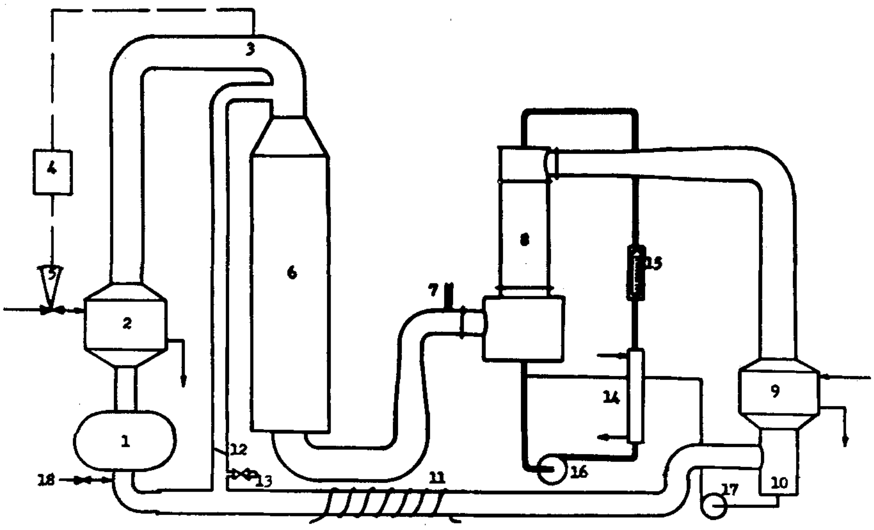

FIG. 2. GENERAL EQUIPMENT LAYOUT.

1. Blower
2. Heater
3. Thermocouple
4. Temperature recorder-controller
5. Motor valve
6. Flow-measuring chamber
7. Wet- dry-bulb assembly
8. Absorption tower
9. Cooling coil
10. Condensate receiver
11. Superheating coil
12. Butterfly valve—by-pass control
13. Vent valve
14. Liquid heat exchanger
15. Liquid flow meter
16. Liquid pump
17. Condensate return pump
18. Steam line for purging

Psychrometry. If the vapor content of a gas stream is known, the psychrometer is a useful instrument for measuring the relative rates of heat and mass transfer from single cylinders. It has a distinct advantage over other methods for determining the dependence of k_G on Schmidt number in that no knowledge of the velocity-term dependence is required. If an exact analogy can be drawn between the two processes of heat and mass transfer, the velocity term should enter into both rates in the same manner, and the ratio of the mass transfer to the heat transfer coefficient should be a unique function of the Schmidt and Prandtl numbers.

Colburn (*10*), and Chilton and Colburn (*7*) have developed equations for the correlation of heat and mass transfer data in terms of the *j* number, a quantity which is analogous to the friction factor in momentum transfer. According to this concept,

$$j_h = [h/C_pG]\ (C_p\ \mu/k)^{2/3} \quad (3)$$

$$j_d = [k_G M_m\, p_f/G]\ (\mu/\rho\ D)^{2/3} \quad (4)$$

where

h = heat transfer coefficient, B.t.u./(hr.) (sq.ft.) (°F.)

C_p = heat capacity of the gas, B.t.u./(lb. mass) (°F.)

k = thermal conductivity, B.t.u./(hr.) (sq.ft.)) °F./ft.)

If j_h is assumed to be equal to j_d, the psychrometric ratio is obtained:

$$(k_G M_m P_f/h)C_p = \left[\left(\frac{C_p\mu}{k}\right)\Big/\left(\frac{\mu}{\rho D}\right)\right]^{2/3} \quad (5)$$

In the above *j*-number analogy the value of k_G is determined directly from the wet-bulb data, as it is multiplied by the term p_f, which corrects for variations in the partial pressure of the inert gas. The value of *h*, on the other hand, is the true heat transfer coefficient which would be obtained if there were no evaporation from the surface. To correct the wet-bulb data it is necessary to include a factor which will allow for the heat being carried away from the thermometer by the mass movement of the vapor molecules.

Figure 1 shows a model in which the transfer of heat and mass occurs only in the normal direction. With the surface temperature as a reference, it is seen that the heat transferred to the surface, q_s, is equal to the heat entering the film, q_a, minus the heat being carried out by the vapor flow, $NC_m(t_a - t_s)$, or

$$q_s = q_a - NC_m\ (t_a - t_s) \quad (6)$$

where

N = number of moles diffusing per unit time per unit area, moles/(hr.) (sq.ft.)

C_m = molar heat capacity of the vapor, B.t.u./(lb. mole) (°F.)

t_a, t_s = gas and surface temperatures respectively, °F.

If there were no flow of vapor, q_s would equal q_a:

$$q_s = q_a = h\,(t_a - t_s) \tag{7}$$

where h = heat transfer coefficient, assumed independent of the mass transfer movement.

The number of moles diffusing may be defined in terms of the mass transfer coefficient as

$$N = k_G\,(p_s - p_a) = k_G \Delta p \tag{8}$$

Substituting from Equations (8) and (7) into Equation (6) gives

$$\begin{aligned} q_s &= h(t_a - t_s) - C_m k_G\,\Delta p\,(t_a - t_s) \\ &= h(t_a - t_s)\,(1 - C_m k_G \Delta p/h) \\ &= h(t_a - t_s)\gamma \end{aligned} \tag{9}$$

$$\text{where } \gamma = 1 - C_m k_G \Delta p/h \tag{9a}$$

If a new pseudo heat transfer coefficient, h', is defined as the total effective heat transfer coefficient including the convective transport of heat, then

$$h' = h_{\gamma} \tag{10}$$

and the correct psychrometric ratio* is:

$$\beta = \frac{k_G M_m p_f \gamma}{h'} C_p = \text{function}$$

$$(\mu/\rho\, D),\ (C_p \mu/k) \tag{11}$$

Colburn and Drew(*10a*) have developed a procedure for correcting the heat transfer coefficient for the effect of mass transfer in the case of condensation of mixed vapors. Their method could also be applied to the wet bulb thermometer. Experimental verification of these possible methods would be desirable.

EXPERIMENTAL

Physical and Thermodynamic Properties of Materials†. *Viscosity.* The viscosities of air, water, vapor, and helium were taken from a review article by F. G. Keyes(*27*) in which the available literature data for several gases are collected and formulated by means of an empirical equation. The viscosity of Freon-12 was taken from a paper by Buddenburg and Wilke(*6*) on the measured viscosity of several gases. It was extended over the desired temperature range by the use of the equation of Hirschfelder, Bird, and Spotz(*21*).

Thermal conductivity. The thermal conductivity of air, water vapor, and helium was taken from the review article by Keyes(*27*) quoted above. The thermal conductivity of Freon-12 is that reported by Sherratt and Griffiths(*39*) over the range from 33.3° to 216.2° C.

Heat capacity, C_p. The heat capacity of air and water vapor was taken from the book by Keenan(*26*) on the thermodynamic properties of air. The heat capacity of Freon-12 was calculated from the work of Justi and Langer(*25*), who reported the heat capacity at constant volume. The heat capacity of helium is reported in Perry's handbook(*36*) as constant for all temperatures.

Diffusivity. The diffusivities used were those measured by C. Y. Lee(*28*). The measured values were corrected to the desired temperature by the equation of Hirschfelder, Bird, and Spotz(*21*).

Gas Specifications. The helium used, supplied by the U. S. Navy, was 99.9% pure, the major impurity being water vapor. The Freon-12 used was supplied by Kinetic Chemicals and was 99.95% pure; the major impurities were Freon-11 and Freon-13.

Equipment.

