Transport Accompanied by Chemical Reaction in Stagnation Flow

There are many industrially important reactions in which the chemical kinetics and external diffusion govern the observed reaction rate, and the observed kinetics differ drastically from the true kinetics. This study shows that the forward stagnation flow region of a circular cylinder can be a convenient system for studying the true kinetics of a solid catalyzed reaction owing to its unique property of uniform accessibility.

K. S. BALARAMAN
R. A. MASHELKAR
and
L. K. DORAISWAMY
National Chemical Laboratory
Pune - 411 008, India

SCOPE

In the experimental measurement of kinetics of fast heterogeneous reactions, it has been observed that mass transfer to the reactive surface determines the rate, once the rate becomes sufficiently high. The rates of most of these surface reactions per unit surface area of catalyst have not been accurately determined owing to the masking of the surface processes by diffusion, and the researchers were forced to operate in the apparently kinetically controlled regime. In this work, attention has been focused on the important interaction between the hydrodynamics, mass transport and heterogeneous catalytic chemical reaction in a well-defined flow field. The kinetics of a diffusionally limited heterogeneous catalytic reaction has been investigated in the stagnation region of a circular cylinder. This geometry was chosen because the entire stagnation region is equally accessible to the diffusing reactants. The concentration profile and the diffusional fluxes then become independent of the position, assuming that the catalyst is equally active at all points.

CONCLUSIONS AND SIGNIFICANCE

Our investigation would appear to be the first attempt wherein the uniform accessibility quality in the stagnation region of a circular cylinder has been used in an experimental study of transport limited heterogeneous catalysis.

Mass transfer experiments have been conducted in a carefully designed stagnation flow setup. Naphthalene vaporization was used as the experimental system. The experimental data agreed very well with the theoretical calculations.

The reaction between hydrogen and oxygen catalyzed by platinum was studied over the temperature range 295° to 333°K and at feed concentrations of hydrogen in the range of 0 to 3%. The reaction was influenced by both diffusion and reaction. The kinetic parameters were estimated from the additivity relation developed. The reaction was found to be first order with respect to hydrogen, and the true activation energy obtained in the study was 48.06×10^3 kJ$(\text{kmole})^{-1}$ over the temperature range investigated.

The characteristic reaction parameters defined by Rosner (1964) were also calculated. The experimental values agreed well with the characteristic plots of Rosner. The present study indicates that stagnation flow region can be used as a useful experimental tool to investigate kinetics of heterogeneous reactions.

BACKGROUND

The use of flow systems in kinetic studies of heterogeneous systems especially for reactions too fast to study by conventional means is well known. These flow systems take diverse forms, but for all practical purposes they could be classified into two categories, depending on whether the chemically active surface is moving or at rest. In the first category, the solid under investigation may be, for example, mechanically rotated. In the second category, we have certain steady flow reactors where the reactant fluid flows past or through the system. It is important to understand the role of fluid mechanics in such systems, since in the absence of correct convective transport information it is not possible to discern the true kinetics.

Two possible ways of discerning true kinetics could be conceived. In the first case, the assessment of diffusional effects can be made by first studying the reaction in a diffusion controlled region, and thereafter the data collected in the intermediate region could be corrected for diffusional effects. The second alternative is to choose a class of reactors, where the convective transport situation is well understood theoretically. The first alternative is time consuming and complex, whereas the latter has a considerable merit. The choice of a simple geometry, wherein the convective transport phenomenon is well understood theoretically, is particularly attractive.

Certain criteria should be applied for the choice of such simple geometries. Ease of experimentation is decidedly an important criterion. However, even more important is the desirability of having a geometry with spatially uniform transport coefficients, that is, a surface which is uniformly accessible. Indeed, in flow systems, nonuniform accessibility is the rule. This is typified by the flow past a flat plate (external flow) or the flow in a tube (internal flow), where the transport coefficient changes in the flow direction. Such variation of transport coefficients implies that a catalyst of uniform activity will usually establish a nonuniform reactant concentration distribution and thereby change the driving force for diffusion. In such cases, the transport problem is coupled with the chemical kinetic problem.

