Mass Transfer in Liquid Metals

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The rate of mass transfer was measured for solid metal shapes dissolving into mercury at room temperature. Sherwood numbers for horizontal tin, cadmium, zinc, and lead cylinders dissolving by natural convection agreed with Nusselt numbers for heat transfer in nonmetallic liquids at the same Rayleigh (Grashof × Prandtl) numbers. Dissolving of zinc tubes by mercury flowing turbulently within them agreed with heat transfer to nonmetals in tubes. Dissolving of random beds of lead spheres by mercury flowing through the bed agreed with similar nonmetal systems. It is concluded that mass transfer processes in liquid metals follow substantially the correlations for other fluids in heat or mass transfer, which with moderate safety factors may thus be used for at least preliminary design purposes.

Cases of mass transfer in liquid metals are becoming more frequent with the increasing popularity of liquid metals as heat transfer media and as solvents. The pyrometallurgical refining of metals has long employed mass transfer between the melted metal and a molten flux, another molten metal, a solid, or a gas. Corrosion of pipes and vessels by contained liquid metals due to temperature differentials plus a variation of solubility with temperature has long been encountered. The purification of molten sodium by the freezing out of sodium oxide is a similar case. Finally, liquid-metal fuel reactors (L.M.F.R.) have been proposed (*20*) which feature mass transfer to accomplish principal objectives. Replenishment of the fissionable element would involve mass transfer, which, however, would be applied principally in the continuous removal of radioactive and neutron-consuming fission products.

In the design of such processes, or in predicting the effect on performance of changes in operating conditions, it is desirable to have available the appropriate dimensionless correlations of mass transfer for each proposed geometry. A few such correlations from studies on nonmetallic liquids have been published. However, before one could confidently employ such correlations for liquid metals it would seem desirable to establish their validity for liquid metals in at least several key geometries. This paper presents the results of such a study employing four binary systems and three rigid geometries. The solids were lead, tin, cadmium, and zinc, and the liquid phase was mercury at room temperature. Natural convection was studied by dissolving horizontal cylinders of all four metals in a stationary pool of mercury, the Grashof × Schmidt number product ranging from 10^7 to 10^9. Forced convection was studied by passing mercury through zinc pipes at Reynolds numbers of 1,200 to 12,000 and through fixed beds of randomly packed lead spheres at void Reynolds numbers of 20 to 1,000. In all cases the Schmidt number was of the order of magnitude of 100.

The differential equations for forced-convection mass transfer with constant physical properties (dilute solutions) are identical with those for heat transfer, provided that the Nusselt and Prandtl numbers in heat transfer are replaced by the Sherwood and Schmidt numbers for mass transfer. It would thus be expected, and has empirically been shown (*1*) *for nonmetals*, that heat transfer correlations obtained for any given geometry are applicable to mass transfer in the same geometry at the same Reynolds numbers when the foregoing replacements are made. Similarly, natural-convection mass transfer would be expected to follow the heat transfer correlations on further replacement of the fractional density change ($\beta \Delta T$) in the Grashof number by the analogous ($\beta_c \Delta c$) for the dissolving of the solute. It is thus additionally desirable to check whether liquid-metal mass transfer obeys the available heat transfer correlations for the same geometries at the same values of the analogous dimensionless ratios. It should be noted that liquid-metal heat transfer results are not helpful in predicting liquid-metal mass transfer because the range of Prandtl numbers for liquid metals is several orders of magnitude smaller than the Schmidt numbers of liquid-metal systems.

PHYSICAL PROPERTIES OF THE SYSTEMS

For reliable correlations it is necessary to employ accurate values of the pertinent physical properties. Thus all the properties were employed as functions of concentration for each solute metal and were taken at the arithmetic-mean film concentration $\bar{c}_f = (c_i + \bar{c}_b)/2$. In the natural-convection tests $\bar{c}_b$ was taken as the calculated instantaneous average concentration of the solute in the bath. In the forced-convection tests the inlet bulk concentration c_1 of solute metal was always zero, and $\bar{c}_f$ was taken as $(c_i/2) + (c_2/4)$.

The density and the coefficient of solution expansion of the amalgams were obtained from Richards (*16*), the viscosity from Von Schweidler (*17*), and the solubility from Hansen (*9*). These properties are all substantially linear over the range of interest and may be linearly interpolated over the concentration and temperature ranges of Table 1. The diffusivities of these amalgams at low concentrations have been tabulated by Furman and Cooper (*5*). These were corrected to the higher concentrations employed by means of the Eyring (*7*) relation:

$$\frac{(D_r)}{(D_v)_0} = 1 + \frac{d(\ln \gamma)}{d(\ln N)} \tag{1}$$

The activity coefficient γ was obtained (*3*) as a function of atom fraction N from Richards' classic data (*16*) on the potentials of amalgam concentration cells. When the deviation between Richards' observed potential and that of an ideal cell, both having the pure solute metal as one electrode, is plotted against the logarithm of the other concentration, the slope multiplied by vF/RT equals $d(\ln \gamma)/d(\ln N)$ at the other concentration. The ratio $D_v/(D_v)_0$ so obtained is given in Figure 1. The results for zinc agree closely with Weischedel's experimental values (*18*), the only literature data that go to reasonably high concentrations in these systems.