The general equipment arrangement is shown in Figure 2. Gas circulation was provided by means of a Roots-Connersville blower, and the flow rate was measured with a nozzle, made to the specifications of Bean, Buckingham, and Murphy(*3*), which was placed in a 20-in.-diam. section of ducting. The gas stream entering the absorption tower (Figure 3) was sampled for moisture content in the gas distributor; the stream leaving the tower was sampled in the exit ducting; and the inlet-gas temperature was measured at the top of one of the risers on the gas distributor.

The large section of the tower served as a liquid reservoir. Liquid circulation was provided by a centrifugal pump, the flow rate being measured by a calibrated rotameter. The liquid entered the top of the tower through a 44-point distributor, which discharged within 1/2 in. of the top of the packing. The temperature of the entering liquid was measured by a 4-couple thermopile placed in four of the distributing tubes. The liquid temperature at the bottom of the packing was measured by a single thermocouple placed in a small cup.

The wet- and dry-bulb thermome-

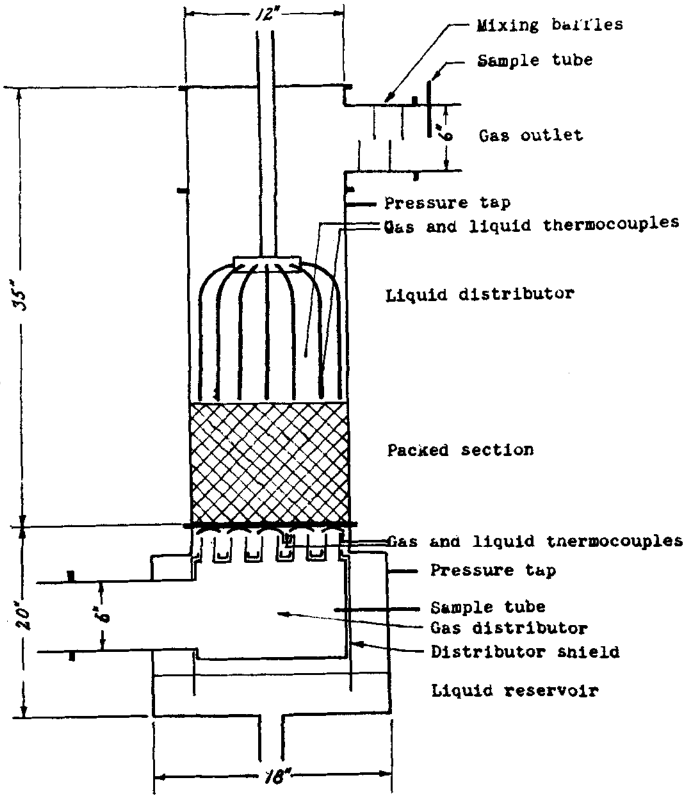

FIG. 3. ABSORPTION TOWER.

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*Heat transfer by radiation has been neglected in formulating the foregoing equations. As written, h is the usual forced convection coefficient for flow normal to cylinders (Ref. *36*, p. 473). To correct for radiation the factor $1 + \alpha$ may be introduced where α is the ratio of the radiation coefficient, h_r, to h, With this addition the psychrometric ratio, Equation (11), becomes

$$\beta = \frac{k_G M_m p_f C_p \gamma (1 + \alpha)}{h'}$$

$$= \text{function}\left(\frac{\mu}{\rho D}\right), \left(\frac{C_p \mu}{k}\right)$$

$$= \frac{M_m\ p_f C_p \gamma\ (t_a - t_s)\ (1 + \alpha)}{M_v\ \lambda_s\,(p_s - p_a)} \tag{11a}$$

where λ_s = latent heat of vaporization of the liquid, B.t.u./lb. mass. The right-hand term has been added to indicate the quantities involved in experimental measurement or use of the psychrometric ratio.

†Properties of pure gases were modified for the presence of water vapor by established methods.

ters for the psychrometic measurements (Figure 4) were placed in the ducting immediately before the tower and were preceded by an orifice which ensured a uniform high-velocity flow across the bulbs. Windows were provided in the duct wall so that positioning and wetting of the wick might be observed. A thermocouple was embedded in the duct wall to give the surface temperature for radiation corrections.

Procedure.

For all runs made with helium and Freon-12 it was necessary first to remove the air from the system. It was found that essentially complete removal could be effected by a single steam purge. The air concentration was checked by means of an Orsat analysis for oxygen, which was sensitive to ± 0.05%. When purging was complete, condensed steam was removed from the lines and the necessary adjustments were made to bring the equipment on stream. To prevent introduction of impurities into the gas stream, the equipment was operated with a slight positive pressure at the blower inlet.

After steady-state conditions were attained, gas samples were withdrawn simultaneously from the top and bottom of the tower by means of aspirator bottles of known volume. The moisture content was determined gravimetrically by drawing the samples through tubes filled with Drierite. During the sampling period the temperature of the inlet liquid stream was carefully controlled, to operate in general with the inlet- and exit-liquid temperatures less than 0.5° C. apart and with a variation of the inlet temperature of less than ± 0.05° C. during the run.

Immediately following sampling, water from an overhead supply was introduced to the wet-bulb wick and allowed to soak the cloth thoroughly. The water-supply rate was then reduced until a pendant drop could be observed on the tip of the wick. The wet- and dry-bulb temperatures were read. The water supply was stopped and the wet-bulb temperature checked for any further change. (Consecutive runs made at the same gas rate indicate that the moisture content of the gas stream remained essentially constant over long periods of time once steady-state conditions had been attained.)

All packed-bed data were taken with a 7 1/2-in. depth of packing with the exception of the runs made with air at heights of 2 and 13 1/2 in., which were used to determine the amount of transfer taking place outside the packed section. All runs were made at a liquid rate of 1,575 lb. mass/(hr.)(sq. ft.) except those made with air to check the effect of varying the liquid rate.

Details of equipment and procedure are available elsewhere(*30*).

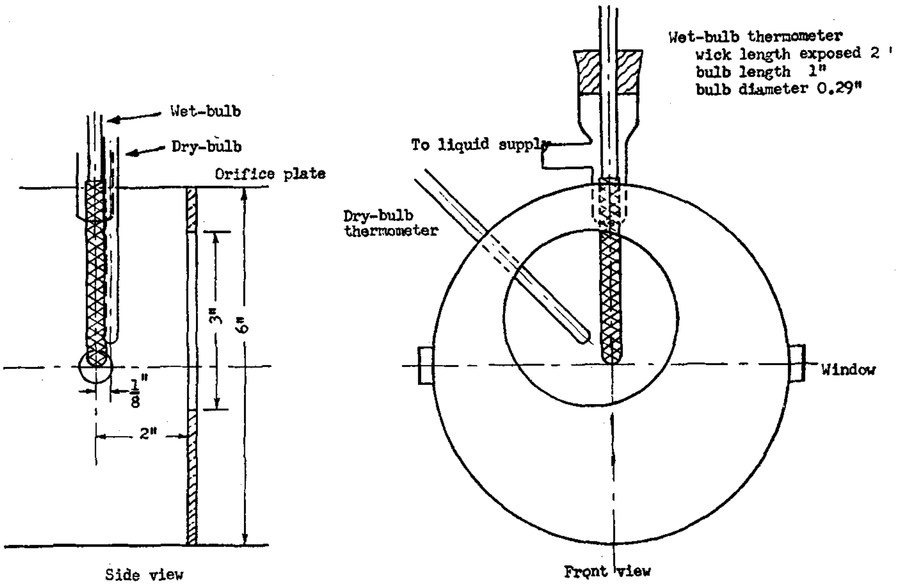

FIG. 4. WET- AND DRY-BULB-THERMOMETER ARRANGEMENT.

