

Subscripts

g = gas phase
 s = solid phase

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Use of Electrochemical Techniques to Study Mass Transfer Rates and Local Skin Friction to a Sphere in a Dumped Bed

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Electrochemical techniques are developed to study the details of the flow around a single particle in a packed bed. Initial work has been done with a dumped bed of 1 in. spheres. Results are presented on the variation of the mass transfer coefficient and the shear stress around the surface of one of the spheres, the overall mass transfer rates to a sphere in a bed of inert spheres, and the effect of Reynolds number on the local mass transfer coefficient and the shear stress.

Present knowledge of the behavior of packed beds is limited because most of the measurements on these systems reflect average happenings over a large number of particles. A considerable amount is known about the flow field around isolated particles, and therefore it has been possible to explain measurements of heat and mass transfer and of drag on them. Similar knowledge is needed about flow around a particle in the presence of other particles in a packed bed.

In recent years a number of contributions have been made to our understanding of this problem. Kramers and Thoenes (10) suggested that the mass transfer coefficient for a single particle in a bed of inert particles is a better measure of what actually occurs between the particle surface and the fluid. They obtained overall mass transfer rates for fluids flowing around single spheres which were part of a regular arrangement of similar, but inactive spheres. Glaser (4) measured the overall heat transfer rate between air and an electrically heated sphere located

in a dumped bed of unheated spheres. Denton (2) and Wadsworth (21) have measured local heat transfer coefficients for air flow with a specially instrumented test sphere located in a regular packing arrangement. Korchak (9) has made turbulence measurements in the interstices of a regular array of spheres. Denton, who carried out his experiments at Reynolds numbers of 5,000, 20,000, and 50,000, located his test sphere at the center of a body-centered cubically packed bed. Wadsworth worked at Reynolds numbers of 8,000 to 60,000 and used the rhombohedral No. 6 blocked-passage array described by Martin, McCabe, and Monrad (11). Rhodes and Peebles (16) studied local mass transfer rates from single benzoic acid test spheres in cubic and rhombohedral arrays for Reynolds numbers of 488 to 3,410 by measuring the local diminution in radius of the test sphere after exposure to a measured rate of water flow for a given length of time.

This paper shows how electrochemical techniques that have been used to study turbulence close to a wall may be applied to the study of the flow around a sphere in a packed bed of spheres. They offer the possibility that in a single experiment one is able to (a) measure the shear

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stress variation and the mass transfer variation around a sphere, (b) monitor continuously the local mass transfer rate and the local shear stress, (c) examine some of the characteristics of turbulence that may exist, and (d) determine the overall rate of mass transfer. Polarized electrodes are imbedded at different locations on the surface of one of the packings to measure local instantaneous values of the mass transfer rate when the entire sphere is a mass transfer surface and when the sphere is inert. From data in the latter mode it is possible to estimate the shear stresses at the surface.

Initial work has been done with a 12-in. glass column containing a dumped bed of 1-in. spheres having a void fraction of 0.41. The Reynolds number (based on the fluid velocity in the empty column) was varied from 5 to 1,100. A transition from laminar to turbulent flow was found to occur in this system over the Reynolds number range 110 to 150 (6 to 8). Results were obtained on the variation of the time-average and fluctuating shear stress and local mass transfer coefficient around the meridian and the equator of the test sphere, the overall mass transfer coefficient, and the effect of Reynolds number on the overall mass transfer coefficient, local mass transfer coefficient, and shear stress.

The results on the variation of the local mass transfer rate and shear stress around the surface of the test sphere augment the studies of Denton, of Wadsworth, and of Rhodes and Peebles in that new types of measurements are obtained, a different packing arrangement is used, and a more detailed study of the effect of Reynolds number is made. The results on the overall mass transfer rates extend Glaser's measurements to higher Schmidt numbers and document the differences in the flow pattern which exist at different spheres in a dumped bed of spheres. These differences appear not only as differences in the pattern of variation around the surface of the sphere but also in the effect of changes in the Reynolds number on the transport rate.

These results have been encouraging with respect to the usefulness of electrochemical techniques in the study of the flow details in a packed bed. However, it has not been possible to exploit fully the detailed knowledge obtained so far because of the complicated flow pattern that exists in a dumped bed. Work is now under way with regular arrangements of spheres.

EXPERIMENTAL PROCEDURE

The system that was used to carry out the experiments is shown in Figure 1. The main section of the packed bed was contained in a 60 in. length of 12 in. diameter glass pipe. The column was packed by lowering 1 in. glass spheres through water in groups of about 100. Periodically during the packing process the upper layers of spheres were stirred and tamped with a long-handled pallet. At the inlet three glass reducers having a length of about 36 in. adapted the 12 in. main section to standard 1 in. steel pipe in the flow system. The entire column and the inlet were filled with glass spheres.

