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Publication Date
1975-08-01

Submitted to Journal of the Electrochemical
Society

LBL-4145 c.1
Preprint

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August 1975

Prepared for the U. S. Energy Research and
Development Administration under Contract W-7405-ENG-48

Mass Transfer to a Rotating Disk in Transition Flow

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August, 1975

Abstract

An experimental study of mass-transfer to a rotating disk electrode has been employed to determine an empirical correlation for the mass-transfer rate in the Reynolds number regime lying between simple laminar and turbulent flows. This domain, apparently characterized by a regular vortex pattern, was found to extend from a Reynolds number of 2.0×10^5 to approximately 3.0×10^5. The resultant correlation for the average transfer rate, in terms of dimensionless Nusselt, Reynolds, and Schmidt numbers, is

$$\overline{Nu} = 0.89\times10^5 Re^{-1/2} Sc^{1/3} + 9.7\times10^{-15} Re^3 Sc^{1/3} .$$

Introduction

The mass-transfer behavior of the rotating disk electrode is of continuing interest. While the bulk of its uses are based upon the uniform-accessibility property described by Levich[1] for disks upon which simple laminar flow[2] prevails, for sufficiently high rotation speeds turbulence may be reached near the edge of the disk. The non-uniform (enhanced) mass-transfer rate resulting from this turbulence may be a desirable feature for some studies. In particular, this behavior has been found useful for studies of corrosion, when the effect of a varying local rate of oxygen transport to the surface of the corroding metal is of interest.[3-6]

An accurate description of the behavior of such an electrode requires knowledge of the local transfer rate for the full range of Reynolds numbers involved. The Reynolds number found appropriate for the disk is $Re = r^2\Omega/\nu$, where r is the radial distance from the center of rotation, Ω the angular rotation speed, and ν the kinematic viscosity of the fluid in which the disk rotates. This behavior for simple laminar flow, as described by von Kármán,[2] is well known, having been first described by Levich,[1] who gave the mass-transfer rate as

$$\overline{Nu}_{lam} = 0.6205\ Re^{1/2}Sc^{1/3} , \qquad (1)$$

where the Nusselt number $\overline{Nu} = \frac{\bar{i}r_o}{nFD\Delta c}$, and the Schmidt number $Sc = \nu/D$. $\bar{i}$ is the average current density on the electrode, r_o the electrode radius, n the number of electrons transferred per ion reacting, F Faraday's constant, D the diffusion coefficient of the active species, and Δc is the concentration driving force for diffusion--bulk concentration minus concentration at the electrode surface.

The transfer rate for well developed turbulent flow has been the subject of several studies,[7-12] and accurate correlations have been proposed for its dependence upon the Reynolds number. The two most useful of these correlations are due to Ellison and Cornet[7] and Daguenet,[9] since both of these also include the Schmidt number influence upon the Nusselt number. Ellison and Cornet report that the mass transfer may be described as

$$\overline{Nu}_{turb} = 0.0117\ Re^{0.896} Sc^{0.249} , \tag{2}$$

while the result of Daguenet is

$$\overline{Nu}_{turb} = 0.00725\ Re^{0.9} Sc^{0.33} . \tag{3}$$

We have chosen to use Daguenet's equation for two reasons. First, the bulk of the data gathered by Ellison were for larger Reynolds numbers than are of interest here $(Re > 10^6)$, while Daguenet thoroughly investigated the flow regime immediately adjacent to the transition region $(3\times10^5 < Re < 10^6)$. Second, in light of the many studies of turbulent transfer in different geometries, it seems that the exponent 1/3 is more realistic than 1/4 for the Schmidt number dependence. This conclusion is supported by the investigations of Donovan et al. for turbulent boundary layers,[13] Hubbard and Lightfoot,[14,15] and Vielstich et al.[16] The influence of the structure of the turbulent flow upon this exponent is discussed by Levich.[17] Also, Ellison and Cornet's data for $Re < 10^6$ are quite scattered and may possibly be better fit by a $Sc^{1/3}$ dependence.

However, we have refit Daguenet's data, as shown in figure 1, according to the correlation

$$\overline{Nu}\ Sc^{-1/3} = 0.0078\ Re^{0.9} - 1.38\times10^5\ Re^{-1/2}\ . \tag{4}$$

This form is based on the supposition that the local asymptotic transfer rate is proportional to $Re^{0.9}$, although overall Nusselt numbers for $Re < 10^6$ are still significantly dependent upon transfer in the laminar and transition zones. The coefficient of the last term in equation 4 will thus be adjusted subsequently by integration of local rates in the laminar and transition zones, while the first term should depend only on the asymptotic behavior in the turbulent regime. This leads to the following correlation for the large Reynolds number asymptotic mass-transfer behavior:

$$\overline{Nu}_{turb} = 0.0078\ Re^{0.9} Sc^{1/3}\ . \tag{5}$$

The problem remains to describe the mass-transfer behavior as a function of Reynolds number for the region in which the simple laminar flow becomes unstable and yields to turbulence. The range of Reynolds numbers over which this change takes place has been termed the "transition region," and the experimental determination

