

Dissolution Rates of Solids in Mercury and Aqueous Liquids: The Development of a New Type of Rotating Dissolution Cell

J. A. R. BENNETT and J. B. LEWIS, Atomic Energy Research Establishment, Harwell, near Didcot, Berkshire, England

A new type of corrosion problem arises when liquid metals, for example, sodium or mercury, are used as heat transfer fluids, since the solubilities of some structural metals in the liquid metals, although extremely small, are often markedly temperature dependent. This may cause a progressive dissolution of metal from the pipe wall in the hot region of the circuit with subsequent crystallization in the colder regions. Although this type of corrosion problem is being studied in many parts of the world, most of the results hitherto published are of an empirical nature; in particular there is little information about whether the rate controlling step during dissolution is a transport or chemical process.

In the present investigation the rate-controlling factors during the dissolution of lead, tin, and zinc in mercury were examined. The dissolution of specimens of these metals in mercury was compared with the dissolution of benzoic acid in water and, in order to extend the range of values of Schmidt groups, in aqueous sucrose solution. It was found that, apart from the dissolution of zinc in mercury, all the processes were transport controlled, the rate factors being correlated by a single hydrodynamic equation involving the Reynolds numbers and Schmidt groups. A chemical rate factor entered into the dissolution of zinc in mercury; the significance of this is discussed elsewhere (1).

To ensure reproducible and uniform hydrodynamic conditions during dissolution a large number of possible stirring conditions were first examined. In the system selected a stationary cylindrical specimen was mounted along the axis of a rotating vertical cylindrical vessel which contained the solvent. This technique, which as far as the authors know has not been used before, avoided the formation of Taylorian vortices at the surface of the specimen which otherwise caused a marked unevenness during the dissolution of the appreciably soluble metals. Since the reverse procedure, rotation of a cylindrical specimen in a stationary vessel, has been used by a number of previous investigators, some experiments were also carried out with the dissolution of benzoic acid in water and sucrose solution in the same apparatus with the vessel stationary and the speci-

men rotating. A different correlation between the dissolution rate and the Reynolds numbers and Schmidt groups was then obtained, which has been compared with the correlations reported by other workers for this type of stirring mechanism.

MECHANISM OF TRANSPORT PROCESSES

As is well known, the turbulence present in the bulk of an agitated liquid is suppressed in the vicinity of any solid surface. There is at present, however, some difference of opinion as to the exact nature of the flow pattern in the zone immediately adjacent to the surface. The earlier and more conventional theories assume that a laminar sublayer is always present in which the flow is parallel to the surface (22). Transport of solute molecules across this region will thus take place by molecular diffusion, which is a much slower process than the eddy or turbulent diffusion operative in the bulk phase. Between the sublayer and the bulk phase there exists a transition region (buffer layer) where turbulence is diminished and both molecular and eddy diffusion are important. The major resistance to transport between the

surface of the solid and the bulk phase, therefore, lies in molecular diffusion across the sublayer. It is convenient in practice to assume that the whole resistance to transfer is due to molecular diffusion alone, the small contribution owing to eddy diffusion being allowed for by taking the diffusional path length to be a fictitious film of a finite thickness, somewhat thicker than the sublayer. The thickness of the film is thus proportional to that of the sublayer, both being dependent on the degree of turbulence in the bulk phase.

Gas absorption and liquid-liquid extraction rates are also usually explained by the concept of the existence of a laminar sublayer at the interface. Alternative theories concerning gas absorption have, however, been proposed by Higbie (10), Kishinevskii (14), and Danckwerts (4) who suggest that the actual surface is constantly being renewed, with turbulent eddies reaching the gas-liquid interface. Kishinevskii further considers that no part at all is played by molecular diffusion. Lewis (15) has reached a similar conclusion, on experimental grounds, for flow at liquid-liquid interfaces. Recently concentration-profile measurements at solid

