Mass Transfer Studies in an Agitated Vessel

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A study of rates of dissolution of organic solid from a flat surface into turbulent liquid in a mixing tank of 6 in. I.D. is reported for five systems—benzoic acid–water, salicylic acid–water, salicylic acid–benzene, succinic acid–*n*-butanol, and succinic acid–acetone. Previous theories for the rates of mass transfer are reviewed and compared, and experimental results analyzed and correlated by an equation in terms of dimensionless groups similar to that of Hixson and Baum, Sherwood and Gilliland, and Rushton and Oldshue. The close agreement between the theoretical and experimental values of the exponent of the Schmidt number in this equation may provide new and significant evidence for the applicability of the surface-renewal theory to mass transfer from a flat solid surface into a turbulent liquid.

For free rotational agitation, a local mass transfer coefficient with respect to the position of a cast solid was detected quantitatively. However, the insertion of four baffles into the tank gave a uniform mass transfer coefficient regardless of the position of the cast solid. A decrease in the mass transfer coefficient was observed when baffles were used.

The advantages of the constant and stationary interface, the stability and simplicity of the apparatus, the possibility of duplicating experimental results, and the success of the detection of a local mass transfer coefficient suggest that the present apparatus and procedures could be used for the study of the theory of mass transfer rates from flat surfaces.

Among the basic concepts of mass transfer between phases which have been proposed, the film theory of Whitman and Lewis (*18*, *19*, *31*) and the Danckwerts surface-renewal theory (*2*) have attracted greater attention from chemical engineers than have others. The first theory pictured a stagnant fluid film at the interface representing the resistance to mass transfer. It was assumed that the solute passed through this film only by molecular diffusion. According to the theory, the mass transfer rate from the interface to the bulk of the phase can be represented by the equation

$$N = \frac{D}{x}(C_i - C)A \qquad (1)$$

Although the film theory has the advantage of supplying a simple picture of a complicated mechanism, it has been doubted whether a stagnant film exists under any turbulent condition, and the usual assumption of a stagnant film at the interface was abandoned by Danckwerts in his surface-renewal theory (*2*). Instead he assumed that the surface was continuously being replaced with fresh fluid. A new term, *surface-renewal rate*, was introduced without specification of the physicochemical properties of the system and the hydrodynamic conditions. Based on this concept, the mass transfer rate equation, similar to Equation (1), can be written as

$$N = \sqrt{Ds}\,(C_i - C)A \qquad (2)$$

Although Danckwerts proposed the surface-renewal theory for gas absorption in a packed tower, a mechanism can be postulated to permit the concept to be applied for solid-liquid or liquid-liquid systems. When the theory was proposed in 1951, Danckwerts made no effort to compare the foregoing equation with published experimental measurements. He regarded the theory as tentative until it was confirmed experimentally. Recently, however, the transient rates of gas absorption by use of a rotating drum for a few systems including a case of mass transfer with chemical reaction were measured by Danckwerts and Kennedy (*3*). Comparing these results with the steady state of gas absorption in the packed tower, they tentatively concluded that none of the present theories by Whitman and Lewis and by Danckwerts gave an adequate description of the absorption process. The recent work of Lynn, Straatemeier, and Kramers (*23*) however shows the applicability of the penetration theory (*11*) of the Higbie

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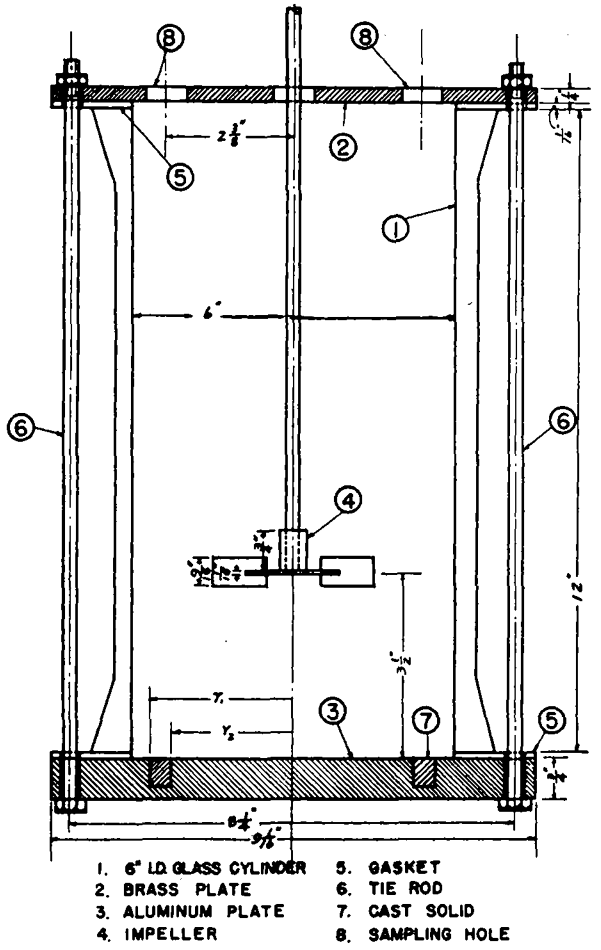


Fig. 1. Agitation tank.

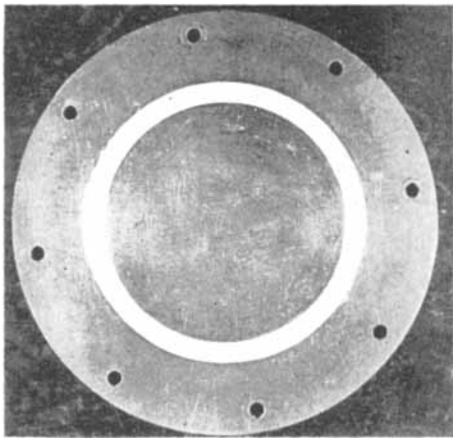


Fig. 2. Bottom plate with cast solid (plate 1).

type to the absorption of sulfur dioxide by water and by aqueous solutions of HCl, $NaHSO_3$, and NaCl in relatively long wetted-wall columns, in short wetted-wall columns, and in wetted spheres singly and in columns. In the presence of small concentrations of a surface-active material the rate of absorption in long wetted-wall columns is very well explained by the penetration theory, which predicts that the rate of absorption is proportional to the diffusivity to the one-half power. It would be premature, therefore, to conclude from these relatively few results that the surface-renewal theory was an unreliable guide. More experimental evidence would be welcomed.