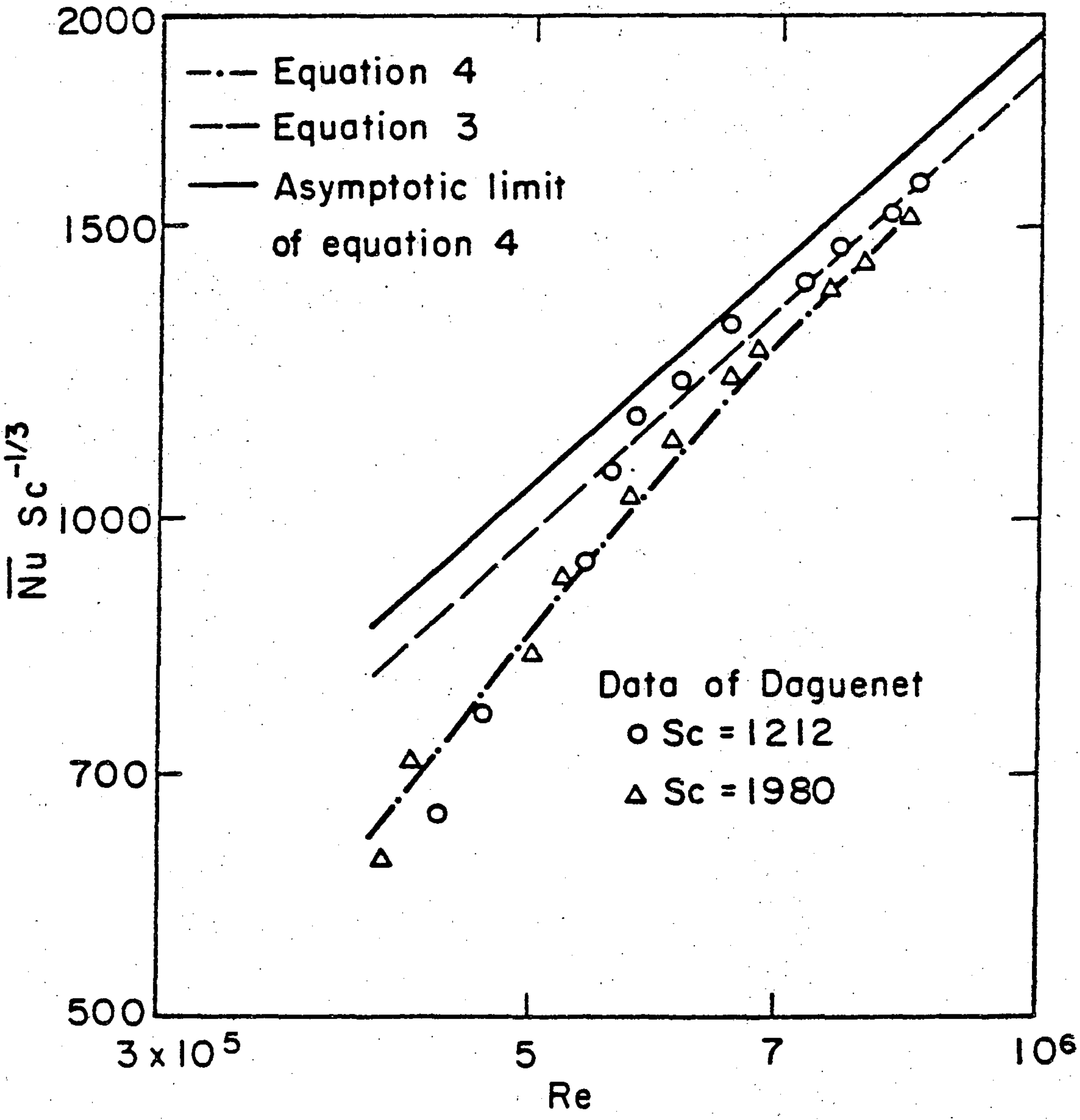

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Figure 1. Correlation of Daguenet's data for turbulent mass transfer to a rotating disk.

of its mass-transfer behavior is the purpose of this investigation.

The nature of this transition region was first described by Gregory, Stuart, and Walker,[18] who demonstrated that the simple laminar flow found near the center of the disk becomes unstable at a Reynolds number equal to approximately 1.8×10^5, and that fully developed turbulence was not reached until $Re \sim 3\times10^5$. Stuart[18] also developed a nonlinear stability analysis which showed that stable periodic solutions to the Navier-Stokes equation could be expected for this range of Reynolds numbers. The experimental and theoretical results both point to the presence of a stable vortex pattern, rather than a predominantly random flow, in the transition region.

Subsequent work by others has served to reinforce the conclusions of Gregory et al. In particular, the work of Chin and Litt[19] has provided perhaps the most accurate determination of the critical Reynolds numbers bounding the transition region. These investigators embedded a small electrode in a disk at some distance from the center of rotation. By varying the rotation speed, the electrode was subjected to a range of Reynolds numbers. A spectral analysis of the current signal to the electrode was used to determine if a regularly varying mass transfer mode was present, indicating vortices moving across the electrode. Their results agreed well with Gregory et al.[18] for the lower bound of the transition region, their value being $Re = 1.7\times10^5$, but they found evidence that the vortices persist to a Reynolds number of about 3.5×10^5, well beyond the value reported by Gregory.

Various studies of mass transfer in the turbulent flow regime also include some data in the transition region. Kreith, Taylor, and Chong[11] report significant deviation from values predicted by the Levich analysis for laminar flow beginning at a Reynolds number of about 2×10^5. The rather scattered data of Cobb and Saunders[12] tend to agree with this. Daguenet[9] reported a lower bound of the transition region of approximately 2.6×10^5, but this value varied with the Schmidt number ($Sc = \nu/D$, where D is the diffusion coefficient of the active species) of the solution. An experimental study performed by Tien and Campbell[10] agreed more closely with Chin and Litt in that they reported enhanced mass transfer beginning at $Re = 1.8\times10^5$. Ellison and Cornet[7] have also performed an experimental (and theoretical) investigation of mass transfer in the turbulent regime. While their data for low $(<3\times10^5)$ Reynolds numbers is somewhat scattered, they state that significant deviation from the Levich theory does not begin until $Re = 3\times10^5$ is reached.