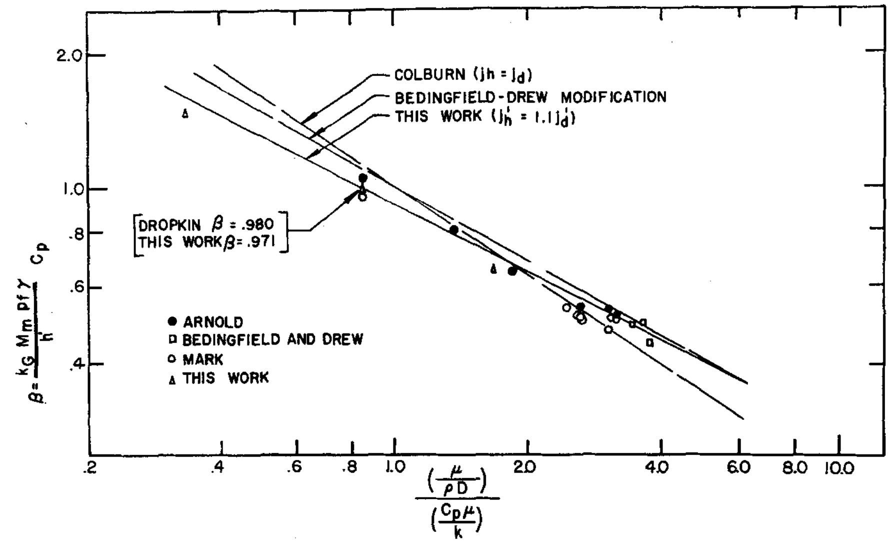

FIG. 5. THE PSYCHROMETRIC RATIO AS A FUNCTION OF *Sc*/*Pr*.

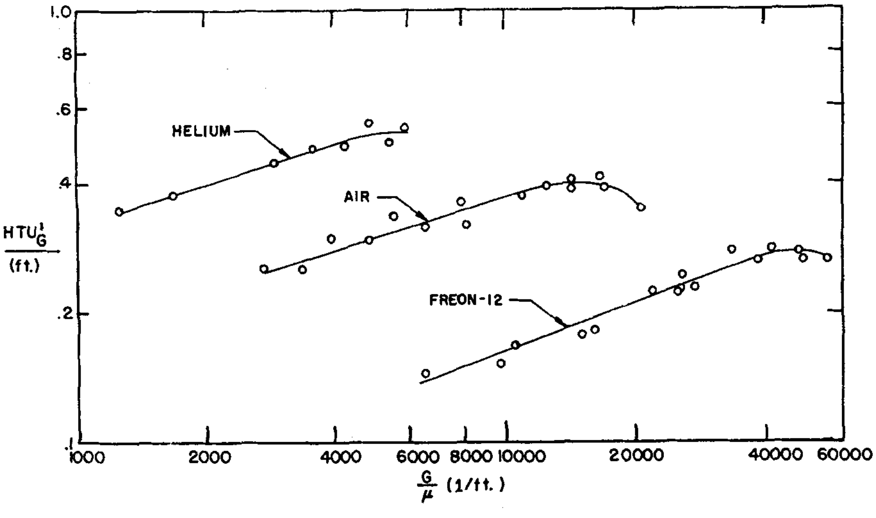

FIG. 6. H.T.U.$'_G$ AS A FUNCTION OF G/μ; $L = 1{,}575$.

RESULTS

Psychrometric.

The values of the psychrometric ratio obtained in this work for the three cases (Table 1) are shown plotted against the ratio *Sc/Pr* in Figure 6. This graph also includes the results of Arnold(*2*), Bedingfield and Drew(*4*), Mark(*31*), and Dropkin(*13*). These data can best be approximated by a straight line having a slope of —1/2, which satisfies a modified *j*-number analogy:

$$j'_h = 1.10\, j'_d$$

$$j'_h = \frac{h}{C_p G}\left(\frac{C_p \mu}{k}\right)^{\frac{1}{2}}$$

$$j'_d = \frac{k_G p_f M_m}{G}\left(\frac{\mu}{\rho D}\right)^{\frac{1}{2}} \qquad (12)$$

The equation for the psychrometric ratio* then becomes

$$\beta = \frac{k_G M_m p_f C_p \gamma}{h'} = 0.91\left(\frac{Pr}{Sc}\right)^{\frac{1}{2}} \qquad (13)$$

Since the value of the Prandtl group is nearly constant for the results shown here, the heat transfer coefficient may also be considered constant. The curve as plotted then represents the variation of the mass transfer coefficient with Schmidt number for flow of gas perpendicular to single cylinders; i.e., k_G is proportional to $Sc^{-\frac{1}{2}}$.

Packed Column

The packed-tower results are presented here in terms of the H.T.U. instead of k_G. In order to calculate the H.T.U. from the data, the following equation was used: (Representative data are presented in Table 2.)

$$H_2 - H_1 = \text{H.T.U.}_G \left[\left(\frac{y_f}{1-y}\right)_{avg} \ln\left(\frac{y_1 - y_i}{y_2 - y_i}\right)\right] \qquad (14)$$

where y = mole fraction and $y_f = p_f/P$. The expression in brackets is the number of transfer units, and its value can be computed from the data on the vapor concentrations at the inlet and exit of the tower and the temperature of the liquid stream. The height of the packed section, $H_2 — H_1$, when divided by the number of transfer units, gives the apparent height of a transfer unit.

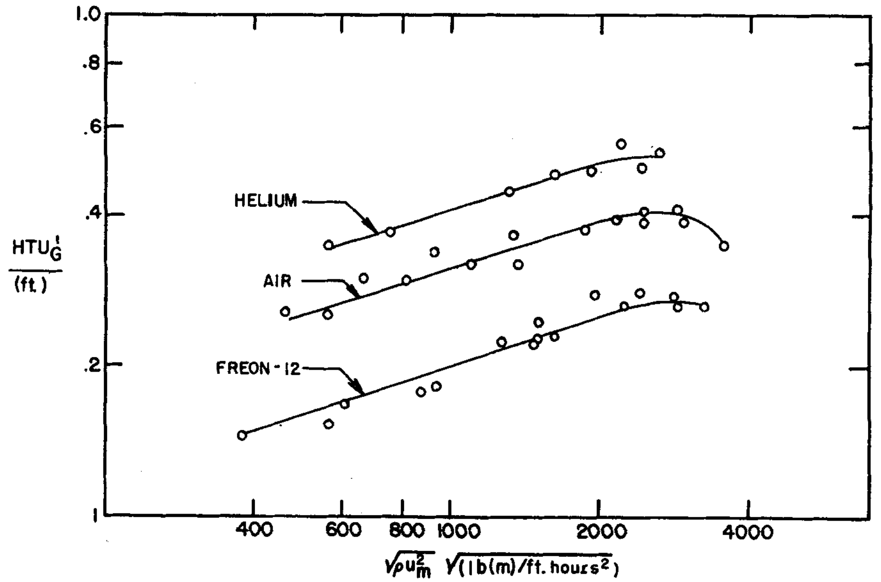

FIG. 7. H.T.U.$'_G$ AS A FUNCTION OF THE INERTIA GROUP; $L = 1{,}575$.