It should be pointed out that both the film and the surface-renewal theories adopted the concentration difference as a driving force; therefore, the equation for rate of mass transfer through a single-phase resistance,

$$N = k(C_i - C)A \qquad (3)$$

is common between the two theories with the different significances attached to the individual mass transfer coefficient k; i.e.,

$$\text{Whitman-Lewis} \qquad k = \frac{D}{x} \qquad (4)$$

$$\text{Danckwerts} \qquad k = \sqrt{Ds} \qquad (5)$$

Up to the present time there have been very few data published which were obtained for the specific purpose of investigating the effect of diffusivity on the mass transfer coefficient. As pointed out by Sherwood and Gordon (*9*), the previous experimental data showed that the effect of diffusivity on the mass transfer coefficient might be represented by the power function $k \propto D^v$ where the exponent v had been reported to be somewhere between 0.15 and unity. A table summarizing these experimental results has been published (*9*). It should be noted that for the study of the additivity of individual mass transfer resistances, Sherwood and Gordon assumed that an individual mass transfer coefficient should vary with molecular diffusivity to the one-half power. Thus it may be concluded that the effect of diffusivity has not been satisfactorily established and that further experimental work for this purpose is desirable.

Besides the two foregoing theories, the kinetic theory of Miyamoto (*24, 25, 26*) and Kishinevskii's surface-renewal theory without penetration (*4, 15, 16, 17*) were also interesting because the existence of a stagnant film at the interface was not assumed. For the process of dissolution of a solid into liquid Miyamoto separated the process into two parts, namely, escape of the solid molecules into the liquid phase and deposition of the dissolved molecules on the solid surface. The further assumption was that, among molecules which made up the solid surface, only those with components of vibration energy at right angles to the interface greater than a threshold value could enter into the liquid phase and, among dissolved molecules, only those which collided with the interface at velocities with components at right angles to the interface greater than a threshold value could deposit on the surface. The mathematical expression for the individual mass transfer coefficient does not include the terms for hydrodynamic conditions of the operations. Thus the application of his theory to equipment design was limited. Kishinevskii's concept was similar to the surface-renewal theory except that the surface became instantaneously saturated with the solute on exposure (*4*). Thus molecular diffusion played very little part in the determination of the rate of absorption. But, as pointed out (*4, 9*), there is strong evidence that the mass transfer coefficient varies with molecular diffusivity.

In chemical engineering research some indirect approaches to evaluating agitation by some measurable quantities have been applied; for example, the studies on the rate of solution of suspended solid pellets were employed for this purpose. The earliest study on the rate at which solids dissolve in their own solution was published by Noyes and Whitney (*27*). The materials (benzoic acid or lead chloride) were cast in the form of cylindrical sticks on glass cores and, after insertion into wide-mouthed bottles containing water, were rotated at a constant rate. The rate of solution was explained on the assumption that a very thin layer of saturated solution was formed at the surface of the solid and that the rate at which the solid dissolved was governed

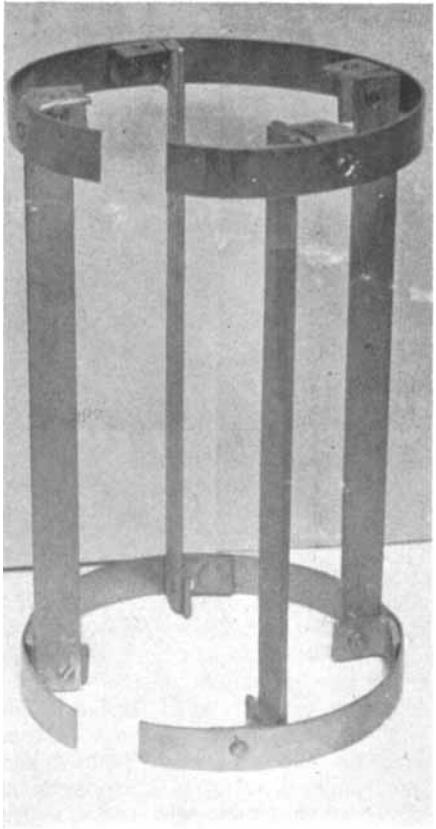


Fig. 3. Baffles.

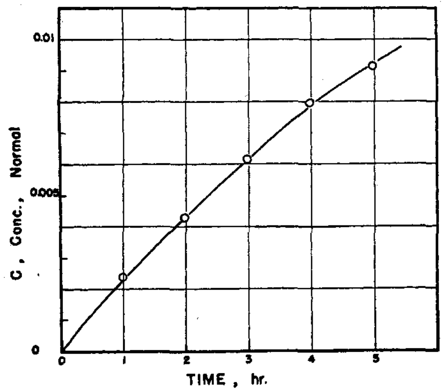


Fig. 4. Concentration of solution.