NATURAL CONVECTION AT HORIZONTAL CYLINDERS

There are few, if any, mass transfer results in the literature for natural convection at horizontal cylinders. There

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TABLE 1. PHYSICAL PROPERTIES OF AMALGAM SYSTEMS STUDIED

Solute,	wt. %	Viscosity, μ lb./(ft.)(sec.) $\times$ 1,000		Density, ρ, g./cc.		Diffusivity, D_v, sq. ft./(sec.)(10^8)		Schmidt No. ($\mu/\rho\ D_v$)		Solubility, wt. %		
		20°	30°	20°	30°	20°	30°	20°	30°	20°	25°	30°
Zinc	0	1.06	1.02	13.545	13.542	1.745	1.850	71.8	65.3			
	0.5	1.094	1.056	13.50	13.497	1.630	1.725	79.6	72.7			
	1.0	1.128	1.090	13.46	13.457	1.510	1.602	88.9	81.1	2.00	2.18	2.40
	1.5	1.162	1.124	13.42	13.417	1.396	1.480	99.4	90.8			
Tin	0	1.06	1.02	13.545	13.542	1.755	1.860	71.4	66.0			
	0.2	1.068	1.029	13.528	13.525	1.602	1.699	78.9	71.8			
	0.4	1.077	1.038	13.516	13.513	1.485	1.574	86.0	78.3	0.64	0.71	0.78
	0.6	1.085	1.046	13.504	13.501	1.404	1.488	91.7	83.5			
Lead	0	1.06	1.02	13.545	13.542	1.212	1.296	103.4	93.3			
	0.5	1.082	1.038	13.541	13.538	1.067	1.142	120.0	107.7			
	1.0	1.100	1.057	13.536	13.533	0.909	0.972	143.2	128.9	1.53	1.67	1.84
	1.5	1.116	1.075	13.532	13.529	0.848	0.907	155.8	140.6			
Cadmium	0	1.06	1.02	13.545	13.542	1.575	1.720	79.6	70.3			
	1	1.088	1.049	13.487	13.484	1.605	1.753	80.5	71.2			
	2	1.117	1.077	13.427	13.424	1.630	1.780	81.8	72.3	5.55	6.20	6.85
	3	1.143	1.106	13.369	13.366	1.653	1.806	82.8	73.5			

are, however, some heat transfer results for sizable cylinders (*12*) and also heat and mass transfer results for natural convection at vertical surfaces, which can be altered to apply roughly to horizontal cylinders by Hermann's conclusion (*10*) for streamline natural convection that the average heat transfer coefficient around a cylinder of diameter D is the same as over a vertical surface of height $2.5D$. For these reasons, plus the mechanical convenience of this shape, horizontal cylinders about ½ in. O.D. were selected for the experimental study of natural convection.

Natural convection is usually associated with streamline flow, and the correlations that have been proposed are given in Table 2. However, from heat transfer results with the same-sized cylinders (*12*) turbulence might be expected to initiate at $(Gr_m \times Sc)$ = about 10^6 and to start to raise the Sherwood number rapidly at about 10^7 or slightly higher.

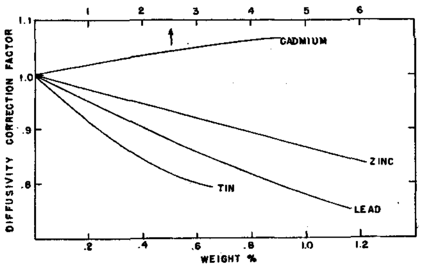

Fig. 1. Diffusivity correction factor for metals in mercury [Equation (1)].

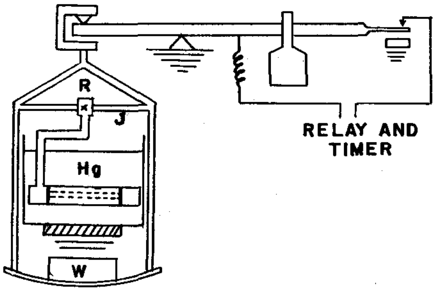

Fig. 2. Schematic diagram of apparatus for mass transfer from a horizontal cylinder by natural convection.

The equipment employed (*3*) is shown schematically in Figure 2. It consisted of a shock-mounted specific-gravity balance sensitive to 0.01 g. modified to support a wire frame. The frame by means of its ballast weight W held the specimen under the surface of the mercury and transmitted changes in the buoyant force to the balance arm. The arm was constrained by set screws to 1/64 in. of movement. An electronic relay indicated the instant that contact was made with the upper set screw.