Correspondence concerning this paper should be addressed to R. A. Mashelkar.

0001-1541-80-3901-0635-$00.95.

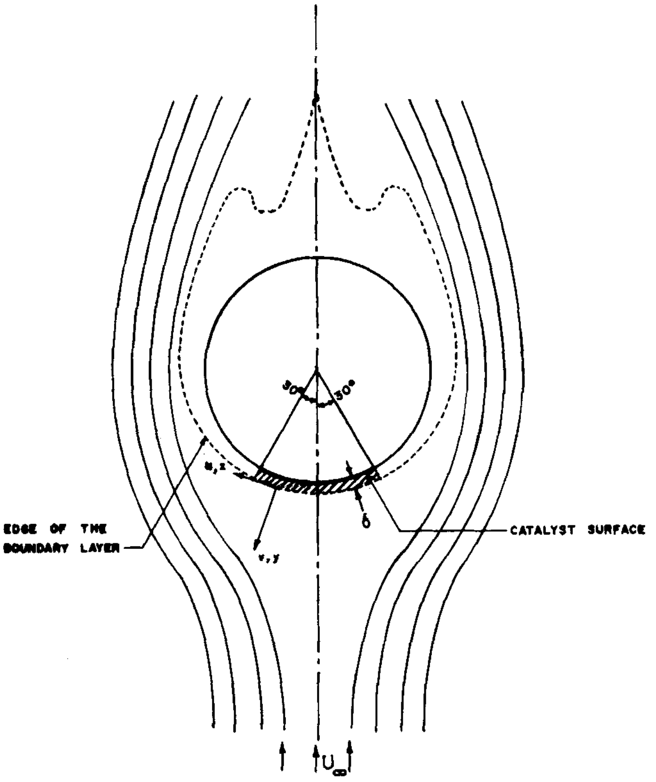

Figure 1. Hydrodynamic model of a circular cylinder showing the forward stagnation region in a flowing stream of reactants.

There appear to be only two hydrodynamic situations where the convective transport is uniform over the entire surface. The first is the laminar boundary layer flow around a rotating disk. The second is the so called stagnation flow. The flow around the forward stagnation point of a cylinder belongs to the latter category.

The flow around a rotating disk for the study of kinetics has received some attention in the past. Serad (1964) presented an experimental and theoretical study for mass transfer with and without chemical reaction on a rotating disk in laminar flow. He developed a model for a rapid second-order irreversible chemical reaction. Heymach (1969) investigated the kinetics of the hydrogenation of α-methyl styrene on both platinum and palladium catalysts deposited as thin films on a disk which rotated in liquid hydrocarbon. White (1972) conducted an experimental study of diffusion limited heterogeneous catalytic reactions on a rotating disk. He investigated the kinetics of the hydrogenation of α-methyl styrene to cumene over palladium and hydrogenation of phenyl acetylene to styrene and subsequently of styrene to ethyl-benzene. Recently, the kinetics of hydrogen peroxide decomposition has been reported on a modified disk by Joksimovic-Tjapkin and Delic (1973).

The unique uniform accessibility property of a stagnation flow has not at all been explored in the past. In the present work, we will present a theoretical analysis and also conduct an experimental investigation of the problem of convective diffusion with and without chemical reaction in this system.

THEORETICAL ANALYSIS

Schlichting (1968) has given an elaborate theoretical treatment of the problem of momentum and heat transfer around a cylinder, and therefore it will not be repeated. However, it will be advantageous to bring out certain salient features of this problem so as to enable us to define the limits in which the assumption of stagnation flow past a cylinder could be made.