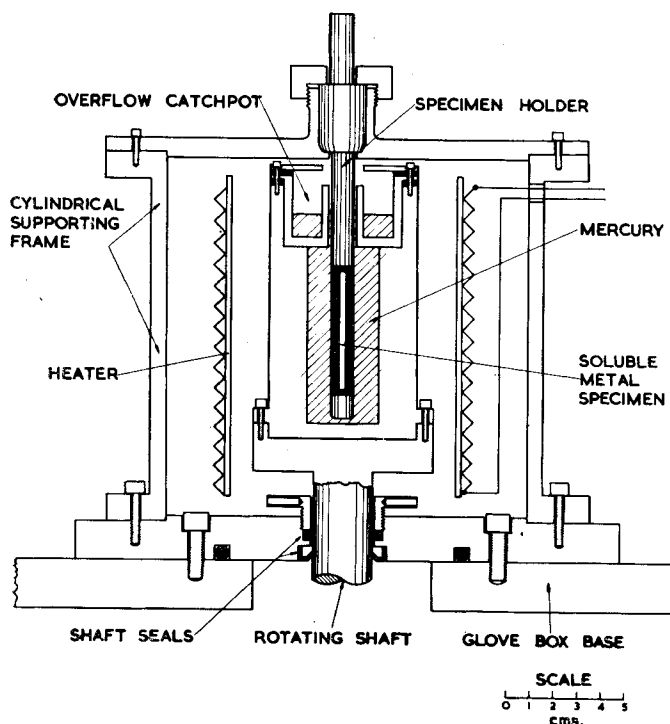


Fig. 1. Rotating cell.

J. A. R. Bennett is at present with Esso Petroleum Company, Fawley, Hampshire, England.

TABLE 1. PHYSICAL DATA

Solute	Solvent	Temp., °C.	Density, g./cc.	Viscosity, centipoises	Solubility	Diffusivity, sq. cm./sec. $\times 10^5$	$\left(\frac{\nu}{D}\right)$
Tin	Mercury	30	13.52	1.49	0.85*	1.60	69
Tin	Mercury	40	13.50	1.44	1.05*	1.50	71
Lead	Mercury	30	13.52	1.49	1.82*	1.25	88
Lead	Mercury	40	13.50	1.44	2.25*	1.07	100
Zinc	Mercury	30	13.52	1.49	2.39*	2.35	47
Zinc	Mercury	40	13.50	1.44	2.86*	2.30	47
Benzoic acid	Water	30	0.996	0.80	4.2†	1.14	708
Benzoic acid	Water	40	0.992	0.66	6.0†	1.43	462
Benzoic acid	Sucrose solution	30	1.18	5.79	3.58†	0.16	31,200
Benzoic acid	Sucrose solution	40	1.19	3.84	4.60†	0.24	13,200

*Solubility expressed as wt. %.
†Solubility expressed as g./liter.

electrodes (16) have been interpreted to indicate that turbulent eddies may reach solid surfaces also (9), and attempts have been made to apply the surface-renewal hypotheses to the description of the flow of mass and heat from solids to liquids (12).

Danckwerts and Kennedy (5) have shown that the laminar sublayer concept yields equations similar to the surface renewal theories. Thus the conventional transport equation can be used irrespective of the validity of the sublayer hypothesis, and the driving force for the transport process will be $(c^* - c)$ whether or not the flow is by molecular or eddy diffusion.

For the rate of transport

$$N = k_d A (c_i^* - c_i) \quad (1)$$

The proportionality constant k_d will be directly proportional to the molecular diffusivity, if the simple sublayer concept is correct but proportional to some lower fractional power in other cases. Under steady state conditions the transport rate will equal the net rate of transfer of solute from the solid specimen to the liquid immediately adjacent to it. This rate can be expressed in a similar manner (18):

$$N = k_c A (\gamma_{sat} c_{sat} - \gamma_i c_i^*) \quad (2)$$

(where γ_i is a function of c_i and not t)

Since the surface concentration c_i^* is generally not known, it is necessary for practical purposes to define an overall mass transfer coefficient, which can be experimentally measured; thus

$$N = K A (c_{sat} - c_i) \quad (3)$$

If k_c is much greater than k_d , one can see that c_i^* becomes equal to c_{sat} and K equal to k_d . The observed transfer coefficient will then have a maximum dependence on the degree of turbulence, that is the Reynolds number. An example of such a transport-controlled system is the dissolution of benzoic acid in water. If, therefore, the dissolution of metals in mercury follows the same general correlation as the dissolution of benzoic acid in water, it may be concluded that these dissolution processes are also diffusion controlled.