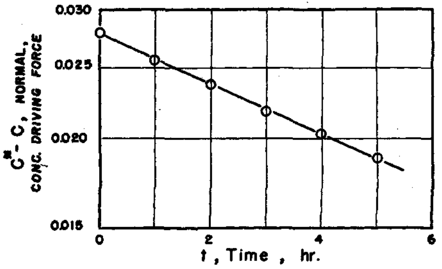


Fig. 5. Determination of mass transfer coefficient *k*.

by the rate of molecular diffusion from this saturated layer into the main body of the solution. Since then most workers have suspended the solid pellets or particles in the solution for the study of mass transfer between solid and liquid systems in a stirred vessel. For this case Hixson and Crowell (*12*) developed the so-called "cube-root law" for the determination of the mass transfer coefficient on the assumption that the surface area of the solid pellets or particles was proportional to the two-thirds power of the volume or the weight of dissolving solids. Later Hixson and Wilkens (*14*) extended the work, still suspending the benzoic acid tablets in water, to include the effects of stirring speed, size of equipment, and the use of baffles. Since the exact expression of the cube-root law was algebraically complex, Hixson and Baum (*13*) proposed an approximate equation with the simplified assumption that the total area of the surfaces of the suspended solid was constant and that the change of concentration as the dissolution proceeded was small so that the average concentration driving force might be expressed by logarithmic means. Mass transfer data for a series of dimensionally similar agitators in which solids were suspended in liquid were reported. Their experimental results were correlated by the similar equation used by Gilliland and Sherwood (*8*):

$$\left[\frac{kl}{D}\right] = K\left[\frac{\mu}{\rho D}\right]^{b}\left[\frac{R\,d^{2}\rho}{\mu}\right]^{c} \qquad (6)$$

The main purpose of the present research was to pursue the Danckwerts type of surface-renewal theory to see whether the theory could be extended to a solid-liquid system. An attempt was made to relate the surface-renewal rate to the operating conditions and physical properties of the agitated system. The present research was limited to the case of mass transfer by physical means only. It was decided to measure rates of solution of organic solid into the turbulent liquid from the flat surface. The operation was made in a mixing tank rather than in a packed tower with its complicated and possibly varying flow patterns or in a wetted-wall column with its rippling or in a mixing tank containing solid particles suspended in agitated liquid with varying surface area. The suspension of the solid particles in the agitated liquid was accompanied by other disadvantages. There was difficulty in obtaining uniform shape of the particles. During agitation the solid particles could be crushed or fractured and the surface area might be changed. A variation in the surface area might also be caused by the agglomeration of the particles. The particles might not have remained suspended in the liquid but have fallen to the bottom of the tank. Thus the advantages of the choice were, first, to have a single fluid resistance to mass transfer and, second, to maintain the interfacial area constant and stationary. An important reason for this system was also that it was intended to be a model of a liquid-liquid system with a fixed interface area.

The second object of the research was to study the method of anticipating the agitator performance by mass transfer data. Specifically, the effects of the position of cast solid, the stirring speed, the position of the impeller, the baffling, and other physicochemical properties on mixing or mass transfer were to be investigated along with the first objective. Since the variation of diffusivities of different organic solids in the same solvent was not great enough for direct determination of its effect on the mass transfer coefficient, several pairs of organic solid-liquid systems were used. If the Danckwerts type of surface-renewal theory could be applied to explain the rate of dissolution, the experimental data could be correlated by Equation (6), in which the exponent to the Schmidt number should be 0.5, as according to Danckwerts's theory k should be proportional to $\sqrt{D}$.

$$\left[\frac{kl}{D}\right] = K\left[\frac{\mu}{\rho D}\right]^{0.5}\left[\frac{R\,d^{2}\rho}{\mu}\right]^{c} \qquad (7)$$

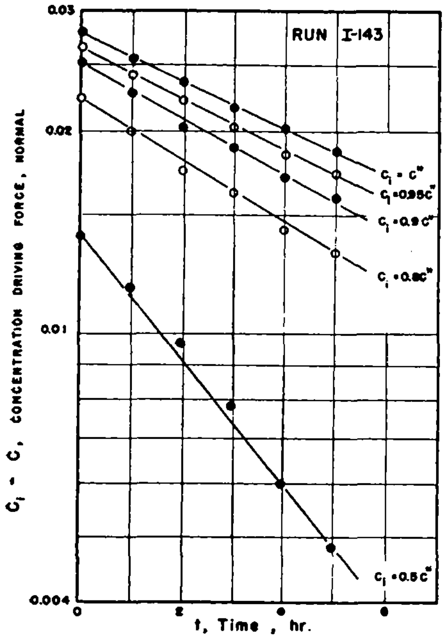
EXPERIMENT

The experimental runs were made in a mixing tank the dimension of which are shown in Figure 1. The bottom plate was aluminum with a ring depression in which organic solid was cast. Four bottom plates with different rings were used. Figure 2 shows the photograph of a bottom plate and Table 1 lists the radii of the circular depressions for four plates.

The solution was agitated by a turbine-type impeller with six flat blades. The impeller was placed at the center of the

TABLE 1. DIMENSIONS OF THE CAST ORGANIC SOLID

Plate	Inside radius r_1, cm.	Outside radius r_2, cm.	Surface area A, sq. cm.	Mean radius r_m, cm.
1	6.568	7.620	46.873	7.094
2	5.387	6.317	34.183	5.852
3	3.810	4.666	22.793	4.238
4	0.635	2.009	11.415	1.322



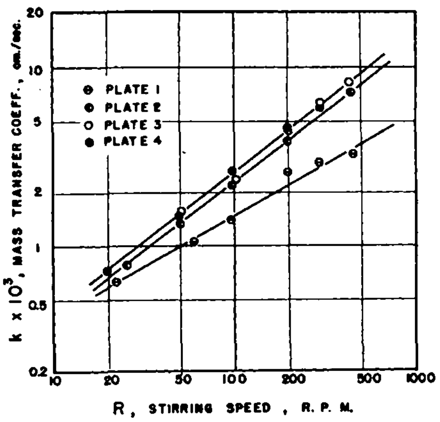


Fig. 7. Effect of stirring speed on rates of dissolution.

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Fig. 6. Effect of interfacial concentration.