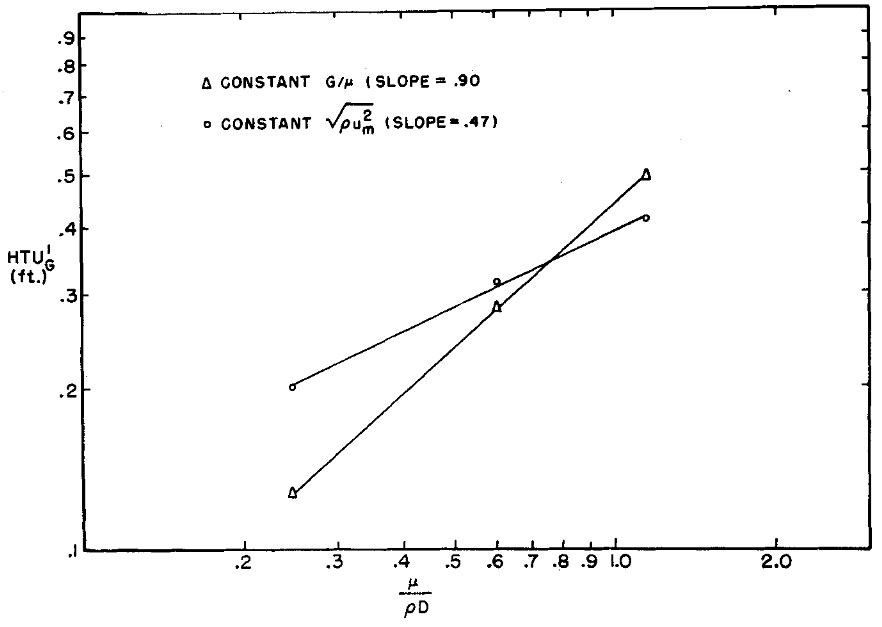

FIG. 8. H.T.U.$'_G$ AS A FUNCTION OF SCHMIDT NUMBER AT CONSTANT VALUES OF G/μ AND INERTIA GROUP; $L = 1{,}575$.

Figure 6 shows the apparent H.T.U. as a function of the group G/μ which is directly proportional to the Reynolds number for a given packing. Since only one packing size and shape was studied, the inclusion of a length dimension in the group was not warranted. The peaks of the curves correspond approximately to the flow rate at which the pressure-drop phenomenon known as "loading" occurs. This was determined by the simultaneous measurement of pressure drop across the packed section. The curves indicate that below the loading point the H.T.U. can be represented as varying with the mass velocity to the 0.33 power, with the power decreasing thereafter as the flooding point is approached. It will be noted that there is a slight difference in the slope for the three curves, but the differences are insignificant in view of the precision of the data.

The more obvious difference among the three curves is the lateral separation of the point of maximum H.T.U. The vertical separation of the curves might be expected and explained on the basis of Equation (*1b*) by the Schmidt-number differences, but the lateral separation would lead one to suspect that the Reynolds number may not be the proper correlating modulus in this case. If, instead, the group suggested by Equation (*2b*) is used, that is, ρu_m^2, the

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*To include radiation effects, see footnote equation (11a).

curves of Figure 7 are obtained. (The group ρu_m^2/length has the dimensional significance of inertia force per unit volume of fluid(*47*), and therefore the group ρu_m^2 will be designated the *inertia group.*) The square root of the inertia group has been used in plotting in order to retain the first-power dependence on the velocity that exists in Figure 6. Similar usage in the correlation of flooding-velocity(*42*) and pressure-drop(*37*) data in packed columns would seem to justify the procedure. This group has also been used in the correlation of plate efficiencies(*20*), where it has been given the name *F* factor. The curves of Figure 7 are identical in shape with those of Figure 6, but with the inertia group as the correlating modulus, the lateral separation has now disappeared and the peaks of all three

TABLE 1.—SUMMARY OF WET-BULB DATA FOR VARIOUS INVESTIGATIONS*

Investigator and system	Pr	Sc	Sc/Pr	$\frac{k_G M_m p_f \gamma}{h'} C_p$
Mark				
air-benzene	0.702	1.71	2.44	0.532
air-chlorobenzene	0.704	2.17	3.08	0.502
air-toluene	0.704	1.86	2.64	0.511
air-carbon tetrachloride	0.700	1.87	2.67	0.499
air-ethylene tetrachloride	0.704	2.15	3.05	0.474
air-ethyl acetate	0.709	1.83	2.58	0.514
air-water	0.704	0.602	0.855	0.955
Arnold				
air-water	0.704	0.602	0.855	1.049
air-methal	0.706	0.970	1.37	0.800
air-propanol	0.705	1.30	1.85	0.642
air-toluene	0.704	1.86	2.64	0.537
air-chlorobenzene	0.704	2.17	3.08	0.531
air-*m*-xylene	0.704	2.24	3.18	0.514
Bedingfield and Drew				
air-naphthalene	0.704	2.56	3.64	0.494
air-*p*-dichlorobenzene	0.704	2.22	3.15	0.509
air-camphor	0.704	2.65	3.76	0.448
air-*p*-dibromobenzene	0.704	2.41	3.42	0.487
Dropkin				
air-water	0.704	0.602	0.855	0.980
This work †				
air-water	0.702	0.602	0.858	0.971
helium-water	0.687	1.15	1.673	0.650
Freon 12-water	0.740	0.248	0.335	1.482

*The physical properties used in the Schmidt and Prandtl numbers were evaluated at 25°C. Diffusivities for the systems studied by Bedingfield and Drew were recalculated by the method of Arnold (*2*).

†Individual results may be found in reference (*30*).

TABLE 2.—SAMPLE EXPERIMENTAL AND CALCULATED RESULTS FOR VAPORIZATION IN PACKED TOWERS*

Exp.	L	G	T_{AB}	T_{WT}	T_{WB}	yi_T	yi_B	Barometer	y_T	y_B	N.T.U.	H.T.U.′	H.T.U.	H.T.U./$Sc^{0.47}$	G/μ	$\sqrt{\rho u_m^2}$
4	1,575	123.2	30.76	29.16	29.43	0.04022	0.04084	753.2	0.03849	0.02153	2.453	0.225	0.375	0.476	2,764	461.4
6	1,575	571.0	40.97	31.30	31.44	0.04540	0.04573	752.2	0.04168	0.02759	1,585	0.394	0.579	0.735	12,686	2,158
9	1,575	292.9	34.14	28.69	28.62	0.03907	0.03891	753.7	0.03690	0.02354	1.958	0.319	0.469	0.595	6,559	1,099
10	1,575	215.5	32.48	28.57	28.15	0.03882	0.03788	753.7	0.03701	0.02305	2,104	0.297	0.437	0.555	4,832	807.7
14	1,575	775.4	40.86	31.41	31.28	0.04567	0.04527	750.4	0.04255	0.02980	1,601	0.390	0.573	0.727	17,227	2,930
15	1,575	934.9	41.10	31.66	31.54	0.04619	0.04578	750.4	0.04377	0.03134	1.787	0.350	0.515	0.654	20,780	3,531
20	1,010	255.3	32.71	28.52	28.52	0.03887	0.03887	750.5	0.03734	0.02363	2.300	0.272	0.400	0.508	5,724	958.7
21	1,995	255.3	32.58	28.37	28.62	0.03853	0.03909	750.5	0.03718	0.02387	2.423	0.258	0.379	0.481	5,724	958.7
22	3,850	255.3	32.24	28.34	28.44	0.03846	0.03868	750.5	0.03739	0.02365	2.643	0.236	0.347	0.440	5,724	958.7
36	575	254.8	33.52	29.40	29.20	0.04081	0.04034	752.4	0.03770	0.02357	1.685	0.371	0.545	0.692	5,712	956.8
1*H*	1,575	137.5	38.78	29.07	28.94	0.03990	0.03959	752.2	0.03691	0.02749	1.398	0.447	0.657	0.615	2,923	1,312
3*H*	1,575	255.5	42.24	30.36	30.24	0.04277	0.04244	752.2	0.03924	0.03010	1.252	0.499	0.734	0.687	5,420	2,431
7*H*	1,575	79.03	34.77	28.70	28.02	0.03898	0.03747	753.7	0.03661	0.02472	1,683	0.371	0.545	0.510	1,683	754.1
8*H*	1,575	58.85	33.36	28.49	27.84	0.03851	0.03708	753.7	0.03627	0.02359	1.796	0.348	0.512	0.479	1,252	562.1
3*F*	1,575	1,514	41.59	34.16	34.18	0.05314	0.05302	753.4	0.04998	0.02634	2.250	0.277	0.407	0.784	48,200	2,820
4*F*	1,575	1,745	42.16	34.39	34.37	0.05375	0.05349	753.4	0.05128	0.02706	2.370	0.264	0.388	0.748	55,430	3,257
9F	1,575	203.8	35.76	32.31	32.67	0.04813	0.04898	752.8	0.04776	0.02148	4.309	0.145	0.213	0.410	6,538	377.9
11*F*	1,575	329.4	34.74	33.48	33.57	0.05111	0.05137	753.5	0.05045	0.02399	3.726	0.168	0.247	0.476	10,574	610.0
14*F*	1,575	874.2	43.00	35.09	35.90	0.05592	0.05844	749.6	0.05395	0.02836	2.726	0.229	0.337	0.649	27,725	1,635
15*F*	1,575	1,216	41.10	34.06	34.12	0.05292	0.05306	749.6	0.05049	0.02738	2.358	0.265	0.390	0.752	38,730	2,269