In view of the lack of agreement about the value of the Reynolds number at which the Levich theory becomes inappropriate and the lack of a correlation for mass transfer in the transition region, an experimental study of this problem was performed. It is hoped that a more exact treatment of the dependence of the local transfer rate upon the Reynolds number, particularly in the transition region, will be found useful for such studies in which variation of the local mass-transfer rate (as a function of position) is desired.

Experimental Work

Electrochemical Systems of Interest

Two reactions were chosen for study: the electrodeposition of copper from a cupric sulfate - sulfuric acid solution onto a rotating copper disk, and cathodic reduction of ferricyanide ions to ferrocyanide ions, also on a copper disk, from a potassium ferricyanide - potassium ferrocyanide - potassium hydroxide solution. Concentrations of the active species were approximately 0.005 molar, while 1.5 molar sulfuric acid and 0.85 molar potassium hydroxide were used as supporting electrolytes. For the ferricyanide reduction, an excess of ferrocyanide was used (c ~ 0.007 molar) to help insure that the limiting current was achieved on the cathode (disk) before excessive oxygen evolution occurred at the anode. Oxygen evolution at the anode of the ferricyanide system is not desirable since it reduces the amount of ferrocyanide oxidized, thus depleting the solution of ferricyanide. A counterelectrode of copper was used for the cupric-acid system to provide the atomic copper for the anodic reaction. A nickel counterelectrode was used for the ferricyanide system anode.

Experimental Apparatus

A series of three PTFE slinger rings on the rotating shaft, each in its own compartment, was used to prevent loss of the solution at high rates of revolution and minimize aeration of the solution during the trials. The electrode consisted of a copper disk approximately 2 1/4 inches in diameter soldered to a 1/4" - 20 machine screw which provided electrical continuity to the shaft. The back and edge of the electrode were insulated with Shell epoxy casting resin which was extended 1/4 inch beyond the edge of the electrode to reduce the effect of the hydrodynamic disturbance due to the edge of the disk upon the flow field over the conducting area. A series of six baffles around the perimeter of the cell were to minimize bulk rotation of the fluid.

The peripheral electronic equipment consisted of a galvanostat (with a built-in current ramp generator) which was built to order for high current applications. Continuous polarograms were recorded on a Hewlett-Packard model 7044A x - y plotter. Current measurements were made by placing a precision resistor (0.1495 Ω, 25 watt maximum power dissipation) in the lead to the counterelectrode, and the voltage drop across it measured directly using the x - y plotter. The rotation speed of the electrode was determined with a "Strobotac" stroboscopic tachometer.

Electrode Preparation

The electrode was polished prior to each series of runs with 4/0 grit emery paper followed by buffing on a canvas polishing wheel coated with six micron diamond paste to a mirror finish. Following both the polishing and buffing, the electrode was washed with Amway L.O.C. cleaning solution followed by rinsing with distilled water and ethanol and drying in a stream of hot air. Due to the low concentrations of Cu^{++} in the cupric sulfate solutions (approximately 0.005 molar) and the short duration of the trials, the surface remained quite smooth and mirrorlike even after a series of fifteen to twenty trials. The trials involving the ferricyanide solutions were observed not to alter the surface detectably during the experiments, although the copper would apparently corrode if allowed to remain in contact with the KOH solution.

Experimental Procedure

For each series of trials, the cell was disassembled and cleaned, and the proper counter and reference electrodes were installed and polished with emery paper. Freshly prepared solution was introduced into the cell, with care being taken to exclude air bubbles. The solution level found to minimize apparent aeration at high rotation speeds was reached when the surface was approximately at the same level as the middle slinger ring, which required about 1.6 ℓ of solution. The polarograms were recorded at various rotation speeds, noting the temperature of the solution at each trial. A series of fifteen trials was found to increase the temperature about 1°C. Typical polarograms for a series of measurements are shown on figure 2. Viscosities of the solutions were determined for the range of temperatures encountered using a capillary viscometer. Diffusion coefficients were measured with the experimental electrode system operated at low revolution speeds using the Levich formula[1] for laminar flow to a rotating disk. For slight temperature variations, the Stokes-Einstein equation $D\mu = CT$, where T is °K and μ = absolute viscosity, was used. During the run, measurements were made at low rotation speeds to monitor possible concentration variation, a problem which occurred in the ferricyanide system due to oxygen evolution at the anode.