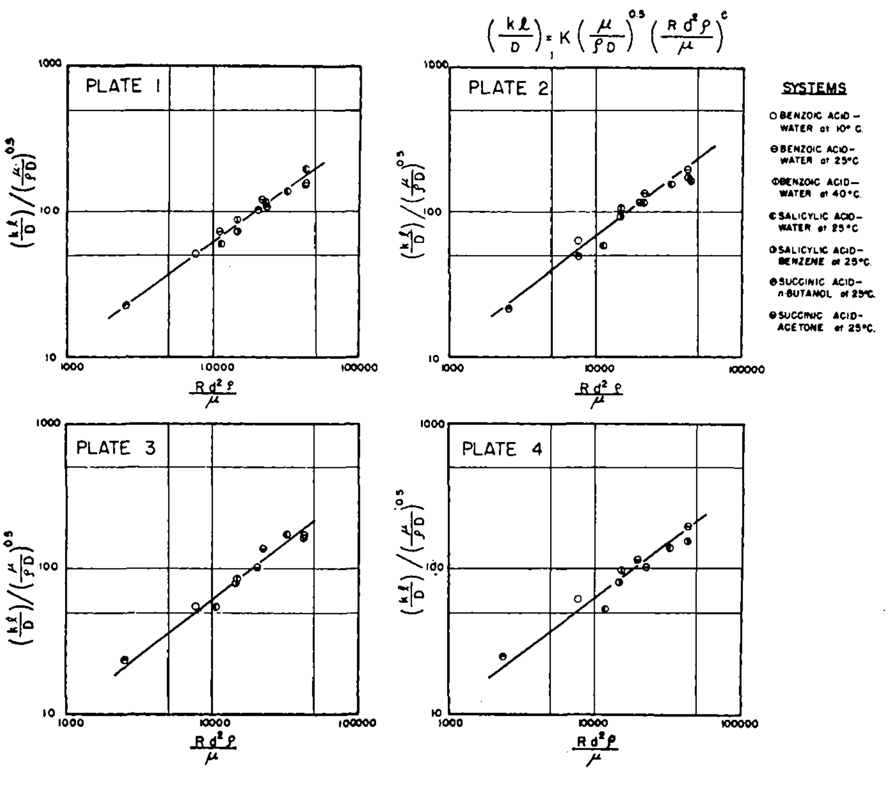

Fig. 8. General correlation for rate of dissolution with baffling.

TABLE 2. RANGE OF EXPERIMENTAL VARIABLES AND PHYSICAL PROPERTIES

Variables	Units	Range
Density	g./cc.	0.7990 ~ 0.9998
Viscosity	centipoises	0.3982 ~ 3.205
Diffusivity	sq. cm./sec.	0.2725×10^{-5} ~ 1.850×10^{-5}
Stirrer speed	rev./min.	50 ~ 500
Temperature	°C.	10 ~ 40
Stirrer position	in.	2.5 ~ 5.5
Mass transfer coefficient	cm./sec.	0.360×10^{-3} ~ 8.897×10^{-3}
Sherwood number		1,070 ~ 13,700
Schmidt number		208 ~ 14,800
Reynolds number		2,250 ~ 48,760

tank and was driven by a variable-speed motor. The speed of the stirrer varied within 3% during a run. For the runs with baffling four narrow vertical baffles ¾ in. wide and ⅛ in. thick were inserted. A photograph of the baffles is shown as Figure 3. The agitation tank was placed in either a constant-temperature bath or a constant-temperature room for each run. The temperature fluctuated in the constant-temperature bath by ±0.2°C. and in the constant-temperature room by ±0.5°C. The selection of the system was one of the most difficult problems in this research because the chosen system had to meet several criteria, i.e., no cracking of the cast solid, adherence of the cast solid to the ring depression, wide range of diffusivities, and large variation of the Schmidt number. The following systems were selected on the basis of these conditions: benzoic acid–water at 25°, 10°, and 40°C.; salicylic acid–water at 25°; salicylic acid–benzene at 25°C.; succinic acid–*n*-butanol at 25°C.; and succinic acid–acetone at 25°C.

The main difficulty with the present work was to prepare a flat surface of organic solid without any trace of cracking. The present authors recommend, after trying several methods, slow melting of the solid and gradual cooling of cast solid. The surface of the organic solid was carefully smoothed to the same level as the aluminum plate with a sharp spatula or a razor blade. Finally the surface was rubbed with fine sandpaper, then washed and dried.

When the agitation tank was set ready for a run, the motor was started and allowed to come to speed. The solvent was added from a funnel of which the end tip was close to the tank wall so that the solvent fell along the wall. In this manner 3 liters of solvent could be admitted with very little initial turbulence caused by pouring. As soon as the solvent was added, the funnel was removed and the timer started. A run was then in progress. At a constant interval of time, e.g., 15, 30, or 60 min., depending on the rate of dissolution, a sample of the solution was removed by a 10-cc. pipette and later analyzed. Ten cubic centimeters of fresh solvent was added immediately after the withdrawal of the sample to make up for the amount which had been removed. The sampling point was kept constant during this investigation. The rotating speed of the stirrer was counted every 10 min. by use of a strobotac. The concentration of the samples was readily determined by a straight titration with sodium hydroxide solution of proper concentration with phenolphthalene used as an indicator.

RESULTS

Mass Transfer Coefficient

The concentration change of the solution in the tank with respect to time can be expressed by the following equation:

$$\ln\frac{C_i - C_n}{C_i - C_{n+1}} = k\frac{A}{V}(t_{n+1} - t_n) \qquad (8)$$

which is developed from the following two equations for a batch operation:

$$N = kA(C_i - C) \qquad (9)$$

$$N\,dt = V\,dC \qquad (10)$$

Therefore, when the concentration of liquid phase in the mixing tank is plotted against time, the line should be an exponential curve, as shown in Figure 4. Furthermore, if the run was made for a longer period, the curve would flatten out and become asymptotic to the saturated concentration. Theoretically, the time necessary to reach the saturated concentration is infinite according to Equation (8).

The initial mass transfer coefficient was not measured because the present experimental procedure was not prepared for that purpose. It was observed that except in a few cases out of one hundred fifty runs the mass transfer coefficient for the first interval was usually the same as that observed in the later intervals. A few deviations were most likely due to the hydrodynamic discrepancy caused by the method of pouring solvent into the tank at the beginning of a run.