The electrochemical techniques developed by Reiss (14, 15), Mitchell (12), and Van Shaw (19, 20) were adapted to this study. An electrochemical reaction is carried out in the system using all or a portion of the test sphere as the cathode and a section of nickel pipe located outside the column as the anode. The electrolyte circulated through the system was an aqueous solution having equimolar concentrations approximately 10^{-2} M. of potassium ferro- and ferricyanide and a concentration of sodium hydroxide about 1 M. The electrochemical reaction consisted of the reduction of ferricyanide ion on a nickel cathode and the reverse reaction on a nickel anode. The properties of the electrolyte are summarized in Table 1. At high enough voltages the reaction rate is fast enough so that mass transfer is controlling; the concentration

of ferricyanide ion is approximately zero over the surface of the cathode. The electric current flowing through the solution to any portion of the cathode surface is directly proportional to the rate of mass transfer to that portion of the surface of the test sphere.

TABLE 1. ELECTROLYTE PROPERTIES

Property		Lowest value	Highest value
ferricyanide conc.	(moles/liter)	0.0046	0.0127
ferrocyanide conc.	(moles/liter)	0.0054	0.0131
sodium hydroxide conc.	(moles/liter)	0.890	1.28
density	(g./liter)	1.037	1.055
viscosity	(centipoise)	1.07	1.18
diffusion coefficient	(sq.cm./sec.)	0.59×10^{-5}	0.65×10^{-5}
Schmidt number		1,580	1,890

The test sphere was located 7 to 8 in. from the top of the packed section. A rest site was formed by arranging three glass balls such that their centers formed a triangle perpendicular to the column axis. The test sphere completed the regular tetrahedron. In placing the other spheres in the column, contact points at or near embedded electrodes were avoided as much as possible.

The test sphere used for the determination of the overall mass transfer coefficient for a single particle in a packed bed was constructed from a 1 in. diameter brass-bronze ball. A hole was drilled into the sphere along a diameter to provide a receptacle approximately $\frac{1}{4}$ in. deep for the lead wire. A 3 ft. length of 0.020 in. diameter nickel wire, which had previously been sheathed in 0.023 in. I.D. polyethylene tubing, was fitted snugly into the well and secured with an electrically conducting cement. The brass ball was chemically cleaned and plated with a mirror-bright surface of nickel by using a method described by Pinner and Kinnaman (13). The surface of the sphere was polished occasionally during the

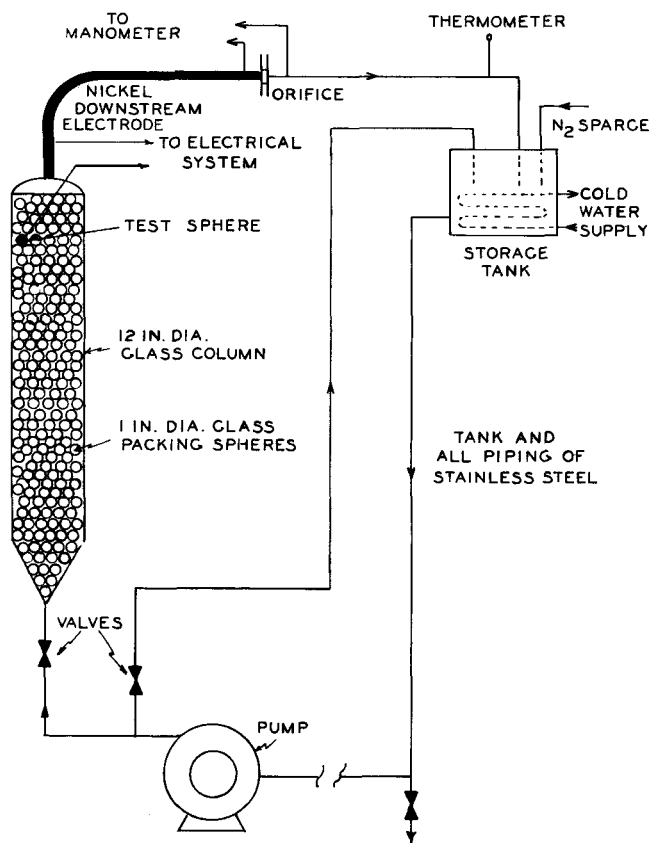


Fig. 1. Flow system.

course of the experimentation. A small drop of epoxy cement was applied to the test sphere at the junction with the lead wire. The hardened epoxy gave strength to the joint and also provided a watertight seal between the sphere surface and the polythylene tubing covering the lead wire. The small loss in the mass-transfer area resulting from the epoxy connection was taken into consideration in the calculation.

The test sphere used to measure the local mass transfer rate or the local shear stress was fabricated from two 1 in. diameter brass-bronze balls. A segment of one of the bronze balls was machined away leaving a flat surface parallel to an equatorial plane and 3/16 in. from it. Without changing the position of the ball in the lathe chuck, a flat-bottomed cavity was drilled into the sphere with its cylindrical walls perpendicular to the flat surface. The cavity had a diameter of 0.680 in. and a depth of 5/16 in. For one of the test spheres six 0.028 in. diameter holes were drilled through the cavity wall on sphere radii through the equatorial plane. These holes were equally spaced around one-half of the great circle. A second test sphere was made with fourteen nickel electrodes embedded at equal spacings around the entire great circle.