A run was initiated by rotating the sample around J until it was immersed in the pool of mercury, tightening the set screw R, and setting the weight on the balance to a value slightly above the balancing point. When the sample had dissolved slightly, the buoyancy decreased, the balance arm rose, and, when the relay indicated contact, the time was recorded. The weight was immediately reset in successive small equal increments and the successive times were recorded. The rates of solution were then calculated over each time interval from the change of buoyancy. These rates decreased slowly and linearly with time t, and thus the initial rate of solution was obtained by extrapolation back to $t = C$, as shown in Figure 3*a*. Different driving forces were obtained by allowing solute metal to build up in the bath over a series of runs. After each series the mercury was completely purified by extended bubbling with air followed by sulfuric acid treatment to remove the gross impurity, with vacuum distillation for final purification.

The metal cylinders were prepared in two ways. Zinc and cadmium cylinders were machined down to size from slugs cast in glass. Tin and lead were extruded at 125°C. and upon cooling were cut off into sections. In all cases the ends were then faced off and an axial hole was drilled to hold the iron end pieces, rubber washers, test cylinder, and wire frame all together. All metals were C.P. or better quality. Castings of zinc were made under various cooling conditions to alter the grain size, but there did not appear to be a large variation in initial solution rate from this cause. However, for uniformity, all cast cylinders were allowed to cool slowly in the room.

TABLE 2. STREAMLINE NATURAL-CONVECTION MASS TRANSFER CORRELATIONS FOR HORIZONTAL CYLINDERS

Relation	Origin	Reference
$Sh = 0.53(Gr_m.Sc)^{1/4}$	Heat transfer tests at horizontal cylinders	*12, 13*
$Sh = 0.53[Gr_m.Sc^2/(0.952 + Sc)]^{1/4}$	Heat transfer tests at horizontal cylinders	*12*
$Sh = 0.418(Gr_m.Sc)^{1/4}$	Pohlhausen's theory for vertical plates, with Hermann's modification	*19*
$Sh = 0.525(Gr_m.Sc)^{1/4}$	Wilke's mass transfer tests at vertical plates with Hermann's modification	*19*
$Sh = 0.522(Gr_m.Sc)^{1/4}$	Ostrach's theory for vertical plates at Sc = 100, with Hermann's modification	*15*

Note: If the physical properties vary over the boundary layer, as a first approximation they should be taken at its arithmetic-mean concentration.

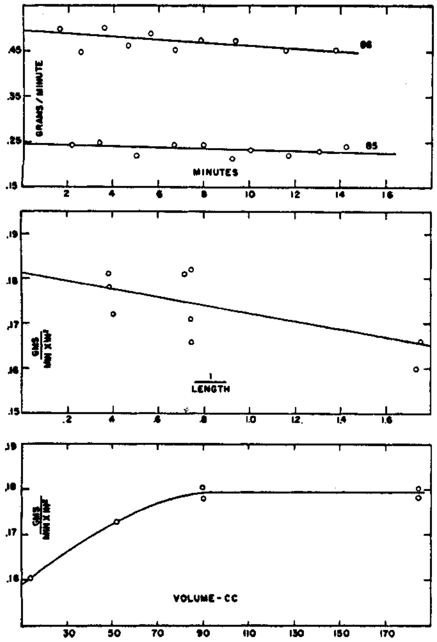

Fig. 3. Effect of time, sample length, and bath volume on the rate of buoyancy decrease of horizontal cylinders in still mercury.

Zinc chill-cast in steel molds, electroplated, or commercially remelted and extruded yielded significantly lower solution rates, owing apparently to traces of iron picked up or to zinc oxide occluded by the metal.

Samples were preamalgamated by being dipped into dilute sulfuric acid to remove the oxide layer and then into mercury. The time elapsed between preamalgamation and the start of the run was found not to affect the rate of solution.

End effects were determined for the most rapidly dissolving metal, cadmium, by varying the length of the samples over the range of ½ to 2½ in. The initial rate of solution was found to increase roughly linearly as $1/L$ decreased, as shown in Figure 3*b*. A 5% increase was obtained by extrapolating to $D/L = 0$ from a cylinder with $D/L = 1/8$. All results herein reported were accordingly corrected to the infinite cylinder values. This end-effect correction was assumed proportional to rate of solution. Thus for the more slowly dissolving systems the correction corresponding to cadmium at the desired L/D was decreased proportionally to the rate of solution of the metal, by weight.

The effect of ambient volume was studied with zinc cylinders. It was found that below 1 kg. of mercury the rate decreased (Figure 3*c*). The amount of mercury used in the runs here recorded was 29 kg., evidently equivalent to an infinite ambient.

One test was carried out to determine whether any appreciable temperature changes occurred at the dissolving surface owing to latent heat effects which might change the solubility c_i. A solid lead bar was bent and one end inserted in a bath of pure mercury and the other in mercury saturated with lead. A copper wire led from each bath to a potentiometer sensitive to 1 μv. The thermoelectric force of this system is roughly 5 μv./°C. There was never any detectable voltage, indicating negligible error due to dissolving.