The removal of dissolved solute owing to intermediate sampling was corrected by the introduction of correction factor ϕ; that is, the true mass transfer coefficient k must be the sum of the apparent mass transfer coefficient k' and the correction factor ϕ.

$$k = k' + \phi \qquad (11)$$

where

$$k' = \frac{\ln \dfrac{C_i - C_n}{C_i - C_{n+p}}}{\dfrac{A}{V}\, p(t_{n+1} - t_n)} \qquad (12)$$

and

$$\phi = \frac{\displaystyle\sum_{m=n+1}^{n+p-1} \ln \frac{C_i - rC_m}{C_i - C_m}}{\dfrac{A}{V}\, p(t_{n+1} - t_n)} \qquad (13)$$

For the determination of correction factor ϕ, a plot of $\ln\,[(C_i - rC_m)/(C_i - C_m)]$ against C_m will facilitate the calculation. However, the correction was usually very small, i.e., within 3% of the true mass transfer coefficient. Particularly when the solution was dilute and the amount of the solution in an agitation tank was large, the correction was usually not necessary.

During a run the mass transfer coefficient was assumed to be constant. The assumption was sound because the plot of log concentration driving force vs. time was straight through the points as shown in Figure 5. If the assumption did not represent the phenomenon observed, there would not be a single straight line through all the points for a run. A single straight line through the plots also implies that the concentration difference between the saturated solution and the solution was the right driving force for the rate of dissolution. The reproducibility of the experimental results was checked at random and was found to be satisfactory. The deviation in mass transfer coefficient was usually within 6%.

In addition to the advantages of constant interfacial area and the simplicity of the apparatus, the possibility of duplicating the experimental results within the experimental error was encouraging. For this reason the present apparatus would be recommended for the study of mass transfer theory in the future.

Several evidences of the existence of an interfacial resistance have been reported (*3, 6, 11, 30*) for gas-liquid mass transfer studies. However, in these reports the authors stated that for a long period of the exposure, say 10 sec. for the carbon dioxide–water system as reported by Higbie (*11*), the effect was not observed. Emmert and Pigford (*6*) showed that after a very short period of exposure $k/k°$ became asymptotic to unity for the same system; here $k°$ was the mass transfer coefficient if no interfacial resistance was assumed. In the study of rates of dissolution of benzoic or salicylic acid into water under free convection, Wilke and his associates (*32*) concluded that the assumption of interfacial equilibrium was not significantly in error for the organic acids involved. In the study of rate of dissolution of solid from the cylindrical surface, Noyes and Whitney (*27*) explained the action on the assumption that the solution at the interface was saturated. It would appear to be safe to assume that there is no appreciable resistance to mass transfer in this study other than that resulting from hydrodynamic and diffusional considerations.

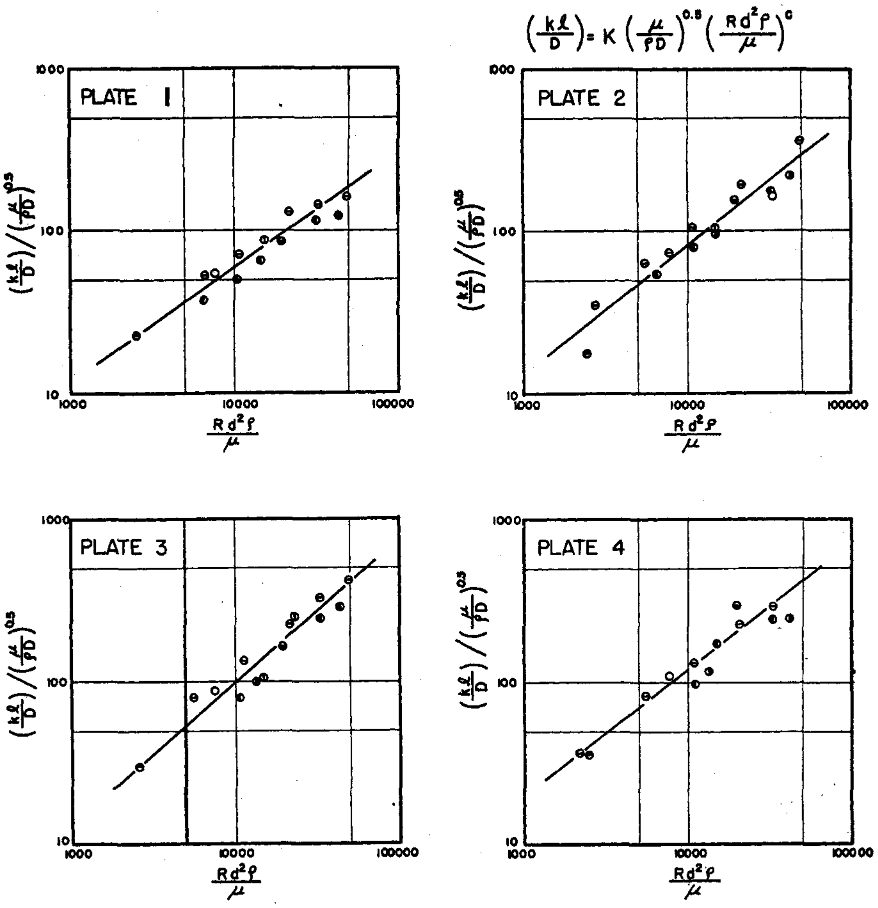

Fig. 9. General correlation for rate of dissolution without baffling.