Short lengths of 0.020 in. diameter nickel wire were given a prime coat of insulation by immersion in freshly made epoxy. By suspending these vertically, the excess epoxy dripped off and a thin coat of insulation remained. The coated wires were inserted through the holes which had been filled with freshly made epoxy. After the epoxy hardened, the outer protrusions of the wire were trimmed and sanded flush with the outer surface of the sphere. When the sanding operation was completed, each electrode had the appearance of a tiny circular nickel disk surrounded by an insulating ring of hardened epoxy. Inside the sphere, the tips of the wires were soldered to 36-gauge conductors which were insulated from one another. In order to provide for electrical connection to the main surface, a 0.020 in. nickel wire was also passed through a port in the cavity wall and soldered to a threaded plug in the cavity floor. After all circuits were checked for continuity and insulation the cavity was poured full of fresh epoxy to within approximately 1/16 in. of the top.

The final steps in the fabrication involved fitting a cap to the semi-sphere and nickel-plating the main surface. A brass cap was machined from rod stock with a cylindrical shoulder that would fit down into the cavity leaving just enough clearance for the hardened epoxy plug. This shoulder rendered the cavity watertight and provided a firm electrical contact between the cap and the main surface of the semisphere. The most satisfactory method found for this was to machine the shoulder a little too small and electroplate both the semisphere and the cap separately until a firm press-fit could be obtained.

The lead wire bundle was brought out of the sphere with six electrodes through a 1/8 in. diameter hole at the midpoint of the other half of the great circle in which the electrodes were located. For the sphere with fourteen electrodes the lead wires came through a hole in the floor of the cavity. The lead wire bundles were sealed in place by using epoxy cement.

Two identical electrical circuits were available for simultaneous use when measuring correlations, and a third circuit could be added to handle the current to the main surface of the test sphere. These circuits are similar to those used by Reiss (14) and Mitchell (12) and are described in detail elsewhere (7).

Overall mass transfer measurements were made with a plated bronze ball. Both overall and local average mass transfer measurements were made with the test spheres with electrodes embedded in them. In the latter case the ferricyanide ion was reacting at the main surface and the surface of all of the test electrodes. The currents flowing to the main surface and to each of the test electrodes were measured separately.

The shear stress distribution around a packing was approximated by operating the 6-electrode sphere or the 14-electrode sphere in such a way that only the test electrodes and not the main surface of the test sphere were participating in the electrochemical reaction. Reiss and Hanratty (14) and Mitchell (12) have shown for the case of fully developed flow in a pipe that the average velocity gradient \bar{S} can be related to the average mass transfer coefficient \bar{K} to small electrodes of

length L in an inert surface by

$$\bar{K} = \frac{3}{2\Gamma\left(\frac{4}{3}\right)} \left(\frac{D^2\bar{S}}{9L}\right)^{1/3} \quad (1)$$

Jolls (7) has shown that Equation (1) can be used to approximate the shear stresses at the surface of a sphere except in the immediate vicinity of a stagnation point. One of the difficulties with this technique is that it does not reveal the direction of flow. Therefore, it is not feasible to map out the field with a few measurements unless there is some *a priori* knowledge of the flow pattern as might be the case with ordered beds.

VARIATION OF THE SHEAR STRESS AND THE MASS TRANSFER COEFFICIENT AROUND THE SURFACE OF A PACKING

Typical profiles for the variation of the mass transfer coefficient around the sphere are shown in Figures 2 and 3 and for the shear stress variation in Figures 4 and 5. As is the case for isolated spheres, the patterns of the mass transfer and shear stress variation can be quite different. These patterns are more sensitive to changes in the packing brought about by repacking the column than to changes in the flow rate. In nearly all of the runs the average of the local measurements was in reasonable agreement with the measured overall mass transfer rate. This would indicate that the measured profiles are representative.

From the shear stress profiles and pressure drop data for flow through dumped beds of spheres one can estimate the relative contribution of the skin drag to the total drag. The ratio appears to decrease with increasing Reynolds number, but an estimated average over the range of flow rates investigated is $\frac{\text{Skin Drag}}{\text{Total Drag}} \sim 0.1$.

This estimate is a rough one since it employed too few points to map out the shear stress with any assurance, and it involved the assumption that the main contribution to the drag on the particles in a bed is coming from the pressure variations around the particle surface rather than from the shear stress.