*Complete results may be found in reference (*30*).

L = liquid flow rate, lb. mass/(hr.)(sq.ft.)

G = gas flow rate, lb. mass/(hr.)(sq.ft.)

T_{AB} = gas temperature at the bottom of the packed section, °C.

T_{WT} = water temperature at the top of the packed section, °C.

T_{WB} = water temperature at the bottom of the packed section, °C.

y_{iT} = interfacial mole fraction of water at the top of the packed section

y_{iB} = interfacial mole fraction of water at the bottom of the packed section

y_T = bulk mole fraction at the top of the packed section

y_B = bulk mole fraction at the bottom of the packed section

N.T.U. = number of transfer units

H.T.U.′ = apparent height of a transfer unit, ft.

H.T.U. = height of a transfer unit corrected for end effects, ft.

Sc = Schmidt number $\left(\frac{\mu}{\rho D}\right)$

G/μ = value proportional to the Reynolds number, 1/ft.

$\sqrt{\rho u_m^2}$ = inertia group, $\sqrt{\text{lb.mass/(ft.)(hr.)}^2}$

curves appear at the same value of the inertia group within the limits of experimental error.

Cross plots of the H.T.U. vs. Schmidt number at a constant value of G/μ of 4,000 in Figure 6 and at a constant value of $\sqrt{\rho u_m^2}$ of 1,000 in Figure 7 are presented in Figure 8. The cross plots indicate the Schmidt-number function by which the H.T.U. must be divided to cause a vertical alignment of the three curves of Figures 6 and 7. The plot based on the Reynolds number would require this function to be $Sc^{0.9}$. Correlation by the inertia-group plot, on the other hand, requires an exponent of 0.47 on the Schmidt group to align the three curves. This exponent is preferred in view of the good agreement with that obtained in the psychrometric study and with some results obtained in the vaporization of liquids and organic solids in packed beds (*9, 43*). A simple power function of the Schmidt group appears to be satisfactory for correlation over the present range of system properties and experimental conditions.

To obtain the true H.T.U., it is necessary to correct for "end effects," that is, that amount of transfer taking place outside the packed section. In the system under study, the amount of transfer is dependent primarily on the area of liquid exposed to the gas, which in turn depends on the liquid distribution and the manner in which the liquid drips from the bottom of the packing. This should be reasonably independent of the gas being used, so that the same end-effect correction might apply to all three gases. If it can also be assumed that the effect of the gas properties on mass transfer is the same in the end section and in the packed section, then the end effect can be expressed in terms of an equivalent length of packing, which should be constant for all gases and independent of the packed height. This procedure has been used by previous investigators (*18, 45*). If the end effect is constant, the apparent H.T.U. values obtained at a given packed height need only be multiplied by a constant value to obtain the true H.T.U. values; therefore the preceding analysis of the Schmidt-number effect is not dependent on

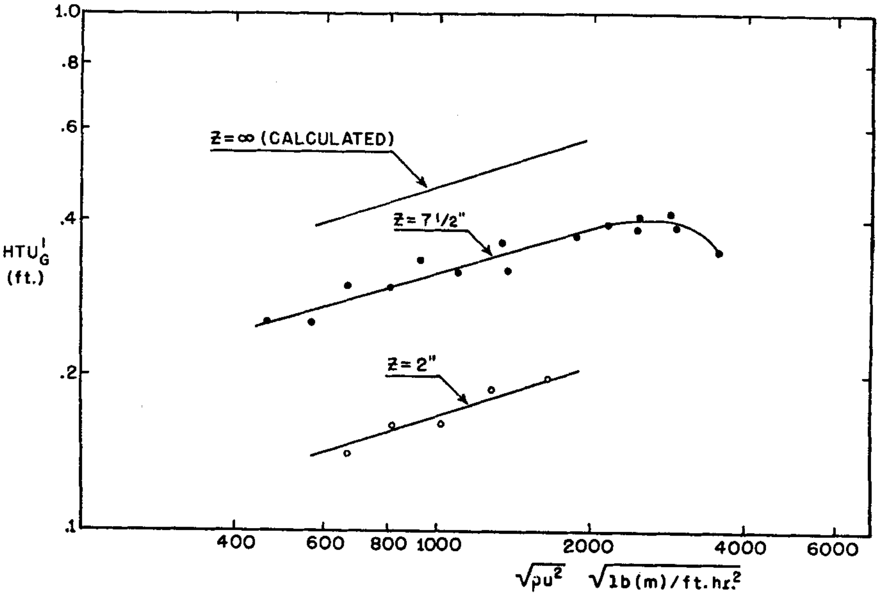

FIG. 9. THE APPARENT H.T.U. AS A FUNCTION OF PACKED HEIGHT FOR VAPORIZATION OF WATER INTO AIR; $L = 1{,}575$.