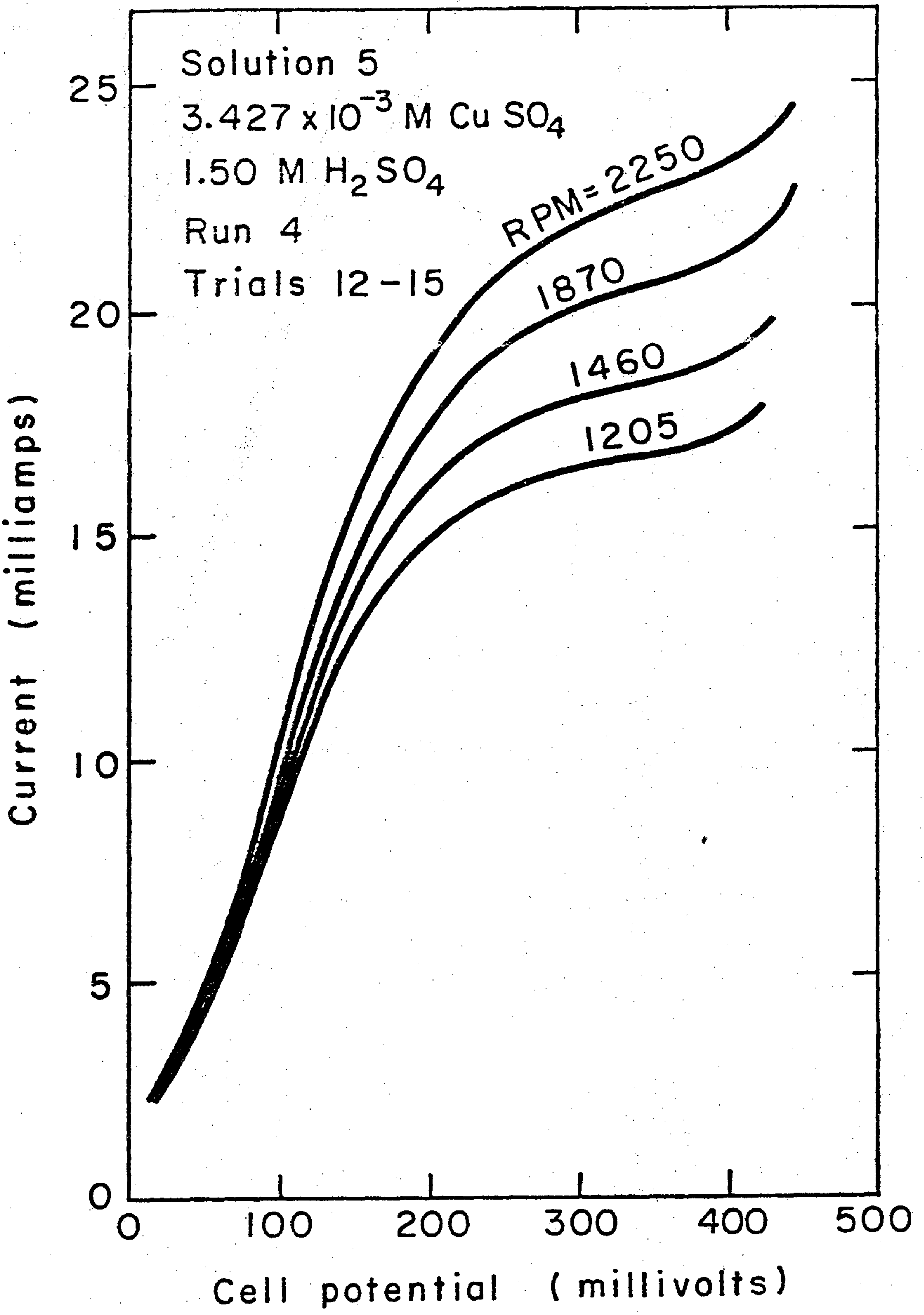

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Figure 2. Typical polarograms for mass transfer to a rotating disk.

Results

The experimental results are presented on figure 3 as a graph of $\overline{Nu}\ Sc^{-1/3}$ versus Re. The data gathered for $Re < 1.4\times10^5$, used to determine the diffusivity of the reacting species and monitor concentration changes, are not shown on this figure. The departure from the predictions of the Levich formula, equation 1, are observed to begin at approximately $Re = 2\times10^5$, in agreement with previous studies. The approach to the values predicted by the equation 5 is also shown.

To determine the local mass-transfer rate as a function of Reynolds number from the experimental data and equations 1 to 5, one may use the expression[4,5]

$$Nu_{loc} = \frac{i_{loc} r}{nFD\Delta c} = \frac{1}{2} \frac{d}{d(Re^{1/2})} [\overline{Nu}\ Re^{1/2}] . \qquad (6)$$

$\overline{Nu}\ Sc^{-1/3} Re^{1/2}$ is shown on figure 4 as a function of the square root of the Reynolds number. While the scatter in the data detracts from the accuracy of the derivatives determined from the curve fit, the results should still be useful in providing an estimate of the local and transfer rates which lie within the data scatter. It should provide an improvement over previous work[4,5] in which the local transfer rate for the laminar region was spliced (with considerable discontinuity) directly to the local transfer rate predicted from the work of Ellison and Cornet, equation 2.