The measurements of the local mass transfer coefficient do not, in general, show as much variation on an equator

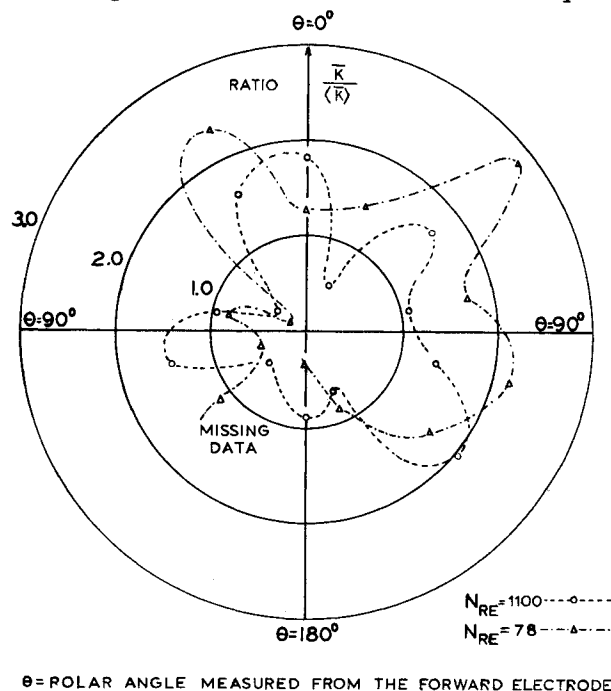


Fig. 2. Distribution of local-average mass transfer rate around a medium.

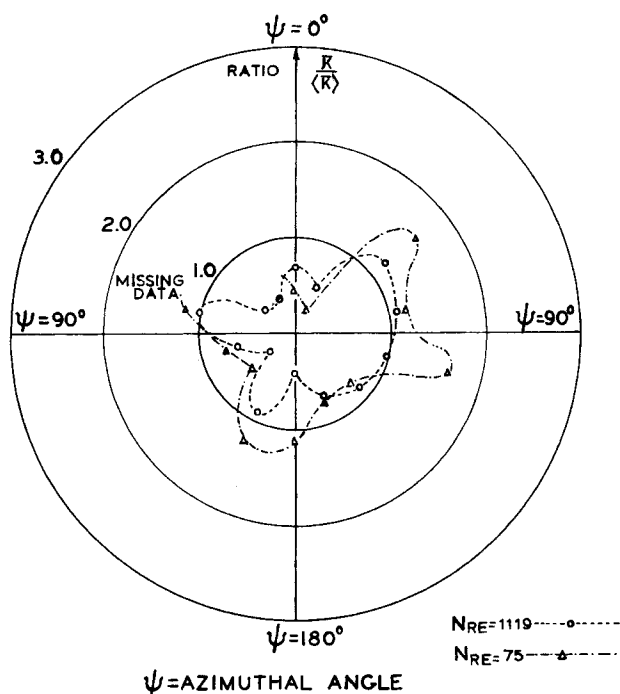


Fig. 3. Distribution of local-average mass transfer rate around the equator.

as they do around a meridian. The variation around a meridian is due to differences in the flow and in the concentration boundary layer at the front and back ends of the sphere as well as to the details of the packing arrangement. For this reason it is of more interest to summarize

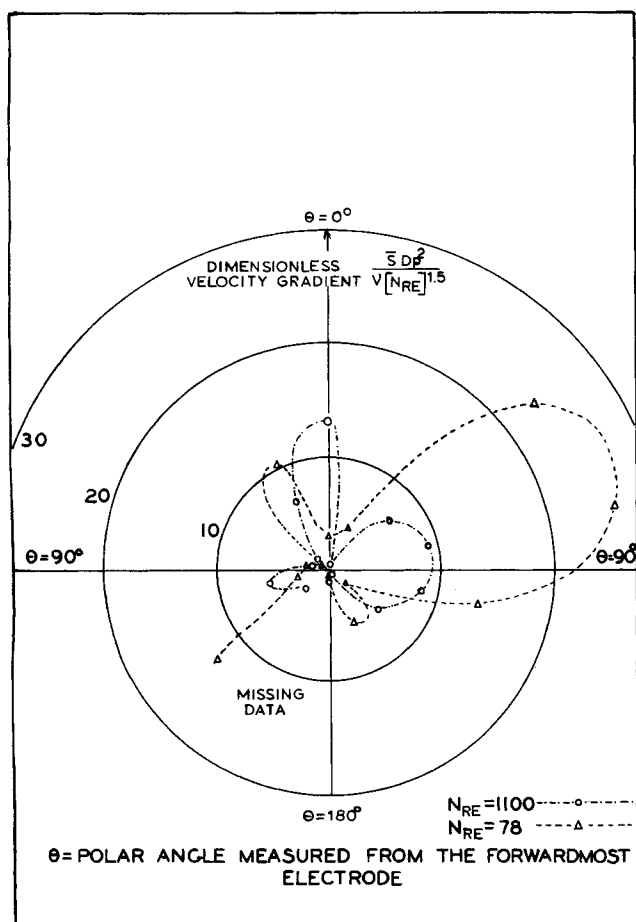


Fig. 4. Distribution of local-average shear stress magnitude around a meridian.