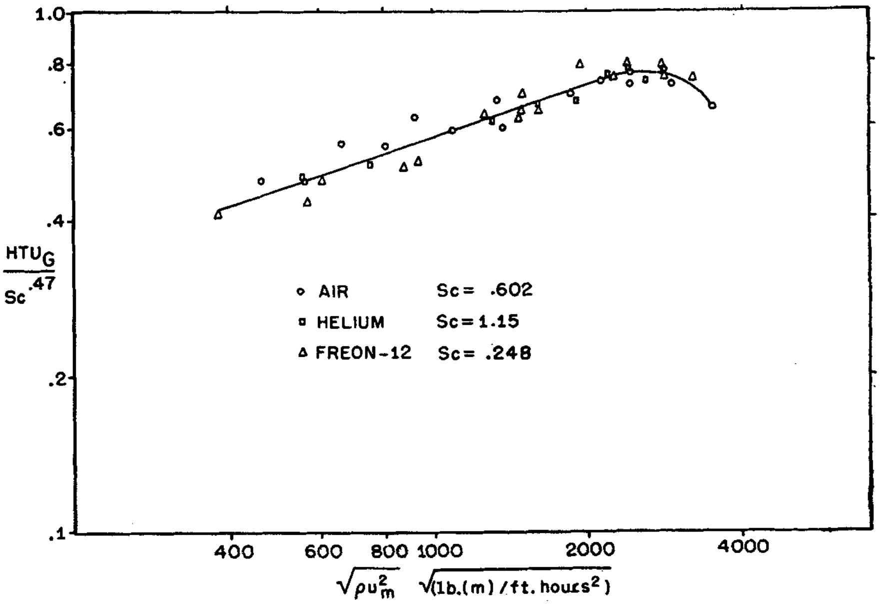

FIG. 10. H.T.U.$_G$ AS A FUNCTION OF THE INERTIA GROUP AND SCHMIDT NUMBER; $L = 1{,}575$.→

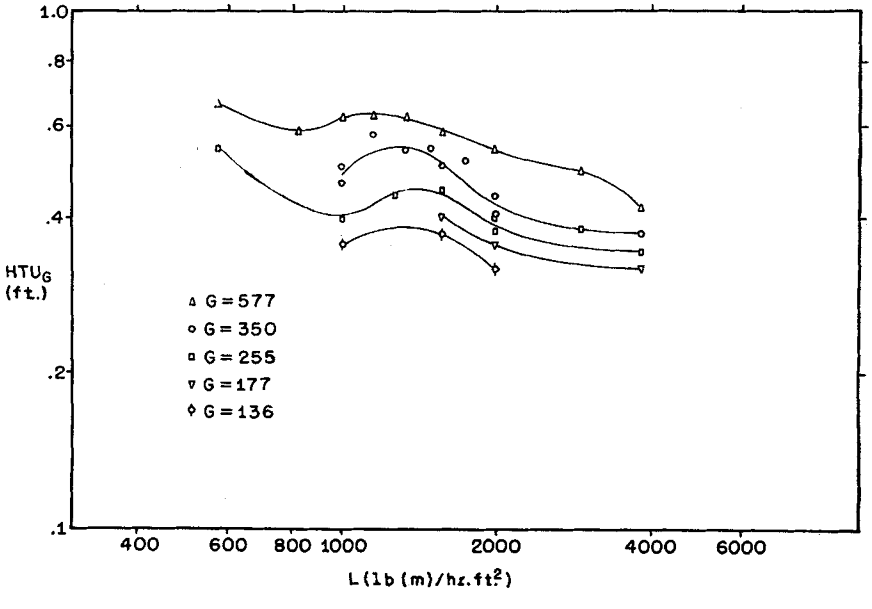

FIG. 11. H.T.U.$_G$ AS A FUNCTION OF THE LIQUID RATE.→

the use of true H.T.U. values, as all the data used were obtained at a single packed length.

It is possible to solve for the true H.T.U. if two apparent values H.T.U.$'_{23}$ and H.T.U.$'_{24}$ are known for two packed lengths Z_{23} and Z_{24} by use of Equation (30):

$$\text{H.T.U.}_{24} = \frac{\text{H.T.U.}'_{24}\ \text{H.T.U.}'_{23}\ (Z_{24}-Z_{23})}{Z_{24}\ \text{H.T.U.}'_{23}-Z_{23}\ \text{H.T.U.}'_{24}} \quad (15)$$

To evaluate the end effect in this work, bed lengths of 13 1/2, 7 1/2, and 2 in. were used. The 13 1/2-in. data scattered badly because the normal analytical errors were so greatly magnified by the approach to saturation at the top of the tower. These data therefore were not used in determining the end effect. The equivalent packed height was found to be essentially independent of gas rate (*see* Figure 9) with an average value of 3.51 in. This value was used to correct all the data for end effects.

All the H.T.U. data taken at a constant liquid rate of 1,575 lb. mass/(hr.)(sq. ft.) may be correlated by the single curve of Figure 10 which shows the effect of both gas velocity and gas properties on the gas film H.T.U. The average deviation of the data points from the line representing the mean is 4.25% with a maximum deviation of 12.5%; however, selection of $\sqrt{\rho u_m{}^2}$ in preference to G/μ as a basis for correlation cannot be conclusively justified with the present results. For example, in the region below the loading point the group $\rho^{0.2}$ (H.T.U.$_{G}/Sc^{0.5}$) can be expressed as a smooth function of G/μ which correlates the data nearly as well as Figure 10. Thus use of $\sqrt{\rho u_m{}^2}$ must be regarded as tentative pending further study. The effects of packing size and liquid properties also must be introduced into any general gas-film correlation.

The curves of Figure 11 show the effect of the liquid rate on the H.T.U. at several air rates. It is rather interesting that the H.T.U. does not bear a simple functional relationship to the liquid rate. It might be expected that as the liquid rate was increased the H.T.U. would decrease owing to the increased wetting of the packing. This would explain the shape of the curves at liquid rates below 900 lb. and above 1,500 lb. mass/(hr.)(sq.ft.). The sudden reversal of the liquid-rate dependence between these flow rates is difficult to explain on the basis of our present knowledge of flow distribution in a packed tower. One possibility is that the increased liquid flow over this range results in the blocking of some of the smaller flow passages, thereby preventing gas flow through these regions. This would effectively reduce the surface area available for transfer and result in higher values of the H.T.U.

It was observed that the average slope of the best straight line that could approximate each of the curves in Figure 11 is approximately the negative value of the slope of the curve of the H.T.U. vs. gas rate (below the loading point). This suggests that the group $(\sqrt{\rho u_m{}^2})_G/(\sqrt{\rho u_m{}^2})_L$ might serve as a parameter for general correlation as illustrated in Figure 12. This modulus has been used extensively for the correlation of flooding data. The average deviation of the data from the line representing the mean is 6.2% and the maximum deviation is 24%. In view of the limited scope of the liquid variables covered in this work, it is not possible to attach a final significance to this correlation. The influence of liquid density could enter in other ways, and the effect of other properties such as viscosity and surface tension remains to be considered.

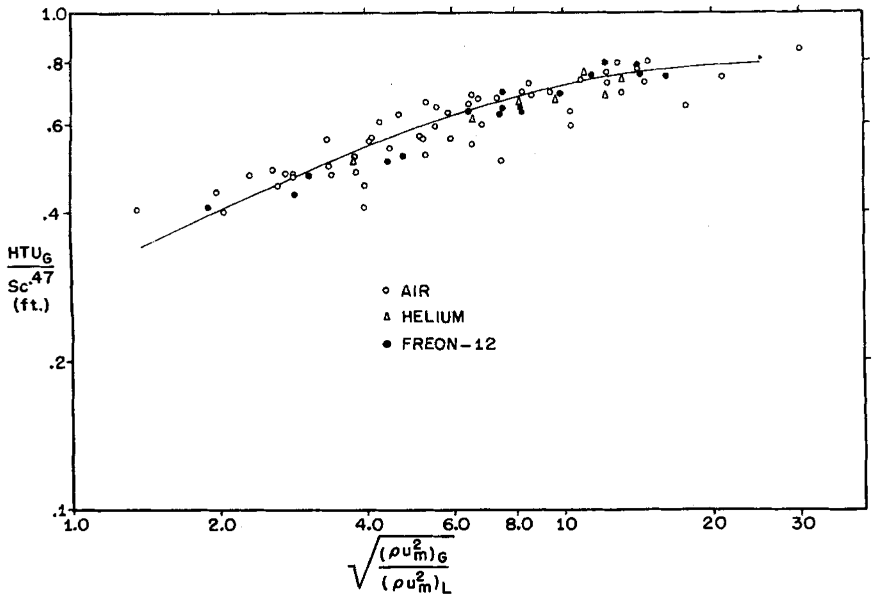

FIG. 12. H.T.U.$_G$ AS A FUNCTION OF THE FLOODING MODULUS.