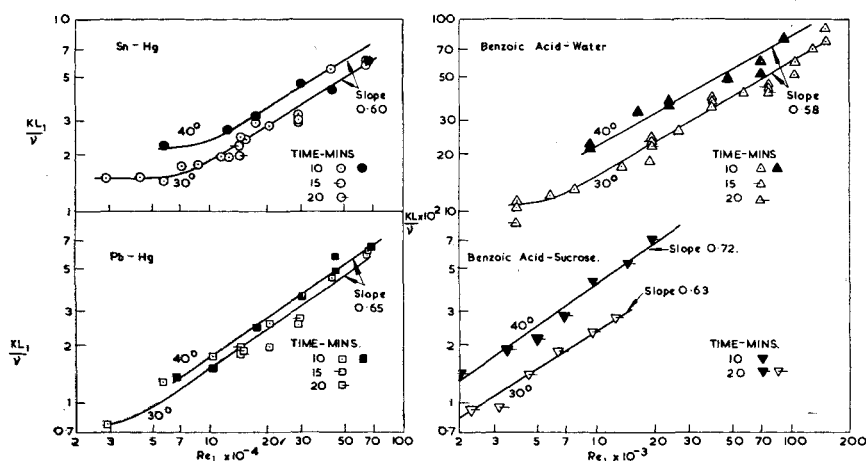
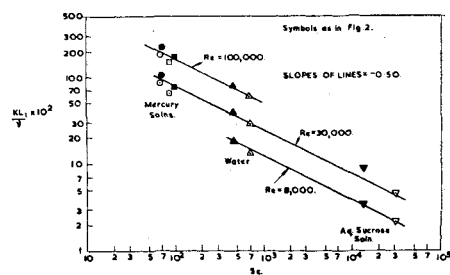
EXPERIMENTAL

Preliminary experiments were carried out with rods of benzoic acid and rods of lead, tin, and zinc, rotating in vessels containing water and mercury respectively. In all cases, even when extensive baffling was used, the surface of the specimens became "corrugated" during dissolution, apparently owing to the presence of Taylorian vortices at the surface (20). The least effect was obtained with the benzoic acid specimens, since the solubility of benzoic acid in water is very small.

When, however, the specimens were kept stationary down the axis of a rotating cylindrical vessel, smooth uniform dissolution was obtained except with zinc specimens, which were invariably etched. Although calculations have been made on the flow pattern between an infinitely long stationary cylinder and an outer rotating cylinder (19, 20), it was clear from the preliminary experiments that the disturbances set up at the top and bottom of the vessel so affected the ideal flow pattern that the hydrodynamics of flow would have to be examined semiempirically.

The metal specimens finally used consisted of tubes 5 cm. long and 1 cm. O.D., which were made from 99.99% pure metal of a very fine grain size. Each tube was threaded onto a stepped stainless-steel holder and held in place by an end cap 1 cm. long, the whole assembly forming a smooth cylinder of uniform external diameter. The specimen holder was accurately positioned along the vertical axis of a cylindrical mild-steel dissolution vessel, 7.3 cm. high and 3 cm. I.D., so that the actual specimen was located in the middle of the vessel. This arrangement minimized the effect on the specimen of the irregular flow patterns at the top and bottom of the vessel. To avoid vortex formation the free surface of mercury was reduced to a minimum by closing the top of the cell with a ring shaped plug which left a clearance of only 1 mm. around the specimen holder. It was found that the volume of mercury remained constant at 49.0 ± 0.1 cc., irrespective of the rotational speed. The vessel was mounted on a vertical shaft would could be rotated at speeds up to 4,000 r.p.m. by means of an electronically controlled motor, the exact speed being measured accurately by a stroboscope. A fixed electrical heater mounted around the vessel enabled the temperature of the mercury to be controlled to $\pm 0.2^\circ\text{C}$. Experiments were carried out at 30° and 40°C . Since the mercury solutions oxidized exceedingly rapidly in air and produced a scum which tended to contaminate the surfaces of the specimens and reduce the bulk concentration, the dissolution vessel was mounted inside a glove box filled with very pure argon. The rotating shaft passed through gas tight seals in the base of the box. A diagrammatic sketch of the transfer cell is shown in Figure 1.

A similar apparatus constructed entirely of stainless steel and mounted in an air thermostat was used for the benzoic acid dissolution experiments. The benzoic acid specimens were prepared by casting molten benzoic acid onto a 1-cm.-diameter metal rod which was recessed from 1 to 6 cm.

Fig. 2. Plot of KL_1/ν vs. Reynolds number—Rotating Cell.Fig. 3. Plot of KL_1/ν vs. Schmidt group—Rotating Cell.