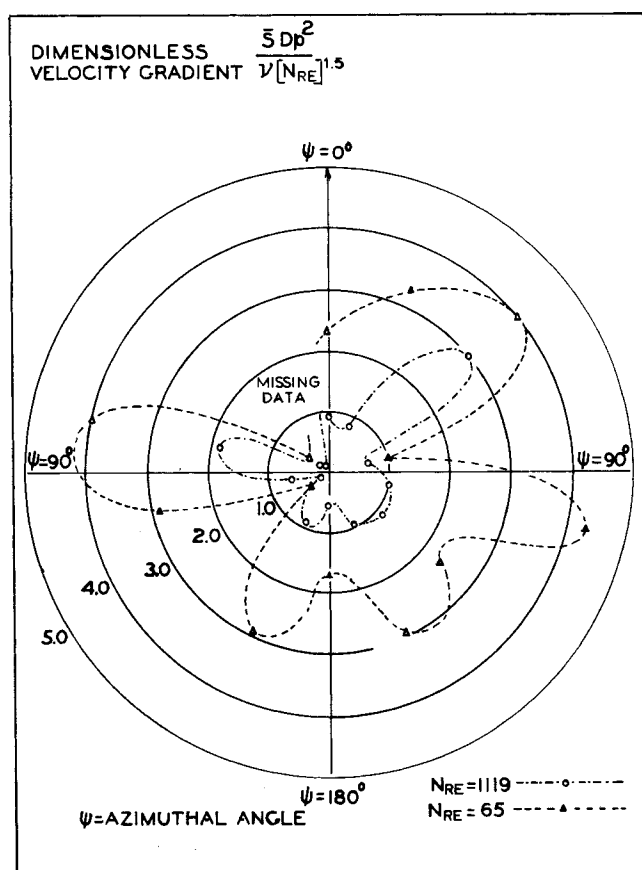


Fig. 5. Distribution of local-average shear stress magnitude around the equator.

some of the features of the seventy-eight profiles that were measured around a meridian.

Both the arithmetic average mass transfer and the variance from this average are significantly greater on the upstream half of the test sphere than on the downstream half. In fifty-two of the seventy-eight profiles the maximum mass transfer rate was recorded at one of the three electrodes in the most forward position on the test sphere, and in forty-one of these the electrode nearest the most forward point on the sphere indicated the maximum value. In an attempt to illustrate the relative contributions made by the forward and rearward hemispheres to the overall rate of mass transfer, arithmetic averages for both halves were calculated for each profile from a summation of the appropriate local values. The ratios $\langle \bar{K} \rangle_f / \langle \bar{K} \rangle_b$ are plotted against Reynolds number in Figure 6, where the results from a number of runs are presented to indicate reproducibility. The smaller values of the time averaged mass transfer rate to the back of the sphere can be explained because the concentration boundary layer will be thicker there and because the velocity gradient at the surface of the sphere is smaller there.

All of the profiles for the shear stress variation that were taken indicate a pronounced maximum in the neighborhood of the equator and relatively low values in the tail region.

At a location on the sphere where the flow is attaching, both the velocity and the concentration boundary layers are being initiated. Such a point could be identified by a location on the packing surface at which the signal from the test electrode was the same, irrespective of whether the remainder of the surface was active or not. In all of the runs there was an indication of attachment close to one of the most forward electrodes. All of the other electrodes gave higher signals when the sphere was inert. No evi-

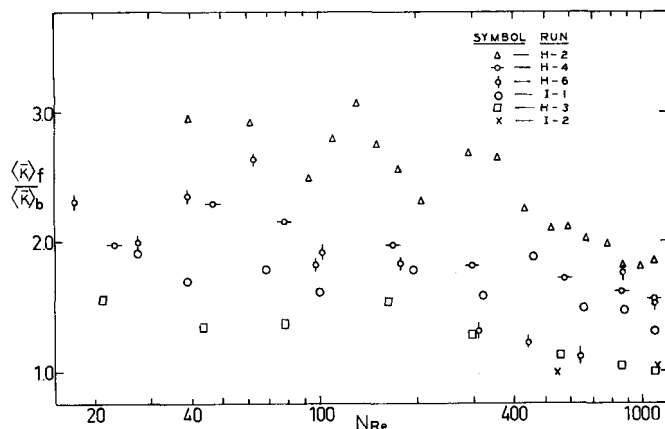


Fig. 6. Ratio of forward to rear mass transfer.

dence was obtained for the existence of an attachment point anywhere else on the surface of a sphere.

For symmetric flow around an isolated sphere the location on the forward regions of the sphere at which the flow attaches is a stagnation point. The external flow that is attaching to the sphere has only a normal component and no tangential component at the point of attachment. It is quite likely that a stagnation point does not exist on the forward part of a sphere in a dumped bed. Attachment does not occur at a point but along a line. At an attachment location the external velocity has a component tangent to the surface of the sphere.