TABLE 4. CORRELATION OF RATE OF DISSOLUTION

$$[kl/D] = K[\mu/\rho D]^b[(Rd^2\rho)/\mu]^c$$

	Plate	K	b	c
With baffles	1	0.1283	0.475	0.693
	2	0.0722	0.523	0.769
	3	0.0973	0.501	0.702
	4	0.2613	0.467	0.630
Without baffles	1	0.2700	0.499	0.596
	2	0.0431	0.526	0.809
	3	0.0915	0.484	0.865
	4	0.1280	0.422	0.948

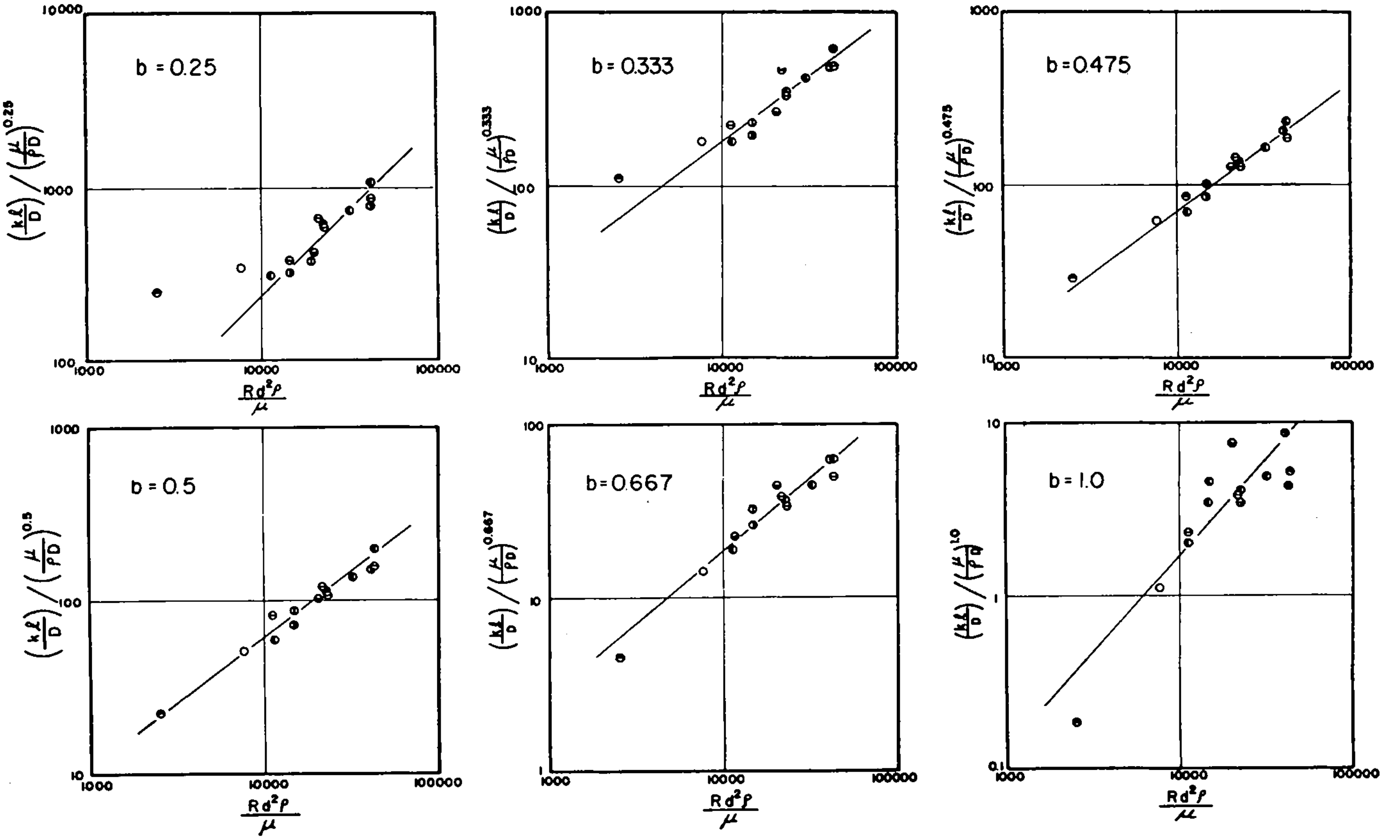

Fig. 10. Comparison of correlations with different exponents of Schmidt number used.

Figure 6 shows the plots of log driving force against time based on the assumption that the interfacial concentration was not at equilibrium. When the solution at the interface was less saturated, the points deviated more from a straight line. As will be shown later, the points for five different systems did not deviate excessively from the general correlation, an implication that the effect of interfacial resistance was negligible in this study or at least that the ratio of k to k° was constant for all the systems.

Effect of the Stirring Speed

Figure 7 shows the effect of the speed of the stirrer on the mass transfer coefficient. For illustration, here the experimental results of the benzoic acid-water system at 25 ± 0.5 °C. without baffling were used. For all runs made, the effect can be generalized that the mass transfer coefficient was proportional to the speed of the stirrer to the βth power.

$$k = \alpha R^{\beta}$$

The constants α and β depended on the plate and on whether baffling was applied. A similar observation for the power-speed study was reported (*28*) with the exponent β between 2 and 3. However, there was no appreciable distinction in α and β between plates 3 and 4, which were the smaller rings in mean radius. With the use of baffles, the effect of the speed of the stirrer on the coefficient could be represented for each system by a single line regardless of the position of a cast solid.

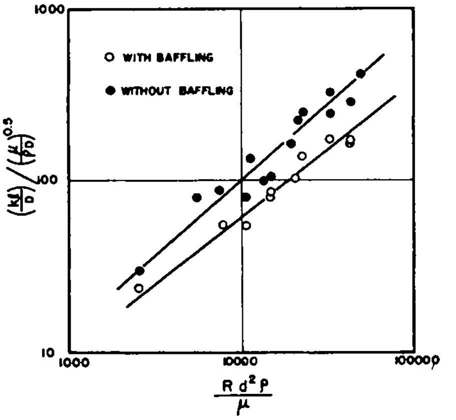

Fig. 11. Effect of baffling (plate 3).

Figure 7 also shows that higher mass transfer rates were obtained with cast solid rings of small diameter than with the large rings. In an unbaffled tank, with the impeller centrally located and the mixing shaft vertical, the entire tank is set in a rotary flow pattern with the highest linear velocities at the center of the tank. As the ring approaches the wall of the tank, the velocity will be lower and therefore the mass transfer coefficient lower.