Consider the flow past a circular cylinder of radius R_1 and free stream velocity U_∞ (Figure 1). The ideal velocity distribution in potential irrotational flow parallel to the x axis is given by (Schlichting, 1968).

$$U = 2\,U_\infty \sin\left(\frac{x}{R_1}\right) \tag{1}$$

It can be readily seen that for $x/R_1 << 1$, $\sin(x/R_1) = x/R_1$. Therefore, the potential velocity distribution in the neighborhood of the stagnation point of a circular cylinder $x/R_1 \simeq 0$ becomes

$$U = \frac{4U_\infty}{D}x = ax \tag{1a}$$

The governing equations of continuity, conservation of momentum and species for a fluid of uniform properties flowing in two-dimensional steady laminar boundary layer are as follows (see Schlichting, 1968):

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0 \tag{2}$$

$$u\frac{\partial u}{\partial x} + v\frac{\partial u}{\partial y} = -\frac{1}{\rho}\frac{\partial p}{\partial x} + \frac{\partial^2 u}{\partial y^2} \tag{3}$$

$$u\frac{\partial C}{\partial x} + v\frac{\partial C}{\partial y} = D_c\frac{\partial^2 C}{\partial y^2} \tag{4}$$

The boundary conditions are

$$u = v = 0,\ C = C_1,\ y = 0 \tag{5}$$

$$u = U,\ C = C_0,\ y \to \infty \tag{6}$$

By introducing nondimensional parameters

$$\eta = y\sqrt{\frac{U}{\nu x}}\ ;\ u = U\frac{\partial f}{\partial \eta} = Uf'\ ;$$

$$\theta = \frac{C - C_1}{C_0 - C_1};\ Sc = \nu/D_c$$

we obtain

$$f''' + ff'' + (1 - f'^2) = 0 \tag{7}$$

and

$$\theta'' + Sc\, f\theta' = 0 \tag{8}$$

The boundary conditions become

$$\eta = 0,\ f = f'' = \theta = 0 \tag{9}$$

$$\eta \to \infty,\ f' = \theta = 1 \tag{10}$$

Equations (7) and (8) could be solved to obtain the velocity and concentration distribution in the boundary layer. The quantity of interest is the local Sherwood number Sh_x. In the neighborhood of the stagnation point it can be shown that (see Schlichting, 1968)

$$\frac{Sh_x}{(Re_x)^{1/2}} = F(Sc) = \left[\int_0^\infty \exp\left\{-Sc\int_0^\eta f(\eta)\right\}d\eta\right]^{-1} \tag{11}$$

Further, it can be easily seen that since $U = 4\,U_\infty/D\,x$, we get

$$\frac{Sh_x}{(Re_x)^{1/2}} = \frac{k_g x}{D_c}\sqrt{\frac{D\nu}{4\,U_\infty x^2}} = \frac{1}{2}\frac{k_g D}{D_c}\sqrt{\frac{\nu}{U_\infty D}} = \frac{1}{2}\frac{Sh_D}{(Re_D)^{1/2}} \tag{12}$$

Therefore

$$\frac{1}{2}\frac{Sh_D}{(Re_D)^{1/2}} = \int_0^\infty \left[\exp\left\{-Sc\int_0^\eta f(\eta)\,d\eta\right\}\right]^{-1} = F(Sc) \tag{13}$$

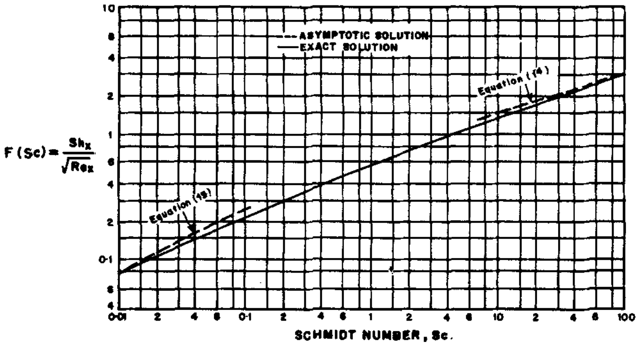

Figure 2. Local Sherwood number as a function of Schmidt number for stagnation flow.