This interpretation is suggested since measurements with isolated electrodes close to the apparent points of attachment often gave the largest mass transfer rate. At a stagnation point the shear stresses over the surface of the test electrode are quite small, and one would expect to find a minimum in the profile obtained from mass transfer mea-

OVERALL MASS TRANSFER

The overall mass transfer rates were correlated by plotting $\langle N_{Sh} \rangle N_{Sc}^{-1/3}$ vs. Reynolds number on log-log graph paper. It was found that each set of data could be fitted by a series of three to four connected straight lines. The data for a given packing arrangement were reproducible and show little scatter. However, a considerable change in the correlation could be effected by repacking the column or by placing the sphere at a different location in the bed. Three typical runs are plotted in Figure 7. The statistical variation is illustrated in Table 2 where the results of all the runs are summarized. The data for different ranges of Reynolds numbers are correlated by an equation of the form

$$\langle N_{Sh} \rangle N_{Sc}^{-1/3} = A N_{Re}^n \quad (3)$$

Data for Reynolds numbers less than 35, the low-range laminar region, exhibit considerably more scatter than do those for higher flow rates. In most cases, a straight line can be drawn through the data in this region, but the coefficients and exponents of the corresponding equations vary widely from run to run. An average value of n of 0.60 and of A of 1.64 is determined. However, the scatter of the data renders this result of questionable significance.

The radial location of the sphere was varied from a position approximately one particle diameter from the center of the column to a position touching the wall of the column. There appears to be no significant effect of the radial location of the test sphere if the Reynolds number is greater than 35. In the low-range laminar region, mass transfer rates for $R^* > 0.75$ are significantly higher and show much less dependence on the flow rate than those with the test sphere located nearer to the center of the column. This effect explains much of the run to run variation in the low-range laminar data shown in Table 2 since data for all radial positions were considered collectively in that case.

TABLE 2. STATISTICAL CHARACTERISTICS OF OVERALL MASS TRANSFER DATA

Region	Quantity	Number of observations	Avg. value	Lowest value	Highest value	% Avg. dev.	% Avg. dev.
Turb.	n	15	0.563	0.449	0.695	0.060	10.7
Turb.	A	15	1.588	0.610	2.804	0.485	30.5
$N_{Re} = 80$	n	17	0.581	0.495	0.643	0.022	3.8
$N_{Re} = 80$	A	17	1.400	1.152	2.040	0.153	11.0
$N_{Re} = 40$	n	17	0.574	0.494	0.715	0.040	7.0
$N_{Re} = 40$	A	17	1.469	0.778	2.149	0.271	18.5
low laminar	n	14	0.600	0.284	0.844	0.134	22.4
low laminar	A	14	1.640	0.585	4.166	0.824	50.2

surements with isolated electrodes. The mass transfer data for those electrodes which appeared to be at a location where the flow is attaching could be correlated with the following equation:

$$N_{Sh} N_{Sc}^{-1/3} = 6.4 N_{Re}^{0.5} \quad (2)$$

The most striking features of these profiles of the mass transfer coefficient and the shear stress around the surface of a packing are the very large variation in the mass transfer coefficient and differences in the behavior of the front and rear halves of a packing.

It is quite interesting that there were no dramatic changes in the average mass transfer rates accompanying the transition to turbulence since both visual studies and mass transfer measurements indicated large flow fluctuations (8). A run at a Reynolds number of 540 showed intensities of the fluctuating signal from electrodes in an inert sphere of from 0.04 to 0.18. At a $N_{Re} = 1,120$ the intensities ranged from 0.04 to 0.68.

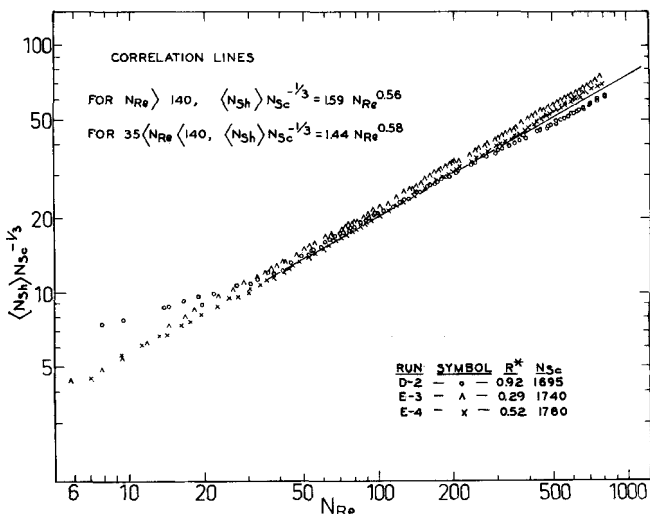


Fig. 7. Overall average mass transfer results.

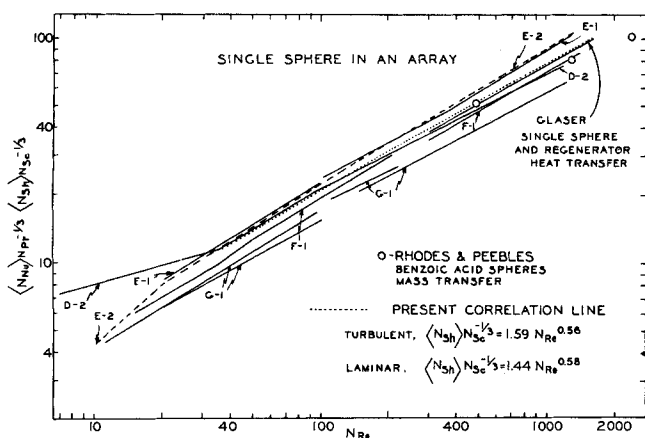


Fig. 8. Comparison of overall average mass transfer results.