DISCUSSION

In spite of the large volume of previous work on the subject, no completely reliable data exist for the resistance to mass transfer in the gas phase in a packed tower. From the numerous studies which have been made, a number of works have been selected to show the variation among investigators that has resulted even though they all used the same packing (1-in. Raschig rings) and inert carrier gas (air) and directed the experiments specifically toward the determination of gas-film data. The results plotted on the graph, Figure 13, for gas-film H.T.U. have been corrected for Schmidt-number effect to the system studied in this work (air-water), assuming the H.T.U. to be proportional to $Sc^{\frac{1}{2}}$. No correction was made for slight variation in packing properties since many of the investigators did not report these quantities.

Van Krevelin, Hoftijzer, and Van Hooren(*47*) reviewed a number of gas-film mass transfer studies to obtain a general equation. Since their equation is based largely on the absorption of NH_3 in water, it gives results similar to those found by Dwyer and Dodge (*14*), Vivian and Whitney(*49*), and Fellinger(*18*). The curve of Taecker and Hougen(*46*) pertains to systems without liquid flow and is not intended to be comparable to the other systems.

It will be noted that the technique of absorbing NH_3 and SO_2 in water has yielded much higher values of the H.T.U. than either the vaporization of liquids or the absorption of NH_3 and SO_2 in strong chemical solutions (absorption followed by a rapid chemical reaction). Fellinger found that absorption of NH_3 in H_2SO_4 gave H.T.U.'s which were 1.5 to 2.5 times smaller than those for the absorption in water. These acid data scattered somewhat and were not published, but these results would seem to indicate that the nature of the liquid-phase resistance to mass transfer in the NH_3-water system may have been improperly evaluated.

An alternative explanation of the discrepancy is the existence of an interfacial resistance, postulated by some authors *(11, 12, 16)*. Resistance to mass transfer usually has been considered to exist only in a gas film and a liquid film where equilibrium exists at the interface between them *(50)*; however it is possible that a large mass transport in one direction might result in an increase in the concentration of vapor molecules at the interface if the accommodation coefficient were very different from unity. Thus the actual partial pressure at the interface could be much higher than the equilibrium value and would result in a lower transfer rate than would be expected. Only fundamental experiments on the rate of NH_3 absorption in water will determine whether either or both of these factors is involved in the wide difference in results obtained from the two types of experiments.

The discrepancy between the various studies based on the vaporization of water is probably the result of the analytical difficulties involved in this type of investigation. Saturation of the carrier gas with water occurs very rapidly so that it is difficult to determine the driving forces for mass transfer at the tower exit with the required precision. To alleviate this condition, it is necessary to use very short packed sections, and therefore large relative errors are introduced in the measurement of the bed length and the effect of the transfer which takes place outside the packed section is magnified.

Although there is a wide variation in the absolute value of the H.T.U., most of the investigators indicate that it will vary with approximately the 0.3 power of the velocity at flow rates below the loading point. There is also the growing realization that this portion of the curve is not exactly a straight line and that above the loading point the value of the H.T.U. undergoes a marked decrease for reasons which are not yet entirely clear. The data of Sherwood and Holloway *(41)* show that the liquid-film H.T.U. is independent of the gas rate until the loading point is reached. At the loading point rapid improvement in the rate of mass transfer would indicate that at this point there is a marked change in the liquid flow pattern, which results in an increased contact area. It is not known whether this change is simply an increase in the wetted area of packing caused by increased gas-pressure drop, a rippling of the liquid surface, or a more severe disintegration of the liquid streams, but it seems reasonable to assume that the effect is also responsible for the improved mass transfer rates in the gas film. Figure 13 gives some indication of the importance of the area effect. The curve of Taecker and Hougen *(46)* was obtained by using completely wetted porous packing without liquid flowing. The values of the H.T.U. that were obtained by this procedure are approximately one half those obtained in this work. This also suggests that there would be a great dependence of the gas-film H.T.U. on the liquid being used, since the fraction of the total available area that is wetted will depend on the characteristics of the wetting fluid.

The dependence of the gas-film H.T.U. on the liquid rate as determined by several investigators is shown in Figure 14. All the curves show a decrease in the H.T.U. with increase in liquid rate but the dependence on liquid varies widely. Dwyer and Dodge, Yoshida and Tanaka *(52)*, and Vivian and Whitney indicate that the H.T.U.

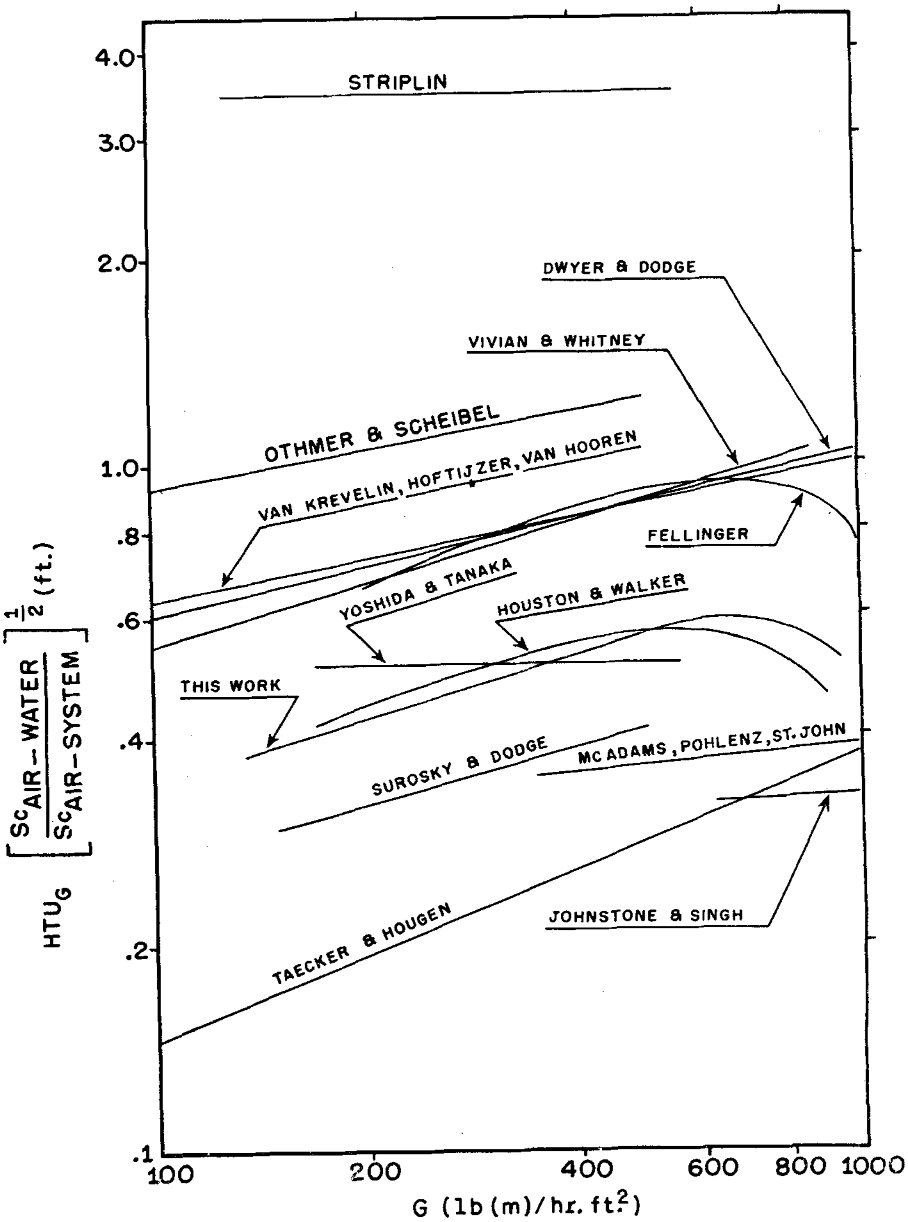

FIG 13. H.T.U.$_G$ AS A FUNCTION OF GAS RATE FOR VARIOUS INVESTIGATIONS.

varies as approximately $L^{-0.2}$ over the entire range of liquid rates measured. The curve of McAdams, Pohlenz, and St. John (*32*) is taken from their heat transfer results as their mass transfer results scattered excessively. The data of Surosky and Dodge (*45*) scatter too much to warrant the assumption that the H.T.U. is independent of liquid rate above a value of 1,200. Actually their data might be fitted as well by either a sloping line or an *S*-shaped curve. The curve of Fellinger based on liquid rates of 500, 1,500, and 4,500 shows the greatest dependence on liquid rate, the H.T.U. varying with $L^{-0.7}$ at low flow rates and changing gradually to $L^{-0.3}$ at high rates. The shape of the curve is almost identical with that suggested by Van Krevelin, Hoftijzer, and Van Hooren. Fellinger's three values could also be fitted by a curve of the type found in this work. It is difficult to draw a general conclusion from these studies or to say whether they offer either supporting or contradictory evidence for the unique liquid-rate dependence found by the authors.