Typical points are given in Table 3, and all the results are plotted in Figure 4 as the Sherwood number *Sh* vs. the Grashof number for mass transfer $Gr_m \times$ the Schmidt number *Sc*. It is seen that the results correlate well among themselves and agree nearly up to $Gr_m \times Sc = 10^9$ with the correlations of Table 2, which agree among themselves.

It should be noted that *Sh* agreed with the laminar correlations up to (at least) $Gr_m \times Sc$ values of almost 10^9, as contrasted with the observed rapid rise in *Nu* at $Gr_h \times Pr$ above 10^7, previously mentioned. However, visual inspection of the samples indicated that turbulence had been present at all but the lowest $Gr_m \times Sc$ values. All the zinc and cadmium specimens ($Gr_m \times Sc > 10^8$) showed a highly roughened surface after a test, probably in these cases an effect of crystal orientation. But all the tin and lead specimens—except the four tin specimens of lowest $Gr_m \times Sc$—showed a smooth lower surface but still displayed a roughened top surface (Figure 5*a*), probably because of turbulent eddies downstream of the cylinders. Figure 5*b* shows the smooth surface and "fish-tail" top edge, as might be expected in com-

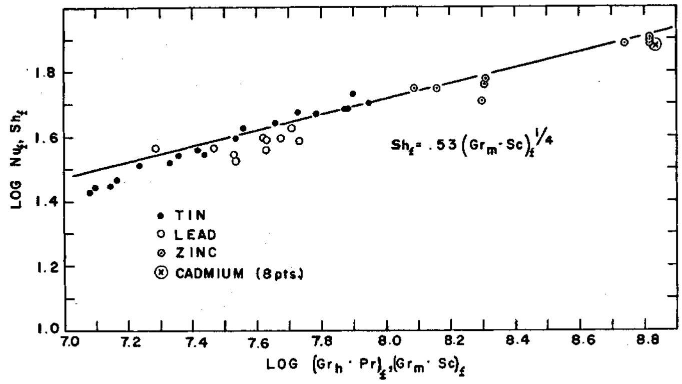

Fig. 4. Correlation of natural-convection mass transfer from horizontal cylinders.

TABLE 3. MASS TRANSFER BY NATURAL CONVECTION FOR HORIZONTAL METALLIC CYLINDERS IN MERCURY

Metal	Run	°C.	Initial diam. D, in.	L, in.	Initial rate, g./min.	Buoyancy factor, $\rho/\Delta\rho$	Initial bath conc., wt. %	Solubility, wt. %	End corr.	log Sh_f	log $(GrSc)_f$	Sc_f
Zn	59s	26.7	0.325	1.028	0.1321	1.113	0.000	2.27	1.032	1.464	8.817	80.4
	64	13.3	0.351	2.516	0.0275	1.113	0.957	1.74	1.003	1.709	8.297	97.9
	68	12.8	0.342	2.530	0.0189	1.113	1.236	1.73	1.002	1.748	8.159	101.9
Sn	40	14.5	0.211	1.512	0.0221	1.173	0.0084	0.560	1.003	1.540	7.356	90.5
	44	15.7	0.174	1.281	0.0213	1.173	0.0557	0.578	1.002	1.468	7.168	91.2
	48	15.0	0.350	2.519	0.0166	1.174	0.095	0.567	1.002	1.701	7.948	92.3
	52	14.8	0.293	2.119	0.0142	1.175	0.159	0.564	1.002	1.640	7.656	93.5
	56	15.9	0.350	2.485	0.0124	1.175	0.196	0.581	1.002	1.681	7.870	93.7
Pb	69	13.4	0.350	2.507	0.0581	5.135	0.000	1.290	1.007	1.625	7.709	138
	73	10.8	0.350	2.531	0.0396	5.137	0.1776	1.230	1.005	1.559	7.628	143
	77	11.7	0.350	2.525	0.0258	5.146	0.5390	1.246	1.003	1.563	7.468	149
Cd	80	12.8	0.350	2.523	0.181	1.767	0.000	4.62	1.023	1.882	8.844	83.5
	84	11.9	0.351	0.575	0.158	1.767	0.139	4.53	1.097	1.879	8.838	83.4
	88	13.3	0.348	1.386	0.175	1.767	0.200	4.68	1.039	1.882	8.832	82.3

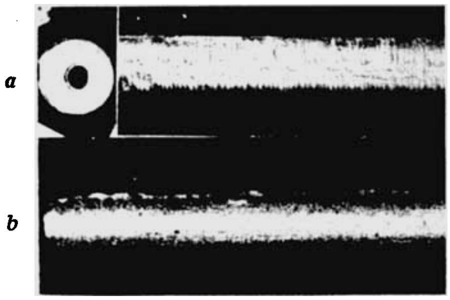

Fig. 5. (*a*) Appearance of top and cross section of horizontal tin cylinder after turbulent natural convection; (*b*) appearance of side of horizontal tin cylinder after streamline natural convection.