It is therefore difficult to present a single correlation for mass transfer to a sphere in a randomly packed bed of spheres because of the statistical variation of the environment from sphere to sphere. However, it is possible to define an average behavior of a large number of experiments for which the bed was repacked before each experiment. The data from this investigation suggest that this average behavior be represented by the following:

$$N_{Re} > 140 \quad \langle N_{Sh} \rangle N_{Sc}^{-1/3} = 1.59 N_{Re}^{0.56} \quad (4)$$

$$140 > N_{Re} > 35 \quad \langle N_{Sh} \rangle N_{Sc}^{-1/3} = 1.44 N_{Re}^{0.58} \quad (5)$$

These equations are plotted in Figure 8 along with the results of several runs to illustrate the statistical variation around this average. It is noted that the differences in the correlations for results above and below the transition to unsteady flow are not statistically significant.

Equations (4) and (5) are in good agreement with the correlation derived by Glaser (4) from his experiments on heat transfer between air and a single sphere in a packed bed.

Results on mass transfer in packed beds, where all of the packings are exchanging mass, have recently been summarized by Gupta and Thodos (5). These data appear to be significantly lower than the results for a single sphere. The difference could be explained if the effective concentration driving force is less when all the spheres are active. In a bed of active spheres each particle is bathed in the concentration boundary layers from the surrounding particles.

It is also of interest to compare the results reported in this paper with those for isolated spheres. In their study of heat and mass transfer in the flow of gases past single

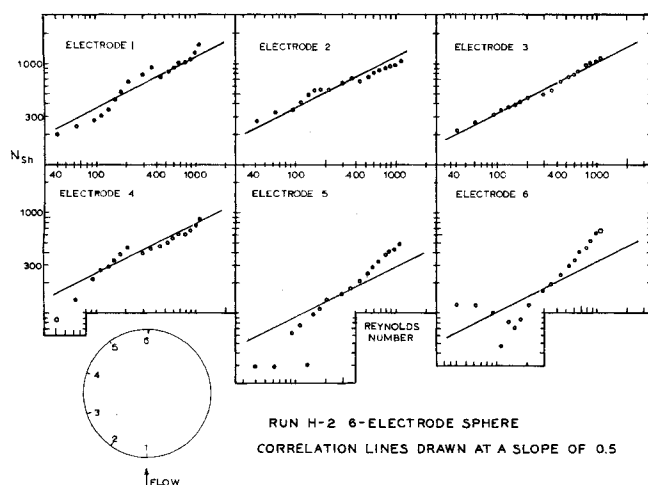


Fig. 9. Variation of the local mass transfer rate with Reynolds number.

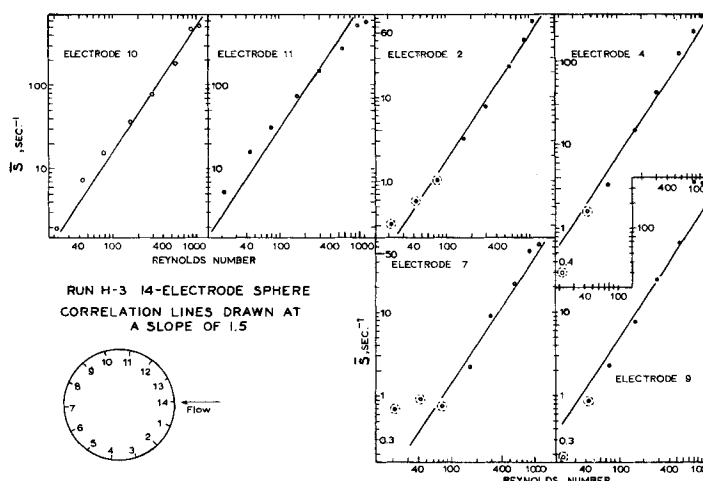


Fig. 10. Variation of the velocity gradient at the surface.

spheres, Evnochides and Thodos (3) found that

$$\langle N_{Sh} \rangle = 0.35 N_{Re}^{0.60} N_{Sc}^{1/3} \quad (6)$$

A comparison of Equations (4) and (5) with (6) would indicate that the transfer rates to single spheres in a packed bed are comparable to transfer rates for isolated spheres if the effective velocity for a packed bed is taken to be about nine times the superficial velocity.

EFFECT OF VARIATIONS IN THE FLOW RATE

The effect of variations in the Reynolds number on the local mass transfer coefficient and on the shear stress was found to be rather complicated. Typical results for the local mass transfer rate are shown in Figure 9 and for the shear stress in Figure 10 and in (8). The derivative of a curve through the data points does not vary in a monotonic fashion with increasing Reynolds number. The interpretation of these results, in contrast to results on an isolated sphere, is complicated by the possibility that a change in Reynolds number could bring about the changes in the basic flow pattern such as shifts in the locations of flow attachments.