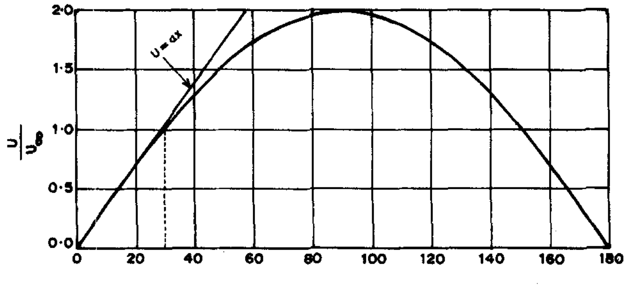

Figure 3. Potential velocity distribution past a circular cylinder.

The function $F(Sc)$ is shown in Figure 2. Approximate analytical solutions for the asymptotic cases of high ($Sc \rightarrow \infty$) and low ($Sc \rightarrow 0$) Schmidt numbers for mass transfer to the stagnation region can be analytically derived. The Sherwood number based on the diameter (Sh_D) is given by

$$Sh_D = 1.322\,(Re_D)^{1/2}\,(Sc)^{1/3} \text{ for } Sc \rightarrow \infty \tag{14}$$

and

$$Sh_D = 1.596\,(Re_D)^{1/2}\,(Sc)^{1/2} \text{ for } Sc \rightarrow 0 \tag{15}$$

The approximate solutions are also shown in Figure 2. They agree favorably with the exact solutions reported.

It should be emphasized that the calculations in the foregoing pertain to the stagnation flow only. The exact limits in which the stagnation flow assumption for flow past a circular cylinder becomes applicable should be discussed. For this, the validity of the potential flow approximation ($U = ax$) and also the consistency of the momentum boundary layer thickness could be taken as criteria. The potential velocity distribution as well as the displacement thickness are helpful for this purpose (see Figures 3 and 4). The displacement thickness is defined as $\delta_1 = \int_{y=0}^{\infty} [1 - u/U]\, dy$. The displacement thickness physically represents the distance by which the external streamlines are shifted owing to the formation of the boundary layer. It is clear that for ϕ lying between 0 to 30 deg., the stagnation flow approximation is a reasonable approximation. This defines the limit for our experimental studies.

Solution of the Convective Diffusion Equation with Chemical Reaction in the Stagnation Region of a Circular Cylinder

Let us consider the steady state flow of a fluid over an impermeable catalyst as shown in Figure 1. The governing conservation equations in this case remain the same as Equations (2) to (6). However, the boundary conditions now change to

$$\text{at } y = 0,\ u = v = 0,\ -D_c\left(\frac{\partial C}{\partial y}\right) = k_s C_1^n \tag{16}$$

$$\text{as } y \rightarrow \infty,\ u = U,\ C = C_0 \tag{17}$$

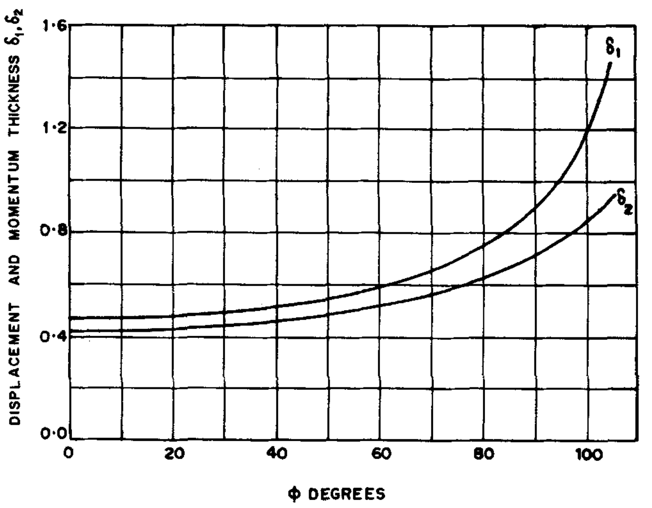

Figure 4. Variation of displacement thickness and momentum thickness with position around the cylinder.