pletely streamline flow, which were obtained in the four lowest $Gr_m \times Sc$ tin samples*. These results thus indicate incidence of turbulence at $Gr_m \times Sc$ = 16,000,000 approximately, but no significant deviation from the streamline relation as far up as 650,000,000. There are thus qualitative agreement with the results so far reported for heat transfer to cylinders larger than wires and rough quantitative agreement on the point of incidence or turbulence. It is difficult to assess the significance of the disagreement on the threshold of turbulence and breakaway of the *Nu* and *Sh* values from the streamline relation. However, it seems established that laminar natural-convection correlations for any geometry and for either heat or mass transfer will hold for liquid-metal mass transfer in that geometry. It also seems probable, particularly in view of the turbulent forced-convection results to be discussed next, that agreement can also be expected, in general, between the corresponding turbulent natural-convection correlations.

FORCED-CONVECTION FLOW IN PIPES

For turbulent forced-convection flow through pipes, the classic Chilton-Colburn equation for heat transfer (*1*) when modified to mass transfer becomes

$$j_D = (k_D/u)(Sc)_f^{2/3} = 0.023(Re)_f^{-0.2} \qquad (2)$$

For the viscous region the Lévêque equation (*9*) when similarly modified becomes

$$j_D = 1.61(Re)_b^{-2/3}(L/D)^{-1/3} \qquad (3)$$

The only previous work on mass transfer inside of pipes that was found in the literature is that of Linton and Sherwood on the dissolving of cinnamic acid, benzoic acid, and β-naphthol tubes in water; *Sc* ranged from 1,000 to 3,000. Though the spread of the j_D values was considerable, the results agreed moderately well with Equations (2) and (3).

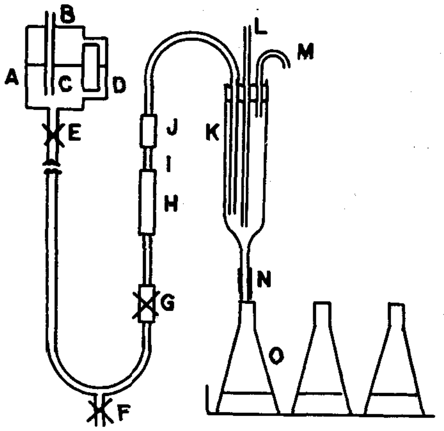

Fig. 6. Apparatus for mass transfer from a zinc tube to mercury.

Various studies on mass transfer wetted-wall columns have shown departures from Equation (2), but this might be expected owing to the motion of the liquid film. It is thus logical to compare liquid metal results in tubes with Equations (2) and (3). Zinc was used for the tubes in this study because it is stronger than the other convenient metals.

The zinc was approximately 99.995% pure, containing roughly equal amounts of bismuth, cadmium, copper, and lead as the principal impurities. After each run the mercury was placed in a suction flask under a layer of 50% sulfuric acid, and room air was bubbled through it from a submerged air-inlet tube by the application of vacuum on top. After an hour or so the acid was decanted and the mercury washed repeatedly with distilled water, trickled through a tall 15% nitric acid tower, washed again, dried in beakers at 120°C., and finally filtered slowly through a dry towel. A bright surface, indicating less than 0.0001% of base metal, was always obtained.

The test apparatus employed (*4*) is shown schematically in Figure 6.

Zinc rods were cast by melting the zinc in an iron crucible, sucking it up into preheated ½-in. Pyrex tubes, and then allowing it to freeze. The rods were cut into 2.1+ in. lengths, drilled through at their axis with an extralong 0.140-in. drill, turned at both ends to a concentric O.D. of 0.350 in., and faced off perpendicular to the axis to a length of 2.1 in. (15 diam.). These specimens (*I* in Figure 6) were forced into heavy-wall steel tubes *H* and *J* having a 0.140-in. bore and a flat-bottomed concentric cup 0.351 in. in I.D. at their nearer ends. Thus the steel-zinc-steel formed a continuous tube of smooth bore. The apparatus contained a closed 3.5-liter steel tank with an air inlet tube *B* to maintain the mercury flow constant during a run, a level gauge *D*, on-off cock *E*, drain pinch clamp *F*, flow-regulating screw clamp *G*, 50-diam.-inlet calming length *J*, glass receiver *K*, thermometer *L*, air vent *M*, and rubber pinch tube *N*. Before a run 30 ml. of 1 *M* hydrochloric acid (more than enough to dissolve the zinc) was added in advance to each of six to ten preweighed sample flasks *O* on a flask holder. A new specimen *I* was next filled and wetted internally with mercury to assure immediate action in the run. It was then inserted between *H* and *J* and the run started at once. Clamp *G* was preset as desired, *E* suddenly opened, and a stop watch started when mercury appeared at *N*. At the end of each preselected time increment (10 to 30 sec. in different runs) *N* was pinched off by hand until the next flask had been slid into position. After the run each flask was weighed to obtain the mercury weight and thus the average flow rate over the interval.