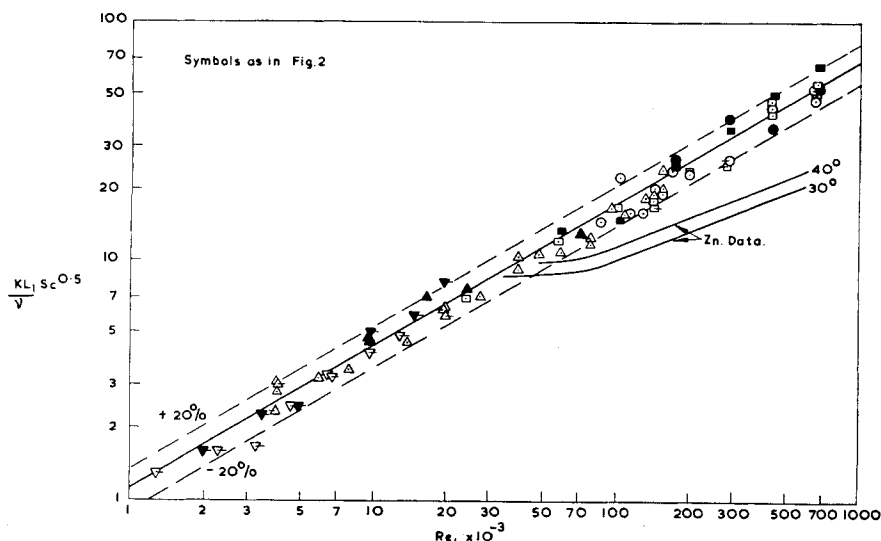


Fig. 4. Plot of $KL_1/\nu \cdot Sc^{1/2}$ vs. Reynolds number—Rotating Cell.

from the end. Surplus acid was machined away leaving a smooth specimen 5 cm. long and integral with the rod. The whole assembly was geometrically identical to the mercury apparatus, but the volume of water was slightly less, being 47.0 cc. Aqueous sucrose solution of concentration 500 g./liter was used.

Owing to the nature of the apparatus it was only possible to take samples of the liquid at the end of each experiment. Normally these were of 10 min. durations, but with the metal experiments some experiments were carried out for 15 and 20 min. to see if the transfer coefficients varied with time and therefore concentration. The lead and tin solutions in mercury were analyzed by evaporating off the mercury from a sample of known weight by heating to 300°C. in a current of nitrogen. This method proved to be quantitatively accurate. A similar technique was adopted with zinc solutions, but as the solid residue invariably contained about 9% by weight of mercury this was dissolved in aqua regia and analyzed polarographically. The benzoic acid solutions were titrated against N/100 sodium hydroxide solutions.

The solubilities of tin and zinc in mercury and benzoic acid in sucrose solution were experimentally determined at 30° and 40°C. All other solubility data were taken from the literature.

The viscosities and densities of all solutions were taken to be the same as those of the solvents, since the concentrations obtained during the experiments were low. No data were available for the diffusion coefficients of the various metals in mercury at 30° and 40°, but some data were available at 25° and lower temperatures. These were plotted as the logarithm of the diffusion coefficient against the reciprocal temperature, which is normally linear, and the best extrapolated values of the diffusivities obtained by inspection. Diffusion coefficients at 30° and 40°C. for benzoic acid in water and sucrose solution were calculated by using the Stokes-Einstein equation and Wilke's (7) data at 25°C. for the diffusion coefficient of benzoic acid in water. All relevant physical data are summarized in Table I

RESULTS

The multicrystalline zinc specimens gave an etched surface on dissolution, apparently owing to the individual crystal grains dissolving at different rates. This suggested that a chemical rate factor entered into the dissolution process and varied with the nature of the crystal face exposed. Confirmatory evidence was obtained from the dissolution of cylindrical specimens consisting of single crystals when it was found that the various crystallographic faces did indeed dissolve at markedly different rates. Similar effects are known to occur during the dissolution of zinc in various aqueous acid solutions; therefore the phenomenon is not peculiar to the dissolution of zinc in mercury. The chemical rate factors have been evaluated for the dissolution of zinc, and the results are given in detail elsewhere (1). Since zinc dissolution is thus not an entirely transport controlled process, the zinc data cannot be correlated along with those for other systems, as is shown later.

The rates of mass transfer were obtained from the final concentration of the mercury solution; thus since

$$N = \frac{Vdc}{dt} t \quad (4)$$

$$\therefore Kdt = -\frac{V}{A} \cdot d \ln (c_{sat} - c_i) \quad (5)$$

and hence K can be obtained by integration. Since samples could only be taken at the end of each experiment, the calculated values of K were mean values over the concentration range $c = 0$ to $c = c_i$. There was, however, no significant difference between values of K obtained after 10 to 20 min.*

*Tabular material has been deposited as document 5768 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for \$1.25 for photoprints or \$1.25 for 35-mm. microfilm.