The number of works devoted to the determination of the Schmidt-number effect is considerably more limited. Houston and Walker (*22*) absorbed NH_3, acetone, methanol, and ethanol from air into water and found that the H.T.U. was proportional to the diffusivity to the —2/3 power. Scheibel and Othmer (*38*) absorbed acetone and methyl ethyl ketone from air into water and obtained data which indicated that the H.T.U. was proportional to the diffusivity to the —1/2 power. However, in trying to correlate their data with the data of other investigators (*14*) on the absorption of NH_3, they were led to the final conclusion that the correct exponent on the diffusivity was —1. The results of both Houston and Walker and Scheibel and Othmer have possible sources of uncertainty in that it is necessary to correct for the liquid-film resistance which is present. In addition, the variation in diffusivity is small.

These difficulties can be avoided by the technique of vaporizing pure liquids into air. Mehta and Parekh (*33*) vaporized water, methanol, benzene, and toluene and found that the H.T.U. was proportional to $D^{0.17}$. Surosky and Dodge (*45*) used water, methanol, benzene, and ethyl butyrate as the liquids and found that their data could be correlated by use of H.T.U. proportional to $D^{0.15}$. The data of Simkin (*43*) and Chrisney (*9*) on the vaporization of solids and liquids from packed beds in the absence of liquid flow indicate that the H.T.U. is proportional to $D^{-0.36}$. This suggests that in the vaporization of pure liquids with liquid flow there are complications being introduced by the manner in which the various liquids wet and flow over the packing. The possible error from this source has been discussed previously.

It is possible to avoid the foregoing difficulties by the technique used in this work, that of using a single liquid for vaporization and varying the Schmidt number through the use of different carrier gases. It is necessary however to ascertain that the various gases do not have different effects on the liquid-flow pattern and effective mass transfer surface. If the gas does cause a change in the liquid flow, it should do so primarily by the pressure it exerts. Elgin and Weiss (*17*) and Jesser and Elgin (*23*) have found that the liquid holdup in a packed tower is independent of the gas-flow rate at constant liquid rate up to the flooding point. In addition, it was found in this work that the pressure drop across the packing was the same for all three gases at the same value of the inertia group. With this group as the correlating modulus, therefore, the relative mass transfer rates should be independent of the influence of the pressure drop on flow pattern and effective surface.

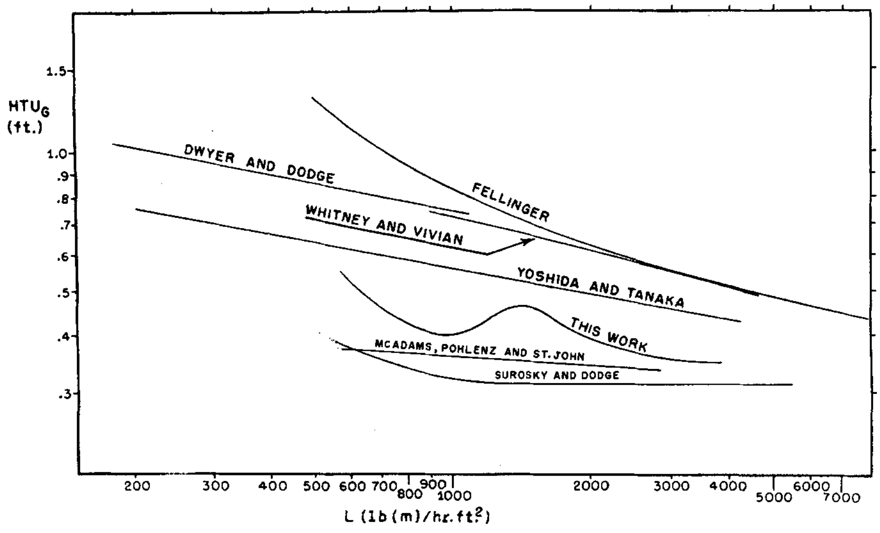

FIG. 14. H.T.U.$_G$ AS A FUNCTION OF LIQUID RATE FOR VARIOUS INVESTIGATIONS; $G = 200$.

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NOTATION

a = area available for mass transfer per unit volume of packing, sq.ft./cu.ft.
C_m = molal heat capacity at constant presure, B.t.u./(lb. mole) (°F.)
C_p = heat capacity at constant pressure, B.t.u./(lb. mass) (°F.)
D = diffusivity, sq.ft./sec.
d = diameter, ft.
G = mass velocity of the gas stream based on the superfiicial area, lb. mass/(hr.) (sq.ft.)
H = height, ft.
h = heat transfer coefficient, B.t.u./(hr.) (sq.ft.) (°F.)
h' = pseudo heat transfer coefficient defined by Equation (10)
H.T.U.$_G$ = height of a transfer unit for the gas film, ft.
H.T.U.$'_G$ = height of a transfer unit for the gas film not corrected for end effects, ft.
j_d = mass transfer factor defined by Equation (4)
j_h = heat transfer factor defined by Equation (3)
j'_d = mass transfer factor defined by Equation (12)
j'_h = heat transfer factor defined by Equation (12)
k = thermal conductivity, B.t.u./(hr.) (sq.ft.) (°F./ft.)
k_G = gas-film mass transfer coefficient, lb. moles/(hr.) (sq.ft.) (atm.)

L = liquid-flow rate based on the superficial tower area, lb. mass / (hr.) (sq.ft.)
M_m = mean molecular weight of the gas stream, lb. mass/lb. mole
M_v = molecular weight of the diffusing (evaporating) species, lb. mass/lb. mole
N = rate of transport of vapor, lb. moles/ (hr.) (sq.ft.)
N.T.U. = number of transfer units
P = total pressure, atm.
p = partial pressure, atm.
p_f = film pressure factor, atm.
q = heat transfer rate, B.t.u./ (hr.) (sq.ft.)
t = temperature, °F., °C.
u = velocity, ft./sec.
u_m = average velocity based on the total cross-section area, ft./sec.
y = mole fraction
$y_f = p_f/P$
Z = packing length, ft.

Greek Letters

$\beta = \frac{k_G M_m p_f \gamma}{h'} C_p$
γ = a heat transfer factor defined by Equation (9a)
μ = viscosity, lb. mass/ (ft.) (hr.)
ρ = density, lb. mass/cu.ft.

Dimensionless Groups

Re = Reynolds number, $du\rho/\mu$
Sc = Schmidt number, $\mu/\rho D$
Pr = Prandtl number, $C_p\mu/k$

Subscripts

a = main stream
avg = average
d = mass transfer
G = gas film
h = heat transfer
i = interface
m = mean
m =molal
s = surface

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