In the forward stagnation region, the mass transfer coefficient is uniform over the surface. This reduces Equation (4) to

$$v_y(y)\,\frac{\partial C}{\partial y} = D_c\left(\frac{\partial^2 C}{\partial y^2}\right) \tag{18}$$

Boundary condition (16) implies that a reactant concentration C_1 is established at each point on the surface such that the local diffusional rate is equal to the local rate of chemical reaction which is exclusively a function of local surface concentration.

Integrating Equation (18) and applying the boundary conditions (16) and (17), we get

$$C_1^n + \frac{D_c}{k_s\delta} C_1 - \frac{D_c C_0}{k_s\delta} = 0 \tag{19}$$

where δ is the concentration boundary layer thickness given by (see Levich 1962)

$$\delta = \left[\int_0^\infty \exp\left\{\frac{1}{D_c}\int_0^\lambda v_y'\,dy'\right\} d\lambda\right]$$

For the general-order case, a mathematical solution has been presented by Levich (1962). For a first-order reaction ($n=1$), Equation (19) reduces to

$$C_1 = \frac{D_c C_0}{k_s\delta + D_c} = \frac{C_0}{k_s\dfrac{\delta}{D_c} + 1} \tag{20}$$

or

$$C_1 = \frac{C_0}{\dfrac{k_s}{k_g} + 1} = \frac{k_g C_0}{k_s + k_g} \tag{21}$$

where k_g is the mass transfer coefficient equal to D_c/δ.

Rate of reaction $\bar{R}$ can be expressed as

$$\bar{R} = k_s C_1 = \frac{k_s k_g}{k_s + k_g} C_0 = K\,C_0 \tag{22}$$

where K is the overall or apparent reaction rate constant given by

$$K = \frac{k_s k_g}{k_s + k_g}$$

or

$$\frac{1}{K} = \frac{1}{k_s} + \frac{1}{k_g} \tag{23}$$

Equation (23) represents additivity of two resistances: one for reaction and the other one for mass transfer. The diffusional and the kinetic terms are thus uncoupled.

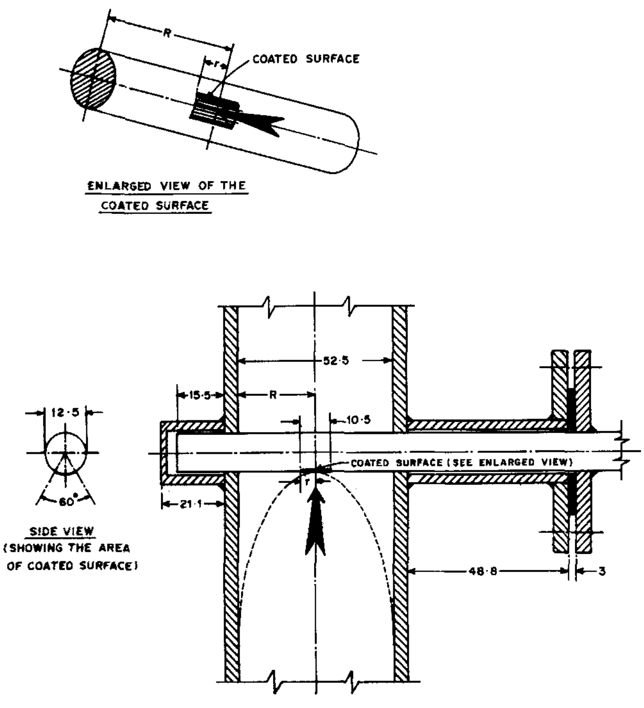

Figure 5. Test section (enlarged view) (all dimensions are in millimeters).

Correction for the Effect of Diffusion in Activation Energy Measurements

Following Rosner (1964), a global catalyst effectiveness factor $\bar{\eta}$ for external diffusion over the impermeable catalyst surface (similar to the effectiveness factor for pore diffusion) can be defined. This will provide a convenient and simple method of correcting the effect of diffusion in the kinetic studies of surface catalyzed reactions.