The zinc was dissolved in the hydrochloric acid by placing the flasks around the edge of an electric hot plate so that strong natural circulation occurred without boiling. The solution was completed when hydrogen evolution ceased and a film of HgCl appeared on the mercury on cooling. The solution and washings were collected, exactly neutralized with potassium hydroxide solution to the methyl red end point, and diluted to 1 liter in a volumetric flask, and a 10-cc. portion was diluted to 100 ml. with 0.1 *N* potassium chloride. This solution was then analyzed by either a manual or, preferably, an automatically plotting dropping mercury electrode, previously calibrated with six known solutions prepared from zinc and mercury in the same way. This was by far the most rapid method of analysis available and yielded reasonable accuracy.

For each interval of every run the following average quantities were calculated, corrections being made where pertinent for the change in average I.D. due to the weight of zinc dissolved up to the middle of the interval: diameter, mercury mass velocity, outlet concentration c_2, rate of solution w per unit wall area, mass transfer coefficient $k_D = w/(c_i - c_2/2)$, mass transfer factor j_D, and film concentration Reynolds number Re_f at $\bar{c}_f$. These values of j_D and Re_f computed over each interval were found to agree reasonably well with Equation (2), though averaging 11% higher (*4*). Plotting j_D and Re_f vs. the median time of the interval and extrapolating back to zero time yielded the points listed in Table 4 and plotted in Figure 7. The extrapolated points are slightly lower, as might have been expected because any roughening of the surface, as observed before in natural convection, would thus be eliminated. The average agreement with Equation (2) is accordingly slightly better. Linton and Sherwood found no difference between L/D values of 6 and 21. Therefore Equation (2) may be considered to apply well to liquid-metal turbulent mass transfer in pipes with fully developed velocity distribution for L/D ratios exceeding 6, and probably with adequate accuracy down almost to 0 L/D. Higher j_D values would be expected if the specimen lacked an inlet calming length. The one streamline run obtained agrees well with Equation (3)*, which may undoubtedly also be accepted for liquid metals.

FORCED-CONVECTION FLOW THROUGH PACKED BEDS

Several studies of mass transfer from packed beds to gases flowing through them have been carried out. In most

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*Figure 5 illustrates the applicability of mass transfer tests in determining local heat transfer coefficients by measuring the rate of mass removal.

*This point is plotted against $(Re)_b$, 6.7% larger than $(Re)_f$.

studies the range of Schmidt number was rather limited, and so its effect was diffcult to assess. Using the analogous mass transfer definition of j to that found (*1*) for heat transfer inside of tubes, Hougen and coworkers (*11*) have adopted $(Sc)^{2/3}$, as in Equation (2). Denton (*2*) deduced a slightly higher power of Sc, *that is, 0.72.*

Experiments with liquid solvents can cover much wider ranges of Sc and thus should be more reliable in yielding a general correlation. Gaffney and Drew (*6*) used beds of succinic and salicylic acid pellets of various quasispherical shapes dissolving in n-butanol, benzene, and acetone. They also took into account the β-naphthol–water data of McCune and Wilhelm (*14*). The total $(Sc)_f$ range thus covered was 160 to 13,000. Reasonable variations of fractional voids X in the bed were included in a single correlating curve by basing the Reynolds number on the average velocity in the voids, that is, by utilizing $(Re_p)_f/X$, where $(Re_p)_f$ is based on the superficial velocity V. The best exponent of $(Sc)_f$ was obtained as 0.58 and the final correlation is shown as the solid line in Figure 8. $(Re_p)_f/X$ ranged from 1 to 5,000 and D_p from 0.01 to 0.042 ft. It seems logical to compare any liquid-metal results with this correlation.

In this study (*8*) the mercury was purified after each run by agitating it with air in a Bethlehem Oxifier, allowing it to stand overnight, and running it through a Bethlehem gold disk "filter." The mercury was then tested with HNO_3 and $K_2Cr_2O_7$ solution as a check that the lead had been completely removed. The spheres were high-sphericity antimony-free No. 8 lead shot from National Lead Co. and No. BB from Remington Arms Co. and were 99.85% or higher in metallic-lead content. The shot was amalgamated within 30 min. before each run by being mixed with $1N HNO_3$ and mercury and drained on a screen with the aid of a suction probe. The shot was weighed beforehand to compute the number for calculating surface area and porosity, but the average diameter was obtained by calipering a number of pellets after amalgamation.