The local Nusselt number derived from equation 1 (using equation 6) is

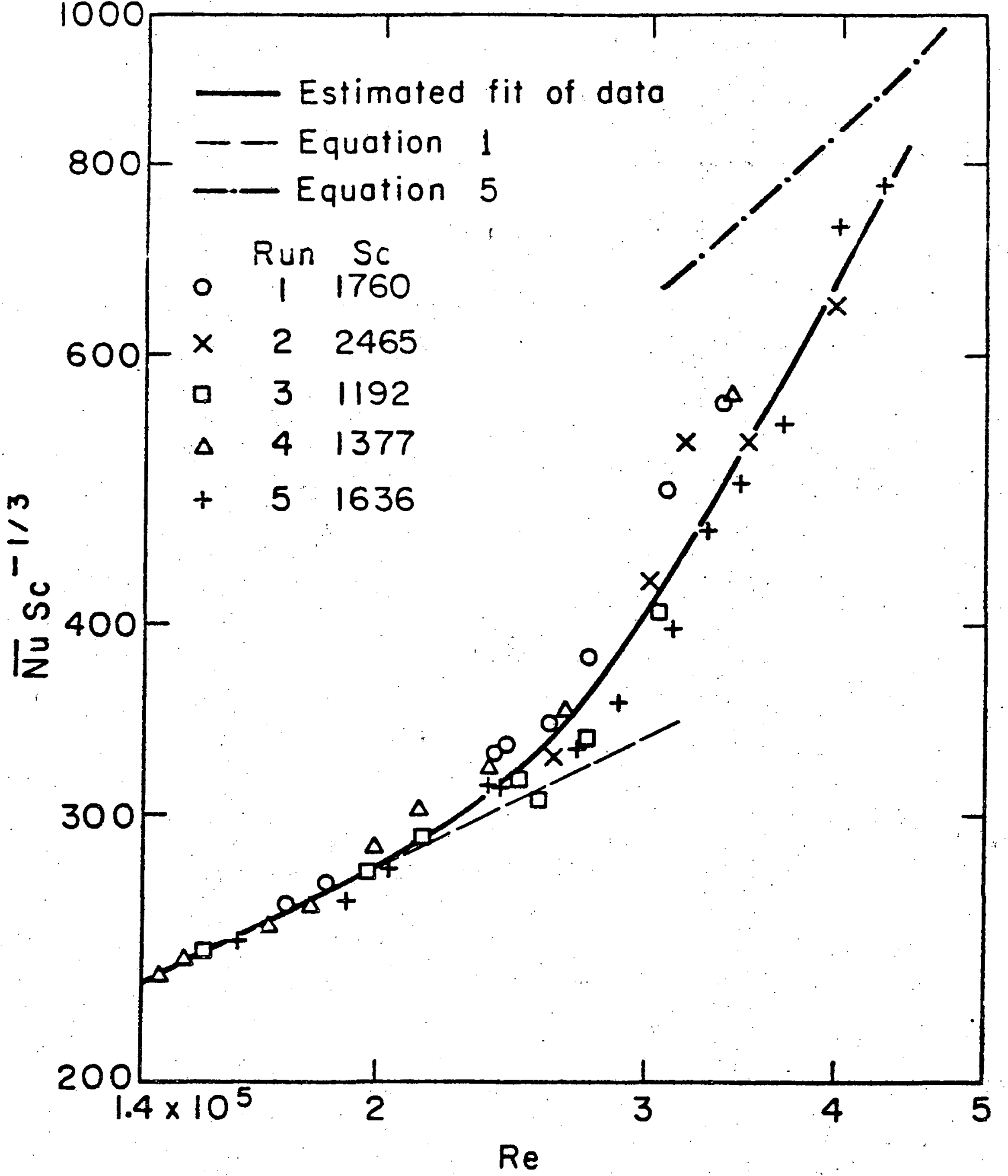

Figure 3. Overall mass-transfer rate ($\overline{Nu}Sc^{-1/3}$) versus Reynolds number for laminar, transition, and turbulent regimes. Standard deviations for the Schmidt numbers are 31, 41, 26, 38, and 57 for runs 1 to 5. Runs 3 and 4 involve deposition of copper; the others are reduction of ferricyanide.