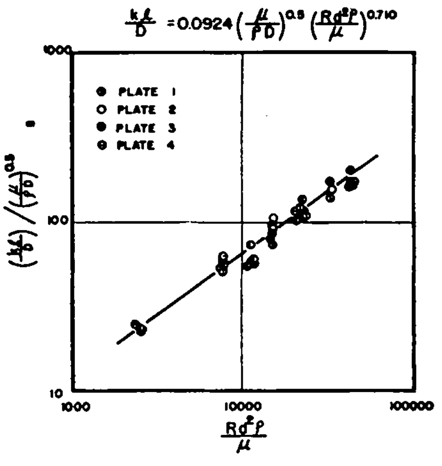

Fig. 12. General correlation for rate of dissolution with baffling.

The range of experimental variables and the physical properties of the systems studied in the present research are summarized in Table 2. The range of the Schmidt number was rather wide, but the Reynolds number was limited.

Correlation of Data

Equation (6) suggested that when $[kl/D]/[\mu/\rho D]^{b}$ was plotted against $[(Rd^2\rho)/\mu]$, a correlation would be obtained with a proper choice of the exponent b. Although Danckwerts did not claim that his theory would work for solid-liquid mass transfer, the surface-renewal theory may be pictured as still applying to solid-liquid interface by the following postulation. A thin liquid layer of saturated concentration was assumed to exist at the interface. Then the controlling mass transfer process between solid and liquid could be pictured as that between the saturated liquid layer and liquid phase. Here the turbulence was assumed to reach the surface of the saturated layer.

The physical properties of the systems for the calculations of the dimensionless groups were obtained from the literature or by measurements. Table 3* tabulates all the experimental results and the calculated dimensionless groups. The experimental results were plotted by $[kl/D]/[\mu/\rho D]^{1/2}$ vs. $[(Rd^2\rho)/\mu]$ on the assumption that the surface-renewal theory might be applicable.

The results seemed to be satisfactory as shown in Figures 8 and 9. For an individual system the previous discussion showed that the mass transfer coefficient varied with the speed of the stirrer to a certain power. This implied the linearity of the plots of the mass transfer coefficient against the stirrer Reynolds number. Then the method of least squares could be applied for the determination of the best values of the exponents b and c of Equation (6). Table 4 lists the constant K and the exponents b and c determined by the method of least squares.

As discussed before, if the mass transfer mechanism follows the surface-renewal theory, the exponent of Schmidt number, b, should be 0.5. The close agreement between the empirical and theoretical values for the exponent b, as tabulated in Table 4, may then support the possible application of the surface-renewal theory to the process of physical dissolution as conducted in the present research. In order to show the differences of the correlation due to different values of the exponent b, Figure 10 was prepared with b equal to 0.25, 0.333, 0.5, 0.475, 0.667, or 1. The experimental data of plate 1 with baffling were taken for comparison. The mean deviation was 3% for $b = 0.475$, which was determined by the method of least squares, where it was 5.8% when the theoretical value of 0.5 was adopted for b.

For solid-liquid mass transfer the relation between the mass transfer coefficient and the molecular diffusivity has not been established as definitely as for other systems. In order to explore the effect of the Schmidt number on mass transfer in turbulent flow, data were obtained on the rate of solution of cast tubes and cylinders by Linton and Sherwood (*22*). Lin et al. (*20*) measured transfer rates from a flowing electrolyte to the surface of an electrode. These data were correlated with respect to Schmidt number to the two-thirds power. Later Lin, Moulton, and Putnam (*21*) presented a theoretical analysis of mass transfer between a turbulent-fluid system and the wall. The fluid was divided into three layers, namely, the very thin sublaminar layer adjacent to the wall, the turbulent core, and the buffer region between the core and laminar layer. Analysis of these showed that there was no definite laminar film for pure molecular diffusion near the wall. The concept was to introduce into the sublaminar layer a small appropriate amount of eddy which decreases rapidly to zero at the wall. The general mass transfer equation included ϕ_D, which is an involved function of the Schmidt number and the friction factor. Eisenberg, Tobias, and Wilke (*5*) studied the rates of mass transfer at circular cylinders rotating about their axes in the center of stationary cylinders by means of solid dissolution and electrolytic redox reactions. The correlation was in terms of Schmidt number to the 0.644 power. They (*32*) also evaluated the mass transfer coefficient from limiting rates of electrolysis and from rates of dissolution of organic solids where no mechanical agitation was applied. For this case the correlation was made with respect to Schmidt number to the 0.25 power. Gaffney and Drew (*7*) determined the mass transfer coefficient for three organic solvents flowing through the packing of organic acid pellets. The height of the transfer unit increased with the 0.58 power of Schmidt number. Only one previous study, that of Hixson and Baum (*13*), correlated data with the 0.5 power to the Schmidt number.

However, Hixson and Baum did not think that in their study the value of the exponent b to the Schmidt number could be accurately determined because of the narrow range of values for the Schmidt number at a given range of the Reynolds number. They selected 0.5 tentatively because this value gave the best correlation for most points and because it was approximately the same as the value 0.44, which was used by Gilliland and Sherwood (*8*) to correlate their data for mass transfer into turbulent air. Their research was not intended to establish this exponent but was to indicate a method for applying the dissolution constant to the problem of designing and determining the operating characteristics of mixing equipment.

Effect of Baffling

The radial flow, as the result of the application of baffles, has vertical and lateral flow currents that are desired for most mixing operations. But the insertion of the baffles was accompanied by the decrease of mass transfer rate. Apparently the surface-renewal rate for the operation without baffles was higher than that with baffles. The same phenomenon was reported by Hixson and Wilkens (*14*) for the rate of dissolution of suspended solid particles into the liquid in a mixing tank.

The magnitude of the decrease in the rate of dissolution upon insertion of baffles was different for each plate, but the magnitude increased as the cast solid ring became small. When the ring was big enough to get close to the wall of the tank, there was almost no effect. The decrease of the mass transfer coefficient due to baffling is shown in Figure 11, results obtained for plate 3 being used.

Different effects with respect to the number of baffles were not studied. According to Hixson and Wilkens, the introduction of more than four baffles appeared not to influence the dissolution rate further for systems of solids suspended in liquid.