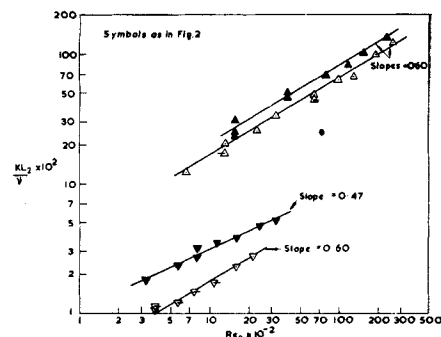


Fig. 5. Plot of KL_2/ν vs. Reynolds number—Rotating Specimen.

A fundamental theoretical approach is rarely possible when considering flow patterns in agitated systems owing to the complexity of the mathematics involved. The data are therefore usually correlated by the use of dimensional analyses, when one assumes that the mass transfer coefficient in a transport-controlled system is a function of the relevant parameters affecting the hydrodynamics of the system. With the rotating vessel experiments these were taken to be the viscosity, density, and solute diffusivity. The same parameters were used with the rotating specimens experiments except that the characteristic velocity and density were different. The diameter was not chosen in accordance with Eisenberg, Tobias, and Wilke (7). Hence one can write

$$k_d = f(\mu, \rho, D, u, L) \quad (6)$$

Three dimensionless groups can be formed from these six variables, and these can be conveniently arranged as follows:

$$k_d L/\nu = f[(Lu/\nu), (\nu/D)] \quad (7)$$

where the kinematic viscosity replaces μ/ρ , and the two groups on the right hand side are the Reynolds number and Schmidt group, respectively. In most instances Equation (7) can be approximated by a product of power functions, that is

$$k_d L/\nu = \text{const. } N_{Re}^a \cdot N_{Sc}^b \quad (8)$$

This expression may also be written in the following alternative forms, the groups on the left hand side being comparable with the Stanton and Nusselt heat transfer numbers, respectively

$$k_d/u = \text{const. } N_{Re}^{a-1} \cdot N_{Sc}^b \quad (8a)$$

$$k_d L/D = \text{const. } N_{Re}^a \cdot N_{Sc}^{b+1} \quad (8b)$$

When Equation (8) was applied to the correlation of the present data, it was necessary to investigate the variations of the experimentally determined over-all transfer coefficients with the Reynolds number and Schmidt group.

INTERPRETATION OF RESULTS

Rotating-Vessel Data

The over-all coefficients in the form

KL_1/ν were plotted against Reynolds number N_{Re} on logarithmic coordinates for each system studied (see Figure 2). If k_d and K are equal, linear plots of slope 'a' should be obtained, since the Schmidt group is a constant in any one system. Linear plots were obtained with slopes in the range 0.60 ± 0.05 for tin and lead in mercury, benzoic acid in water, and benzoic acid in sucrose solution at 30°C.; a higher exponent 0.72 was found for benzoic acid in sucrose solution at 40°C. When the zinc data were plotted in this manner, linear plots with slopes of 0.45 were obtained, the lower slope being due to the existence of a significant chemical step. The graphs flattened out at low values of the Reynolds number, probably owing to the proportionally greater contribution from convection currents at low stirring speeds. Because of the experimental scatter of the results a mean exponent of 0.60 on the Reynolds exponent has been adopted.

The Schmidt group exponents 'b' were similarly obtained by plotting KL_1/ν against the Schmidt group on logarithmic co-ordinates with the Reynolds number used as a parameter (see Figure 3). The data were well correlated by straight lines of slope -0.50 which extended over three orders of magnitude of the Schmidt groups. All the experimental data were therefore plotted in the form $(KL_1/\nu) N_{Sc}^{0.5}$ against N_{Re} . A reasonably good correlation with an over-all scatter of about $\pm 20\%$ was obtained (Figure 4). The equation to the solid line is

$$(KL_1/\nu) \cdot N_{Sc}^{0.5} = 0.018 N_{Re}^{0.6} \quad \text{i.e.} \quad (9)$$

$$(KL_1/\nu) = 0.018 N_{Re}^{0.6} \cdot N_{Sc}^{-0.5}$$

The zinc-mercury data were also plotted in this manner, and smoothed curves representing the results are included in Figure 4. These curves lie below the general correlation and also, unlike the other data, are temperature dependent. The benzoic acid-sucrose data fit the correlation to a fair degree of approximation, although, since at 40°C. the Reynolds exponent is greater than

0.60 with this system, the 40° points lie across the correlation at a shallow angle.