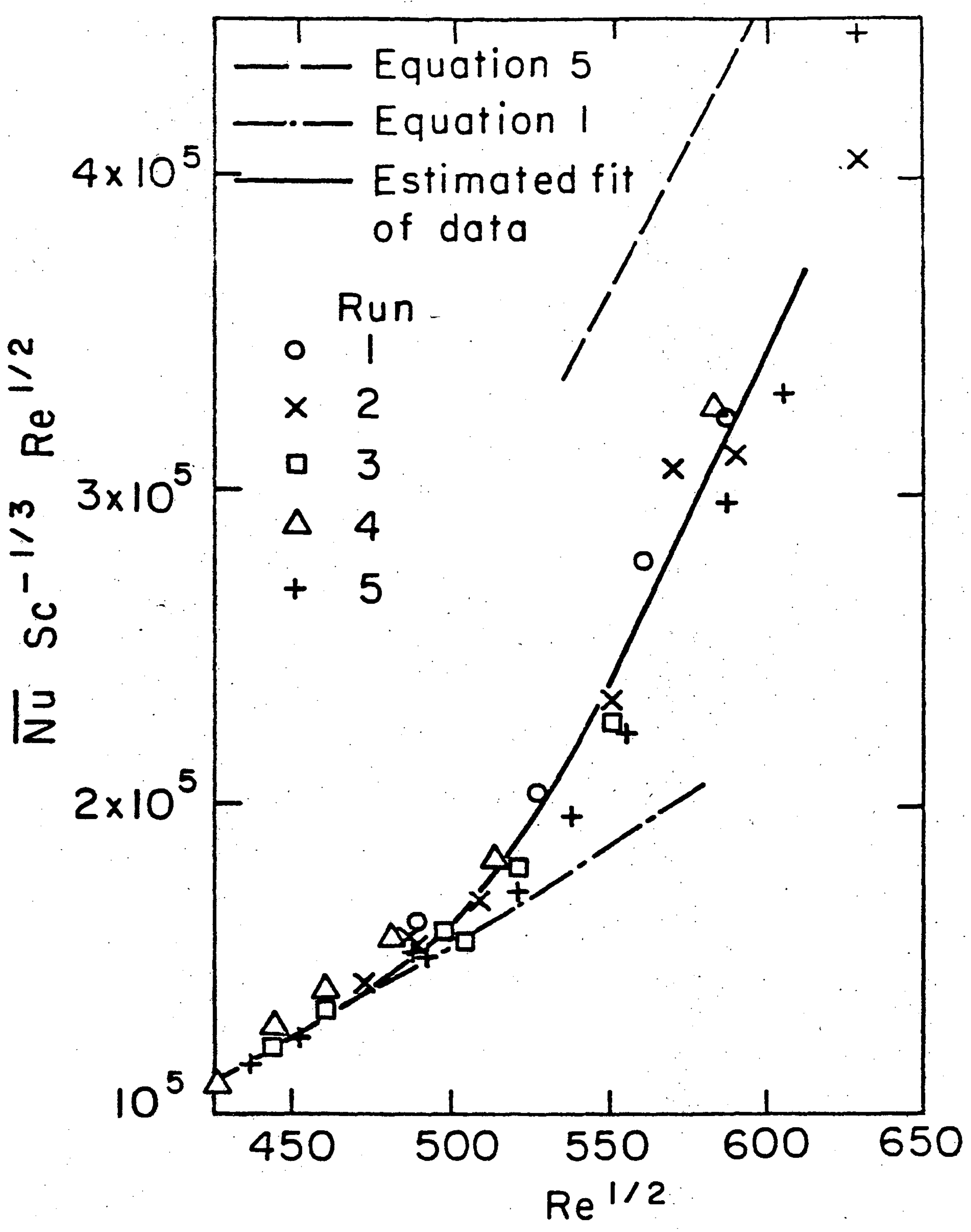

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Figure 4. Overall mass-transfer rate versus square root of the Reynolds number.

$$Nu_{lam} = 0.6205\ Re^{1/2} Sc^{1/3} . \tag{7}$$

In a similar manner, equation 5 yields

$$Nu_{turb} = 0.01092\ Re^{0.9} Sc^{1/3} . \tag{8}$$

These equations, along with the local Nusselt numbers for the transition region determined by differentiation of the curve fit given in figure 4, are given in figure 5. A straight-line fit of the data for the transition region seems reasonable; the one shown dotted in figure 5 leads to a correlation for the local Nusselt number

$$Nu_{trans} = 3.4\times10^{-14} Re^{3} \tag{9}$$

for Reynolds numbers between 2.0×10^5 and 3.0×10^5 and is continuous with the laminar and turbulent equations 7 and 8 at these values of Re .

Given these results for the local transfer rate in the three regions, we can reconstruct the overall transfer-rate behavior by integrating equation 6. For the laminar region, equation 1 is unchanged. In the transition region, $2\times10^5 < Re < 3\times10^5$, integration of equation 9 leads to

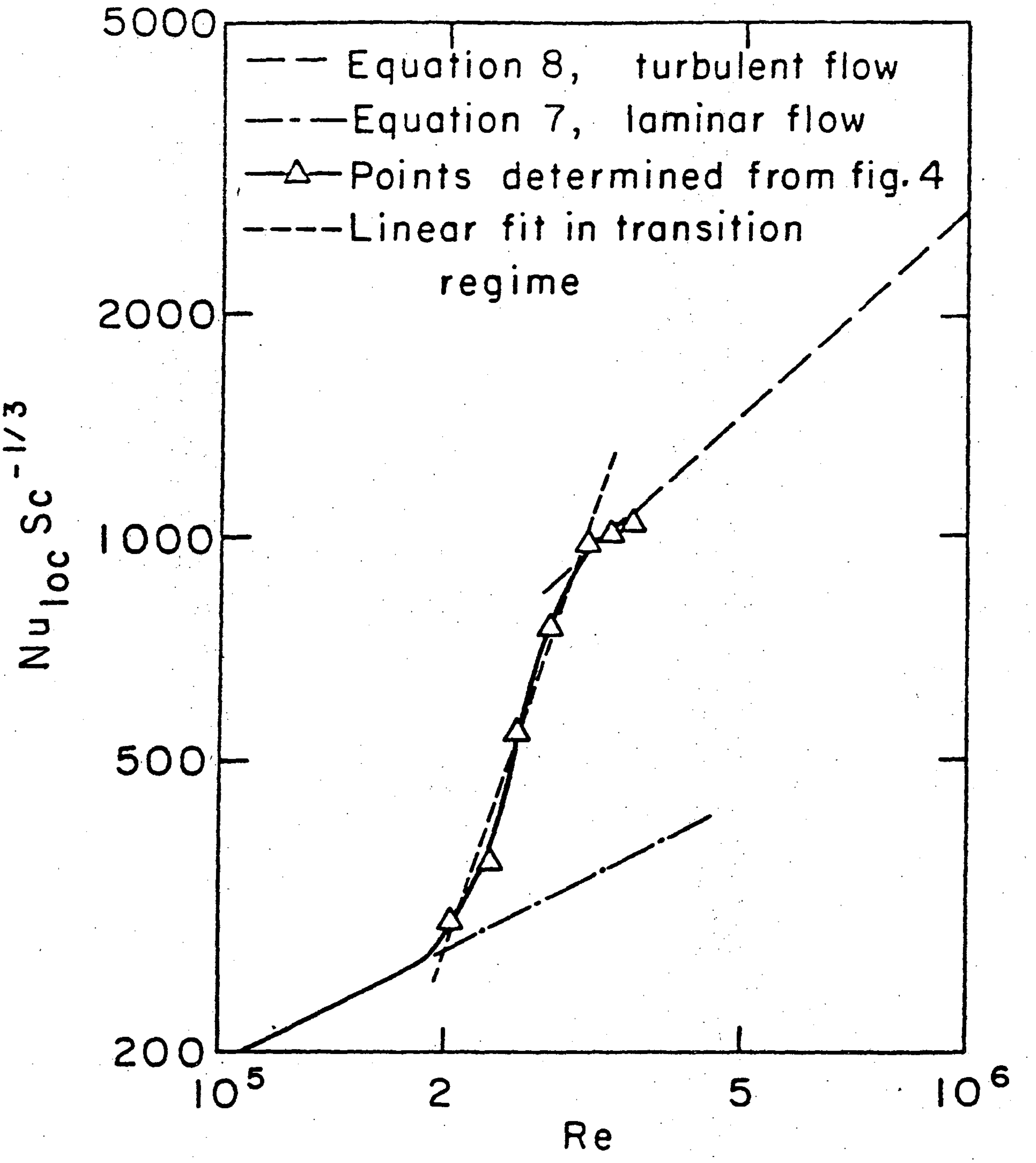

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Figure 5. Local mass transfer rate versus Reynolds number for transition, laminar, and turbulent regimes.