It has been recognized (*3, 10*) that the local value of mass transfer coefficient k would probably vary from one region to another in a packed tower. There are regions in the packing where the surface-renewel rate is high and other parts where the liquid is relatively stationary. This uncertainty and nonuniformity of local k value is the fundamental disadvantage of the use of a packed tower in the study of mass transfer theory. In this research the local k value was detected quantitatively as shown in the correlation of the data.

There was a much greater deviation in the exponent of the Schmidt number for the unbaffled tank than for the baffled tank, as shown in Table 4. With the Reynolds number used as the measure of the hydrodynamic condition in the tank, wider variation in the exponent of the Reynolds number also indicated strongly the variation of hydrodynamic conditions with respect to position. However, when the baffles were inserted, the variation of the local mass transfer coefficient was eliminated, as the experimental results of all four plates could be represented by a single line, as shown in Figure 12. Then a single correlating equation was sufficient to cover all plates if baffling was provided.

$$\left[\frac{kl}{D}\right] = 0.0924\left[\frac{\mu}{\rho D}\right]^{0.500}\left[\frac{R\,d^2\rho}{\mu}\right]^{0.710} \tag{14}$$

As compared with the packed tower, the success of the quantitative detection of the local k value would lead to the recommendation of the use of this apparatus for the study of mass transfer mechanism.

Effect of Surface Structure

The question of the effect of the different crystal structure at the interface was very difficult because of the impossibility of casting exactly the same surface each time. However, if the method of casting

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*Table 3 may be obtained as document 4957 from the American Documentation Institute, Photoduplication Service, Library of Congress, Washington, 25, D. C., for $3.75 for photoprints or $2.00 for 35-mm. microfilm.

was repeated in the same manner described in the experimental details, the results could be reproduced, an implication that although the surface might not have the same crystals at any point, its population would be almost the same for each casting. The constant k value during a run and the success of the general correlation for different systems might indicate that minor change in the crystal structure and its population on the surface did not play a dominant role in the present study. Similarly the small deviation of the smoothness of the surface has little effect on the rate of dissolution in the mixing tank, where the hydrodynamic conditions have a greater effect.

Effect of the Position of the Impeller

A change of the flow pattern in the tank caused by the change in the height of the impeller was observed as the change in the dissolution rate. For agitation with baffling, however, the change of the transfer coefficient was insignificant. The application of the baffles masked out the effect of the position of the impeller. Even in the case without baffling, plates 1 and 2 showed insignificant change in the mass transfer coefficient. Near the wall the flow would have greater influence from the wall than would other factors. Appreciable effects of the impeller position were observed for plates 3 and 4 when no baffles were attached. Qualitatively it could be stated that the dissolution rate decreased as the position of the impeller was raised. For power-consumption study (*28*) the effect of the stirring position has been observed but no definite quantitative expressions were obtained. For any quantitative conclusion concerning the effect of the impeller position, more data are needed.

CONCLUSIONS

The following conclusions can be reached from the results of the present investigation.

This study proposed the correlation of the experimental results for mass transfer from a flat solid surface into a turbulent liquid in terms of dimensionless groups with 0.5 used as the exponent of the Schmidt number. The fact of this satisfactory correlation may provide new and significant evidence for the possible applicability of the Danckwerts type of surface-renewal theory to the mass transfer from a flat solid surface into a turbulent liquid in an agitated vessel.

Nonuniformity and instability of local mass transfer coefficient is one of the disadvantages for the use of a packed tower for the study of the theory of mass transfer rates. Furthermore, a local mass transfer coefficient cannot be detected in a packed tower. In this study the local mass transfer coefficient was quantitatively detected with respect to the position of a cast solid; however, insertion of four baffles into the tank gave a uniform mass transfer coefficient regardless of the position of a cast solid. Since the present system can be considered to be a model of a liquid-liquid system, it is concluded that, in order to obtain the uniform mass transfer rate across the liquid-liquid interface in an agitated tank, baffles should be applied.

The advantages of the constant and stationary interface, the stability and simplicity of the apparatus, the possibility of duplicating experimental results, and the success of the detection of the local mass transfer coefficient suggest the use of the present apparatus and procedures for a further study of the mass transfer theory. In particular, a study of the mass transfer rates accompanied by chemical reaction is recommended. The surface-renewal rates obtained in the present work could be compared with the rate obtained for the case accompanied by chemical reaction as a further confirmation of the surface-renewal theory.

NOTATION

A = interfacial area, sq. cm.
C = concentration of solution, g. equivalent/liter or g. equivalent/cc.
C^* = saturated concentration, g. equivalent/liter or g. equivalent/cc.
C_i = interfacial concentration, g. equivalent/liter or g. equivalent/cc.
D = molecular diffusivity, sq. cm./sec.
K = constant
N = rate of mass transfer, g. equivalent /sec.
R = stirring speed, rev./sec. or rev. min.
T = temperature, °C.
V = volume of solution, cc. or liters
a = volume of a sample, cc.
b = exponent of Schmidt number
c = exponent of Reynolds number
d = diameter of impeller
k = mass transfer coefficient, cm./sec.
k' = apparent mass transfer coefficient, defined by Equation (12), cm./sec.
k° = mass transfer coefficient, no interfacial resistance assumed, cm./sec.
l = diameter of the mixing tank, cm.
p = number of sampling
r = constant = $(V - a)/V$
r_1 = inside radius of ring depression for casting solid, cm.
r_2 = outside radius of ring depression for casting solid, cm.
r_m = mean radius of ring depression for casting solid, cm.
s = fractional rate of surface renewal, $(\text{sec.})^{-1}$
t = time, sec.
v = exponent
x = fictitious film thickness, cm.
α = constant
β = constant
ϕ = correction factor defined by Equation (13), cm./sec.
ϕ_D = function of Schmidt number and friction factor, f, proposed by Lin, Moulton, and Putnam (*21*)
ρ = density, g./cc.
μ = viscosity, centipoises or g./(cm.)(sec.)

Subscripts

m = integer
n_1 = an arbitrary integer

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