Rotating-Specimen Data

The data for the dissolution of rotating specimens of benzoic acid in water and aqueous sucrose solution were treated in the same manner as the rotating-cell data. A linear logarithmic plot was obtained between KL_2/ν and the Reynolds number N_{Re} in all cases (see Figure 5). The slopes of the lines were again 0.60 for water at 30° and 40° and for sucrose solution at 30°C.; with aqueous sucrose at 40°C. the slope was 0.45. In Figure 6 a logarithmic plot is given of (KL_2/ν) against the Schmidt group, and the data are best correlated by lines of slope -0.6.

The data are replotted in Figure 7, and, with the exception of the sucrose solution at 40°C. which again crosses the main correlation at a shallow angle, are correlated to within $\pm 20\%$:

$$(KL_2/\nu) N_{Sc}^{0.6} = 0.135 N_{Re}^{0.6} \quad \text{i.e.} \quad (10)$$

$$(KL_2/\nu) = 0.135 N_{Re}^{0.6} N_{Sc}^{-0.6}$$

DISCUSSION

The correlation of the tin-mercury and lead-mercury data along with the benzoic acid-water data is conclusive evidence that the rates of dissolution of these metals are also transport controlled over the range of turbulence investigated. The 40° benzoic acid-sucrose data tends to diverge from the correlation because the exponent on the Reynolds number is different, probably since the use of a simple power function is a good approximation only over the range of Schmidt groups obtained with mercury and water, that is 47 to 708. The extension of this expression to the much higher Schmidt groups obtained with sucrose solution, 13,200 and 31,200 at the two different temperatures, is much less valid. It is not clear however why the sucrose data at 30° fit the correlation better than those

obtained at 40°C. The divergence of the zinc data arises from the presence of a slow chemical step at the surface of the crystal lattice during dissolution. These results have been discussed in detail elsewhere, and values have been derived for the chemical rate constant.

It is interesting to note that the same exponent of the Reynolds number was obtained for both the rotating vessel and the rotating specimen experiments. The 0.6 power falls in the 0.5 to 1.0 range of Reynolds exponents previously reported by various authors for a variety of flow condition [Vielstich (21) and Bircumshaw and Riddiford (2)]. Eisenberg, Tobias, and Wilke have recently carried out a comparable rotating specimen experiment (7) and obtained a correlation which, when expressed in the form of Equation (8a), gives a Reynolds exponent of 0.7; however the use of a simple power function relation to cover the whole range of experimental results is of limited validity. Over the range of Reynolds numbers equivalent to those used in this paper the Reynolds exponent is somewhat lower.

The Schmidt group exponent is of considerable theoretical interest as it indicates the relative importance of molecular and turbulent diffusion. The well-known Chilton-Colburn equation (3) derived from the relationship between heat transfer and fluid friction gives an exponent of $2/3$ on the Schmidt group. This can be written

$$(K/u) = f(N_{Re}) \cdot N_{Sc}^{-2/3} \quad (11)$$

Linton and Sherwood (17) have shown that this expression can be used to correlate their experimental results for mass transfer in pipe flow. Eisenberg, Tobias, and Wilke (7) reported a slightly lower exponent 0.644 on the Schmidt group for rotating specimen experiments; other authors however have found values of the Schmidt exponent significantly greater or smaller than two thirds. King and Howard (13) measured the mass transfer coefficients for the dissolution of thin metal wires in various liquid reagents by using stationary specimens mounted

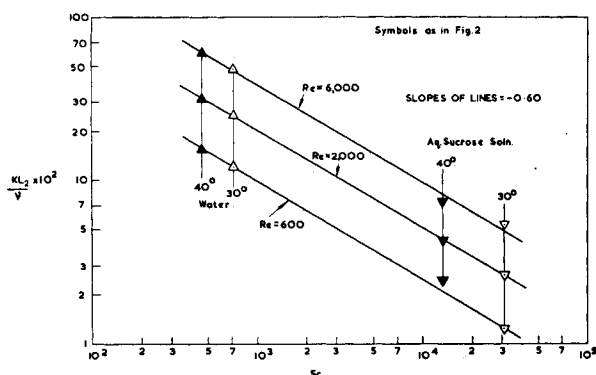


Fig. 6. Plot of KL_2/ν vs. Schmidt group—Rotating Specimen.