$$\overline{\mathrm{Nu}}_{\mathrm{trans}}\mathrm{Re}^{1/2} = 2\int_{0}^{\sqrt{2\times10^5}} \mathrm{Nu}_{\mathrm{lam}}\,d(\mathrm{Re}^{1/2}) + 2\int_{\sqrt{2\times10^5}}^{\sqrt{\mathrm{Re}}} \mathrm{Nu}_{\mathrm{trans}}\,d(\mathrm{Re}^{1/2})$$

$$= (0.89\times10^5 + 9.7\times10^{-15}\mathrm{Re}^{3.5})\mathrm{Sc}^{1/3} \qquad (10)$$

For the turbulent region, $\mathrm{Re} < 3\times10^5$,

$$\overline{\mathrm{Nu}}_{\mathrm{turb}}\mathrm{Re}^{1/2} = 2\int_{0}^{\sqrt{2\times10^5}} \mathrm{Nu}_{\mathrm{lam}}\,d(\mathrm{Re}^{1/2}) + 2\int_{\sqrt{2\times10^5}}^{\sqrt{3\times10^5}} \mathrm{Nu}_{\mathrm{trans}}\,d(\mathrm{Re}^{1/2})$$

$$+ 2\int_{\sqrt{3\times10^5}}^{\sqrt{\mathrm{Re}}} \mathrm{Nu}_{\mathrm{turb}}\,d(\mathrm{Re}^{1/2}) = (0.0078\ \mathrm{Re}^{1.4} - 1.30\times10^5)\mathrm{Sc}^{1/3} \qquad (11)$$

when the appropriate expressions are substituted for $\mathrm{Nu}_{\mathrm{lam}}$, $\mathrm{Nu}_{\mathrm{trans}}$, and $\mathrm{Nu}_{\mathrm{turb}}$.

The overall Nusselt numbers given by equations 1, 10 and 11 are shown on figure 6 as a function of Reynolds number. The curve representing the fitted experimental data is also shown, along with the modified correlation of Daguenet for the turbulent region. The graph supports the statement by Ellison and Cornet[7] that for $\mathrm{Re} > 10^6$ only the contribution to the overall mass transfer due to the turbulent region is of consequence. At $\mathrm{Re} = 10^6$, the difference between the correlation of Daguenet, equation 5, and the results which include the contribution of the laminar and transition regions, equation 11, is less than seven percent. This is within the range of the maximum deviation of the data from the fitting curve. The