The results that have been presented on the profiles of the variation of the mass transfer coefficient and shear stress suggest that flow over much of the surface of the sphere could be described by a three-dimensional boundary layer flow. According to boundary layer theory the local mass transfer coefficient should vary with the 0.5 power of the Reynolds number, and the shear stress should vary with the 1.5 power of the Reynolds number.

The lines drawn in Figures 9 and 10 have the slopes predicted by boundary layer theory. It is seen that reasonable agreement is obtained between the average trend of the data and the prediction except in the rear of the sphere. The agreement shown in these Figures is typical of all of the results that were obtained.

It is to be noted that in the rear of the sphere some of the measurements of local mass transfer indicate a larger variation with Reynolds number than would be indicated by boundary layer theory.

On the basis of these measurements of local mass transfer coefficients one could propose an explanation for the observed effect of Reynolds number on the overall mass transfer coefficient which is the same as that given for isolated spheres (18). The effect of Reynolds number on the overall mass transfer coefficient to a sphere in either a bed of inert spheres or in a bed of active spheres indicates a slightly larger power on the Reynolds number dependency than the 0.5 predicted by boundary layer theory. It would seem that over most of the surface the flow is of

a boundary layer type. However, there exists a small region in the rear of the sphere where the mass transfer rate shows a much greater dependency on Reynolds number than is predicted by boundary layer theory. Boundary layer theory is invalid in the rear of an isolated sphere because of flow separation. It is not clear as yet whether there is a region of separated flow in the rear of a sphere in a packed bed. It would seem that this is an interesting question to which future experiments should seek an answer.

CONCLUDING REMARKS ABOUT THE MASS TRANSFER RATE

Measurements of the overall rate of mass transfer to a single sphere in a dumped bed of inert spheres are presented for a Schmidt number of about 1,700. Results obtained by other investigators in which all the spheres in the bed are active are significantly lower since each particle is bathed in the concentration boundary layers from the surrounding particles. This supports the suggestion by Kramers and Thoenes that studies of mass transfer to single spheres in a packed bed offer a better opportunity to observe fluid-solid interaction. However, it has been found in this research that the variation in the flow pattern from sphere to sphere can cause considerable variation, not only in the magnitude but also in the effect of Reynolds number on the measurements obtained at different spheres. In order to obtain meaningful results it is necessary to average measurements from a large number of experiments. Such average measurements have been compared with the heat transfer study by Glaser at a Prandtl number of 0.7. The transfer coefficients obtained from the two investigations are in reasonable agreement if it is assumed that Sherwood number varies with $N_{Re}^{0.58}$ to $N_{Re}^{0.5}$ and with $N_{Sc}^{0.33}$. A comparison of this result to what has been reported for isolated spheres by Evnochides and Thodos indicates that the same equation can be used for isolated spheres and for spheres in a dumped bed if the effective velocity in a dumped bed is taken to be about nine times the superficial velocity. The measurements of the local mass transfer coefficients reported in this paper also show some similarities to what has been observed for isolated spheres in that the Sherwood number varies with $N_{Re}^{0.5}$ over all of the sphere with the exception of the very rearward portions.

CONCLUDING REMARKS ABOUT THE FLOW PATTERNS

Although the results of this study are insufficient to come to any definitive conclusions regarding the flow patterns in a packed bed, certain generalizations are suggested. The large variation in the local mass transfer coefficient and the shear stress around the surface of a sphere is in striking contrast with the assumption of a uniform mass transfer rate which is sometimes used to analyze the performance of packed beds. The distinct difference in the behavior of the front and back portions of a sphere in a packed bed seems to contradict the notion that a packed bed may be viewed as a system of interconnected channels. A model which emphasized the flow around individual particles is more consistent with the measurements obtained in this study (1, 17). With the exception of the very rearward portion of the spheres the effect of Reynolds number on the local mass transfer rate and on the local shear stress is what is predicted by boundary layer theory for isolated spheres. This would seem to suggest that flow over most of the surface of the sphere could be described by a three-dimensional boundary layer flow. An analysis based on boundary layer theory is com-

plicated by the very complex external flow patterns. This is emphasized by the observation that the pattern of the variation in the shear stress and the mass transfer rate is more sensitive to changes in the packing arrangement than to changes in the flow rate. A model of the flow is needed in order to carry out a more detailed analysis than is presently possible. It would seem that more information on the overall flow patterns is needed in order to guide the formulation of such a model.

ACKNOWLEDGMENT

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NOTATION

- D = diffusion coefficient
- D_p = particle diameter
- \bar{K} = time-averaged mass transfer coefficient
- L = length of the electrode
- N_{Nu} = Nusselt number based on the particle diameter
- N_{Re} = Reynolds number using a superficial velocity based on the empty column and the particle diameter
- N_{Sc} = Schmidt number
- N_{Sh} = Sherwood number based on the particle diameter
- \bar{S} = velocity gradient at the surface of the sphere
- R^* = dimensionless radial distance from the center of the column ($R^* = 1$ is at the wall)
- ν = kinematic viscosity
- $\langle \rangle$ = average over the surface of the particle

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