The apparatus was similar to that of Figure 6 but employed a thick-walled gauge-glass column between end plates in place of items H, I, and J. The column had seven to ten horizontal thicknesses of 14- and 24-mesh 0.02-in. iron wire cloth at both top and bottom, to distribute the flow and to hold the bed compact by its elasticity. The procedure also was similar to that of the previous test. Five time intervals, usually of 10 sec. each, were employed in most runs. The sample flasks in this case contained 50 or 100 ml. (excess) of 50% by volume glacial acetic acid. After weighing, the flasks were loosely stoppered and gently shaken for over 6 hr. to dissolve the lead totally. The lead was determined gravimetrically in the solution and wash waters by the usual chromate method. The constant conditions of each run and the observed results are given in Table 5. In the last three runs, at low Re, the total change in average shot diameter was under 1%, and average results for the run are reported. Over the first three runs the change in average diameter was 6, 7, and 2% respectively, and all results reported were obtained by extrapolation back to zero time.

The number of transfer units N.T.U. was calculated as $c_2/(\Delta c)_{lm} = \ln [c_i/(c_i - c_2)]$, from which the height of a transfer

TABLE 4. MASS TRANSFER IN FORCED CONVECTION OF MERCURY THROUGH ZINC TUBES

Run	°C.	Tube length L, ft.	Initial diam. D, ft.	Sc_f	Initial solution rate, g./sec.	Solubility, wt. %	Initial efflux conc., wt. %	$j_D \times 10^3$	$(Re)_f$	Initial mass velocity G, lb./(hr.)(sq. ft.)
4	25.0	0.176	0.0120	83.9	20.6	2.18	0.0183	2.95	4,560	22,767
5	23.0	0.182	0.0120	83.1	70.0	2.11	0.0275	5.25	1,140	7,624
6	24.7	0.182	0.0118	82.6	53.6	2.17	0.0235	3.00	12,400	58,400
7	24.6	0.187	0.0119	82.7	36.5	2.17	0.0264	4.30	7,890	39,190
8	24.8	0.184	0.0117	82.5	14.5	2.18	0.0310	4.57	3,350	16,530
9	24.1	0.186	0.0117	82.7	48.6	2.15	0.0302	4.00	11,120	53,790
11	23.4	0.186	0.0118	83.1	37.5	2.13	0.0279	4.00	8,620	42,110

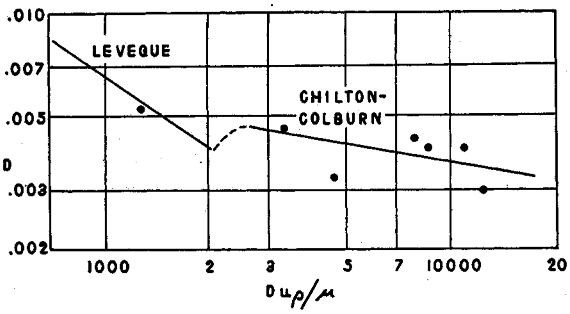

Fig. 7. Correlation of mass transfer in a tube with forced convection.

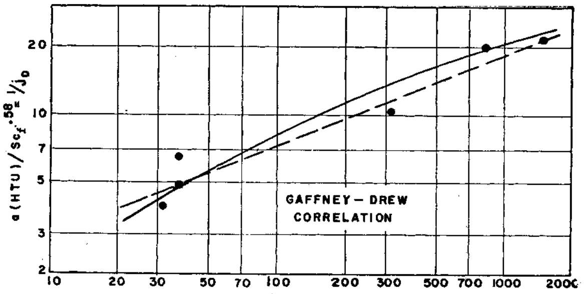

Fig. 8. Correlation of mass transfer from a bed of lead shot to flowing mercury.

TABLE 5. MASS TRANSFER IN FORCED CONVECTION OF MERCURY THROUGH PACKED BEDS OF LEAD SPHERES

Run	°C.	Efflux conc., wt. %	Solubility, wt. %	Transfer units, N.T.U.	Bed ht., ft.	H.T.U., ft.	Superficial velocity V, ft./sec.	Fractional voids, X	Shot diam., in.	Bed diam., in.	Re/X	Sc_f	$1/j_D$	$\frac{D_c}{D_p}$	$\frac{H}{D_p}$
1	26.5	0.341	1.721	0.2215	0,164	0.740	0.0721	0.499	0.0825	0.76	820	124.1	19.8	9	24
2	27.8	0.346	1.771	0.2167	0.174	0.803	0.1246	0.486	0.0845	0.76	1498	123.5	21.5	9	25
3	24.0	0.571	1.642	0.428	0.156	0.364	0.0242	0.446	0.0853	0.76	314	127.0	10.2	9	22
4	29.7	1.255	1.833	1.155	0.223	0.193	0.00325	0.507	0.0840	1.02	36.9	133.2	4.82	12	32
5	29.5	1.527	1.829	1.805	0.510	0.283	0.00371	0.555	0.0807	1.02	36.7	136.3	6.53	13	76
6	29.1	1.444	1.813	1.593	0.512	0.321	0.00134	0.498	0.1721	1.43	31.5	135.7	3.95	8	36

unit H.T.U. is obtained as H./N.T.U. Then j_D by the Gaffney and Drew definition was computed as $(Sc)_f^{0.58}/(a$ H.T.U.), which equals $(k_D/V)(Sc)_f^{0.58}$. For consistency with usual practice, the physical properties are taken at $\bar{c}_f$ rather than $\bar{c}$, though Gaffney and Drew did not correct to $\bar{c}_f$.