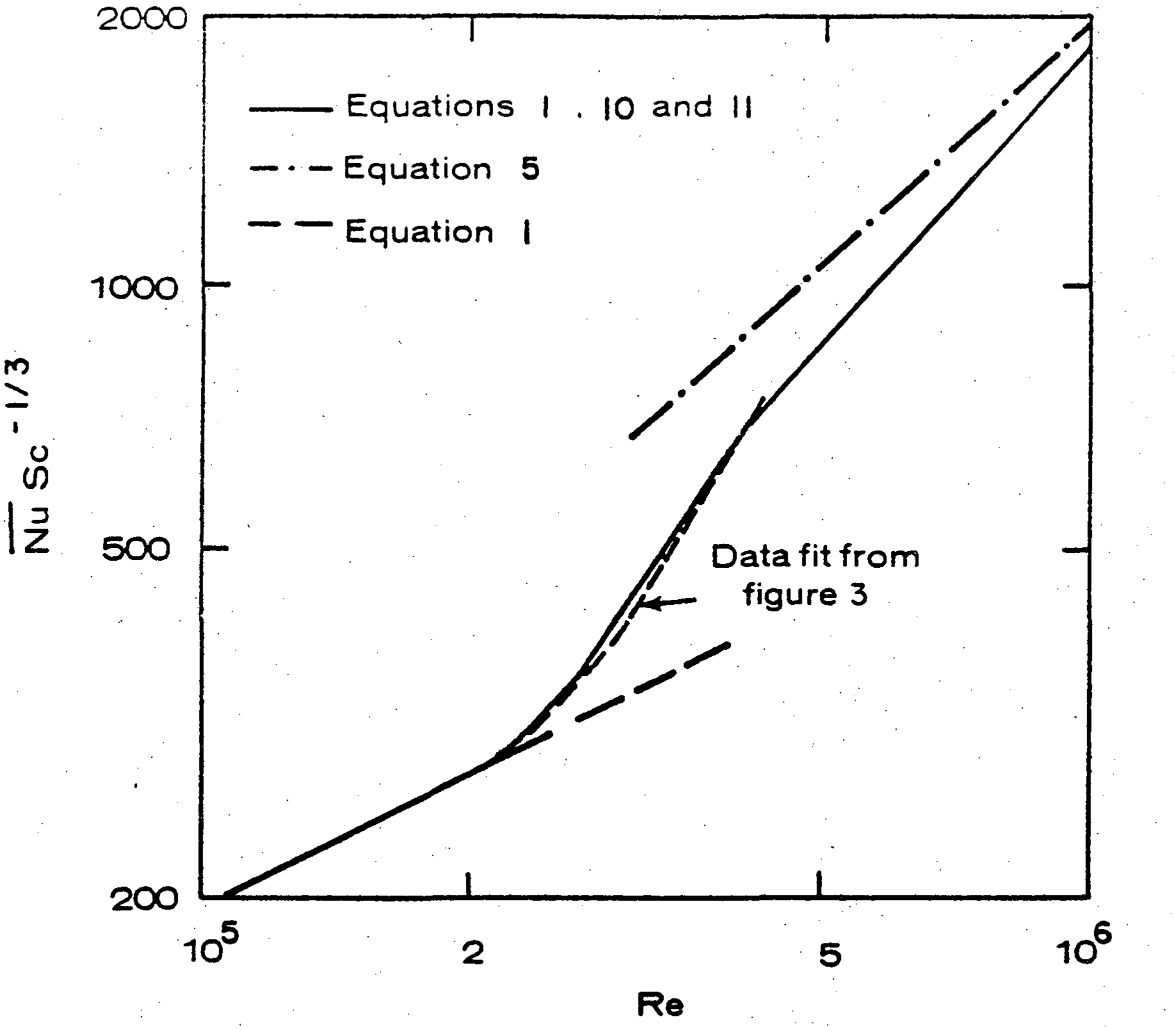

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Figure 6. Overall mass-transfer rate as predicted by correlations for the transition and turbulent flow regimes.

estimated local Nusselt numbers in the transition region differ from the experimentally determined values, shown on figure 5 by at most ten percent near $Re = 3.5\times10^5$; the fit is improved for larger Reynolds numbers.

Acknowledgment

This work was supported by the U.S. Energy Research and Development Administration.

List of Symbols

Δc	concentration difference between bulk and surface, mole/cm^3
D	diffusion coefficient of limiting solute, cm^2/sec
F	Faraday's constant, 96,487 C/equiv
$\bar{i}$	average current density, A/cm^2
n	number of electrons transferred per ion or molecule reacting
Nu_{loc}	local Nusselt number
$\overline{Nu}$	average Nusselt number
r	distance from axis of rotation, cm
r_o	electrode radius
Re	Reynolds number
Sc	Schmidt number
T	absolute temperature, deg K
μ	viscosity of solution, g/cm-sec
ν	kinematic viscosity of solution, cm^2/sec
Ω	rotation speed, sec^{-1}

References

1. B. Levich, "The Theory of Concentration Polarization," Acta Physicochimica U.R.S.S., 17, 257-307 (1942).
2. T. von Karman, "Über laminare und turbulente Reibung," Zeitschrift für angewandte Mathematik und Mechanik, 1, 233-252 (1921).
3. F. L. LaQue, "Theoretical Studies and Laboratory Techniques in Sea Water Corrosion Testing Evaluation," Corrosion, 13, 303 (1957).
4. N. Vahdat and J. Newman, "Corrosion of an Iron Rotating Disk," Journal of the Electrochemical Society, 120, 1682 (1973).
5. N. Vahdat, "Corrosion of an Iron Rotating Disk," M.S. Thesis, LBL-873, University of California, Berkeley (1972).
6. I. Cornet and U. Kaloo, "Temperature and Velocity Effects on the Cathodic Protection of a Steel Disk Rotating in Sea Water," Third International Congress on Metallic Corrosion, Extended Abstracts of Papers Presented, 213 (1966).
7. B. T. Ellison and I. Cornet, "Mass Transfer to a Rotating Disk," Journal of the Electrochemical Society, 118, 68 (1971).
8. B. T. Ellison, "Mass Transfer to a Rotating Disk," PhD. Disseration, University of California, Berkeley (1969).
9. M. Daguenet, "Etude du Transport de Matiere en Solution a l'Aide des Electrode a Disque et a Anneau Tournant," International Journal of Heat and Mass Transfer, 11, 1581 (1968).
10. C. L. Tien and D. T. Campbell, "Heat and Mass Transfer from Rotating Cones," Journal of Fluid Mechanics, 17, 105 (1963).

11. F. Kreith, J. H. Taylor, and J. P. Chong, "Heat and Mass Transfer from a Rotating Disk," Transactions of the American Society of Mechanical Engineers, Journal of Heat Transfer, 81, 95 (1959).

12. E. C. Cobb and O. A. Saunders, "Heat Transfer from a Rotating Disk," Proceedings of the Royal Society of London, A236, 343 (1956).

13. L. F. Donovan, O. T. Hanna, and S. Yerazunis, "Similar Solutions or Turbulent Boundary Layer Heat and Mass Transfer Problems," Chemical Engineering Science, 22, 595 (1967).

14. D. W. Hubbard, PhD. Dissertation, University of Wisconsin, Madison (1964).

15. D. W. Hubbard and E. N. Lightfoot, "Correlation of Heat and Mass Transfer Data for High Schmidt and Reynolds Numbers," Industrial and Engineering Chemistry Fundamentals, 5, 370 (1966).

16. W. Vielstich, A. Inocencio, and J. Heitbaum, Berichte der Bunsen Gesellschaft fur Physikalische Chemie, 78, 1252 (1974).

17. V. Levich, Physicochemical Hydrodynamics, Prentice-Hall, Inc., Englewood Cliffs, N.J. (1962).

18. N. Gregory, J. T. Stuart, and W. S. Walker, "On the Stability of Three-Dimensional Boundary Layers with Application to the Flow due to a Rotating Disk," Philosophical Transactions of the Royal Society of London, Mathematics and Physical Sciences, A248, 155 (1956).

19. D. T. Chin and M. Litt, "An Electrochemical Study of Flow Instability on a Rotating Disk," Journal of Fluid Mechanics, 54, 613 (1972).

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