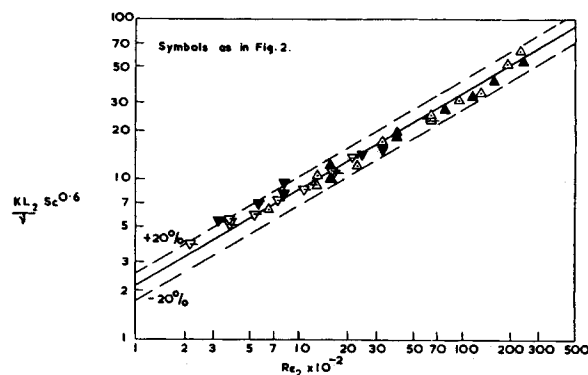


Fig. 7. Plot of $KL_2/\nu \cdot Sc^{0.6}$ vs. Reynolds number—Rotating Specimen.

inside a rotating cylindrical stirrer and the rate of heat transfer from electrically heated platinum wires to aqueous sucrose solutions. Exponents of about 0.70 were found on both the Schmidt groups and Prandtl numbers. These results cannot be compared directly with stirred vessel experiments discussed in this paper since the cylindrical stirrer was grooved, open at both ends, and immersed in a large vessel of liquid. Longitudinal flow past the specimen would thus take place, the flow pattern being further complicated by vortex formation at the free surface of the liquid. A different type of turbulent flow existed in the stirred vessel, which approximated that investigated by Taylor (20). Higher Schmidt exponents, up to 0.83, have been obtained by King (13) in earlier experiments with rotating cylindrical metal specimens.

On the other other hand Davion (6) found an exponent of 0.63 for the dissolution of single crystals of salts in water, while Gaffney and Drew (8) reported that the best exponent on the Schmidt group for the dissolution of organic acid pellets in a packed extraction column was 0.58. Hixson and Baum (11) and Johnson and Huang (12) reported values of 0.50 for mass transfer in paddle-stirred systems. The latter authors suggest that this low value indicates that the Danckwerts surface-renewal theory can be applied to solid-liquid dissolution as well as to gas-liquid absorption. Two different exponents on the Schmidt group were obtained in the experiments conducted by the authors of this paper, 0.50 with the cell rotating and 0.60 with the specimen rotating.

Since values of the Schmidt exponent ranging from 0.50 to 0.82 have been reported by various authors, it is clear that the Danckwerts theory cannot generally be applied in its original and simplest form which would give a 0.50 exponent only. If it is assumed that solutes can penetrate through the laminar sublayer before the sublayer is destroyed by an eddy, then exponents greater than 0.50 can be postulated. The exponents should be greater the greater the local turbulence in the apparatus used. This is indeed the case in the authors' experiments, 0.60 with the actual specimen rotating as compared with 0.50 when the cell rotated. Since values of the Schmidt exponents in the range reported can also be postulated by modifications of the conventional boundary layer theory (2, 21), these results do not themselves confirm or disprove the surface renewal hypothesis. It is clear that the geometry and stirring conditions used by the various workers have a direct bearing on the value of the Schmidt exponent obtained by them. More data, especially on the exact nature of the turbulence present at the surfaces of the various dissolving specimens, are needed before the theory can be rigorously proved.

CONCLUSIONS

1. A comparison between the dissolution of lead and tin in mercury with the dissolution of benzoic acid in water, which is known to be transport controlled, indicated that the metal dissolution rates are also transport controlled.

2. The dissolution rate of zinc in mercury is lower than would be anticipated from the data for the other metals, and this has been shown to be owing to a slow chemical step at the surface of the crystal lattice.

3. Taylorian vortices are inevitably obtained at the surfaces of rotating specimens. They can be prevented only by circulating the solvent around the specimen, as for example by rotating the containing vessel. Data for the dissolution of stationary rods under these conditions of stirring are correlated to $\pm 20\%$, over a range of Reynolds numbers from 1,000 to 440,000 and a range of Schmidt groups from 47 to 700, by the relation

$$(KL_1/\nu) = 0.018(L_1u_1/\nu)^{0.60}(\nu/D)^{-0.50}$$

This correlation can be extended to Schmidt groups of 31,000, that is dissolution of benzoic acid in aqueous sucrose solution, with a slight loss of accuracy.