The agreement between these results and the Gaffney and Drew correlation is seen in Figure 8 to be roughly within the accuracy of the results. The dashed line, which would be more convenient for mathematical manipulations and should be adequately reliable, at least for liquid metals from $(Re_p)_f/X$ of 20 to 2,000, has the equation

$$j_D = \frac{k_D}{V}(Sc)_f^{0.58} = \frac{(Sc)_f^{0.58}}{a \times \text{H.T.U.}} = 0.87\left(\frac{(Re_p)_f}{X}\right)^{-0.40} \tag{4}$$

No significant effect of column diameter D_c seems to be present. The points at the two highest D_c/D_p values (the two upper points at low Re/X in Figure 8) lie above a smooth curve through the other points, but that is opposite to the expected deviation. Thus these correlations should be suitable for D_c/D_p down to somewhat below 8 with reasonable accuracy. The beds were at least twenty-two particles thick, and the results should be applicable to any bed thicknesses above a few particles.

CONCLUSIONS

1. Experiments on the dissolving of horizontal metal cylinders by mercury in natural convection and of zinc tubes and beds of lead shot by mercury in forced convection agreed within the experimental uncertainty with the best available dimensionless correlations for mass transfer in nonmetals under the same conditions (Figures 4, 7, and 8 respectively).
2. The logical supposition that mass transfer in all liquid-metal systems follows the same correlations as in nonmetals is strongly corroborated.
3. Under all the conditions investigated, mass transfer in liquid-metal systems follows, within the experimental uncertainty, the same correlations as does heat transfer in nonmetals, provided that the analogous mass transfer quantities are substituted for the heat transfer quantities (Schmidt number for Prandtl number, Sherwood number for Nusselt number, and solution expansion for thermal expansion in the Grashof number).
4. A preliminary acid treatment and amalgamation is in general desirable to eliminate surface oxide films and to obtain immediate uniform wetting of base metals by mercury at room temperature; otherwise consistent mass transfer results are not obtained.
5. With well-wetted metal surfaces there is no surface or "chemical" transfer resistance in addition to the usual liquid-phase transfer resistance.

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NOTATION

a = surface area per unit volume of packed bed, sq. ft./cu. ft.
c = concentration of solute, lb./cu. ft.
C = specific heat, B.t.u./(lb.)(°F.)
d = differential operator
D_v = diffusivity, sq. ft./sec. except where noted
D = diameter, ft.
F = the faraday, coulombs/g. equivalent
g = acceleration of gravity, ft./sec.^{2}
h = heat transfer coefficient, B.t.u./(hr.)(sq. ft.)(°F.)
H = height of bed, ft.
H.T.U. = height of 1 transfer unit, ft.
k = thermal conductivity, B.t.u./(sq. ft.)(hr.)(°F.)
k_D = mass transfer coefficient, ft./sec.
L = length, ft.
N = atom fraction
N.T.U. = number of transfer units
R = the gas constant, joules/°R.
t = time
T = absolute temperature, °R.
u = average velocity, ft./sec.
v = valence
V = "superficial" velocity through a packed bed, ft./sec.
w = rate of solution per unit area, lb./(sec.)(sq. ft.)

Greek Letters

β = coefficient of thermal expansion, °F.$^{-1}$
β_c = coefficient of volumetric expansion due to solution, cu. ft./lb.
γ = activity coefficient of a solute
Δc = concentration driving force, lb./cu. ft.
ρ = density, lb./cu. ft.
μ = viscosity, lb./(ft.)(sec.) or lb./(ft.)(hr.), as required in the dimensionless ratio

Dimensionless Groups

Gr_h = Grashof number for heat transfer $(D^3g\beta\Delta T\rho^2)/\mu^2$
Gr_m = Grashof number for mass transfer $(D^3g\beta_c(c_i - c)\rho^2)/\mu^2$
j_D = j factor for mass transfer, Sh $(Sc^{1/3}Re)^{-1}$
Nu = Nusselt number hD/k
Pr = Prandtl number $C\mu/k$
Sc = Schmidt number $\mu/\rho D_v$
Re = Reynolds number $Du\rho/\mu_{(pipes)}$ or $D_pV\rho/\mu X$ (packed beds)
Sh = Sherwood number k_DD/D_v
X = fractional voids

Subscripts

b = ambient or bulk average
c = column
f = film
h = heat transfer
i = interface
m or D = mass transfer
p = packing particles
0 = zero solute concentration
1 = inlet stream
2 = outlet stream

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