4. Data for the dissolution of rotating specimens of benzoic acid in water are correlated by the following expression which differs somewhat from that of Wilke:

$$(KL_2/\nu) = 0.135(L_2u_2/\nu)^{0.60}(\nu/D)^{-0.60}$$

5. There is insufficient evidence to confirm or disprove the validity of the surface renewal theory for the dissolution of solids in liquids.

NOTATION

- A = total surface area of specimen, sq. cm.
 c = concentration, wt. % for mercury solutions; g./litre for aqueous solutions
 D = diffusivity, sq. cm./sec.
 f = function of
 k = individual mass transfer coefficient, cm./sec.
 K = observed 'over-all' mass transfer coefficient, cm./sec.
 L_1 = characteristic dimension of rotating cell hydraulic diameter of the annulus, cm.
 L_2 = characteristic dimension of rotating specimen diameter, cm.
 N = rate of mass transfer, g./sec.
 t = time of dissolution, sec.
 u_1 = characteristic velocity of rotating cell peripheral velocity of the inner surface cm./sec.
 u_2 = characteristic velocity of rotating specimen peripheral velocity of the specimen, cm./sec.
 V = volume of solvent, cc.

- γ = activity coefficient
 μ = viscosity of solvent, g./sec.(cm.)
 ν = kinematic viscosity of solvent, sq. cm./sec.
 ρ = density of solvent—g./cc.
 N_{Re} = Reynolds number, Lu/ν
 N_{Sc} = Schmidt number, ν/D

Subscripts

- c = chemical rate
 d = diffusional rate
 sat. = at saturation
 t = value at time t

Superscript

- * = value at solid-liquid interface

LITERATURE CITED

- Bennett, J. A. R., and J. B. Lewis, *J. Chim. phys.*, p. 83 (1958).
- Bircumshaw, I. L., and A. C. Riddiford, *Quart. Rev.*, **6**, 157 (1952).
- Chilton, T. H., and A. P. Colburn, *Ind. Eng. Chem.*, **26**, 1183 (1934).
- Danckwerts, P. V., *ibid.*, **43**, 1460 (1951).
- Danckwerts, P. V., and A. M. Kennedy, *Trans. Inst. Chem. Engrs.*, **32**, S49 (1954).
- Davion, Maurice, *Ann. Chim. (Paris)*, **8**, 259 (1953).
- Eisenberg, M., C. W. Tobias and C. R. Wilke, *Chem. Eng. Progr. Symposium Series*, No. 16, **51**, 1 (1955).
- Gaffney, B. J., and T. B. Drew, *Ind. Eng. Chem.*, **42**, 1120 (1950).
- Hanratty, T. J., *A.I.Ch.E. Journal*, **2**, 359 (1956).
- Higbie, Ralph, *Trans. Am. Inst. Chem. Engrs.*, **31**, 365 (1935).
- Hixson, A. W., and S. J. Baum, *Ind. Eng. Chem.*, **33**, 478, 1433 (1941).
- Johnson, A. I., and Cheng-Jung Huang, *A.I.Ch.E. Journal*, **2**, 412 (1956).
- King, C. V., and P. L. Howard, *Ind. Eng. Chem.*, **29**, 75 (1937).
- Kishinevskii, M. Kh., and A. V. Pamfilov, *Zhur. Priklad. Khim.*, **22**, 1173 (1949).
- Lewis, J. B., *Chem. Eng. Sci.*, **3**, 248 (1954).
- Lin, C. S., R. W. Moulton, and G. L. Putnam, *Ind. Eng. Chem.*, **45**, 636, 640 (1953).
- Linton, W. H., and T. K. Sherwood, *Chem. Eng. Progr.*, **46**, 258 (1950).
- Moelwyn-Hughes, E. A., "Kinetics of Reactions in Solution," p. 375, Oxford University Press (1947).
- Schlichting, Hermann, "The Boundary Layer Theory," Pergamon Press, London (1955).
- Taylor, G. I., *Trans. Roy. Soc. (London)*, **223**, 289 (1923); *Proc. Roy. Soc. (London)*, **A.151**, 494 (1935).
- Vielstich, Wolf, *Z. Elektrochem.*, **57**, 646 (1953).
- Whitman, W. G., *Chem. Met. Eng.*, **29** (4), 146 (1923); Lewis, W. K., and W. G. Whitman, *Ind. Eng. Chem.*, **16**, 1215 (1924).

Manuscript received January 7, 1958; revision received April 21, 1958; manuscript accepted April 22, 1958.