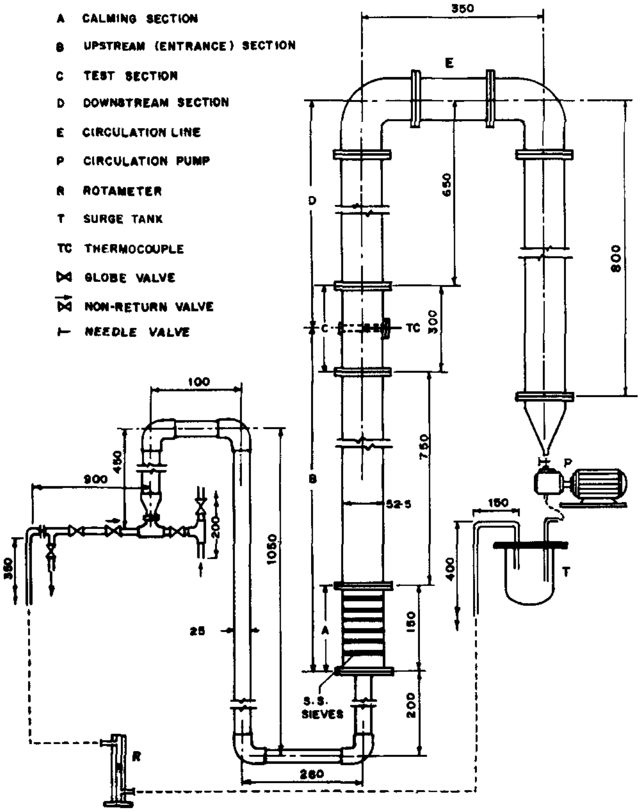

Figure 6. Experimental setup for kinetic studies.

$\bar{\eta}$ = integrated diffusion (effectiveness) factor

$$= \left(\frac{C_1}{C_0}\right)^n \tag{24}$$

$\bar{C}_n$ = catalytic parameter

$$= \frac{\text{characteristic rate of interfacial reaction}}{\text{characteristic rate of convective diffusion}}$$

or

$$\bar{C}_n = \frac{k_s}{k_g} \tag{25}$$

$\bar{\eta}$ is related to $\bar{C}_n$ by

$$\bar{\eta} = (1 + \bar{C}_n)^{-n} \tag{26}$$

and for the first-order reaction this reduces to

$$\bar{\eta} = (1 + \bar{C}_1)^{-1} = \left(1 + \frac{k_s}{k_g}\right)^{-1} = \frac{k_g}{k_s + k_g} \tag{27}$$

One can correct for the effect of convective diffusion in activation energy measurements only if the order of the reaction is known and if the value of k_g is determined. Such plots are helpful in experiments designed to obtain the value of the true activation energy E, particularly at elevated surface temperatures.

EXPERIMENTAL

Simulation of a Two Dimensional Stagnation Flow

The first problem encountered in this experimental investigation was the simulation of a two dimensional stagnation flow in the laboratory. The chief requirement was the generation of a uniform flow in which the cylinder could be placed. This problem was solved in the following manner.

It is known that under conditions of fully developed laminar flow, the steady state velocity distribution in a pipe of circular cross section is given as

$$u = 2\,\bar{U}\left[1 - \left(\frac{r}{R}\right)^2\right] \tag{28}$$

Let us examine the flow at the center, that is, $(r/R << 1)$. For $0 \le r/R \le 0.2$, the variation of the local velocity would be from $2\,\bar{U}$ to $1.92\,\bar{U}$, which is of the order of 4%. Indeed, if the values of the velocity in this region were to be averaged, then the mean velocity would be $1.96\,\bar{U}$, and the variation in the local velocity is of the order of 2%. Thus, if the exposed surface either for mass transfer or for the catalytic reaction were coated to this extent, then the resulting error due to nonuniformity of flow would be negligible.

From Equations (14) and (15), it can be seen that Sh_D is proportional to $Re_D{}^{1/2}$. Thus the mass transfer coefficient varies as the square root of the velocity. Therefore, a 2% variation in velocity in the radial direction would cause approximately 1% variation in the calculated Sherwood number. Furthermore, when mass transfer is accompanied by reaction [Equation (23)], the error will be even less. A schematic diagram of the test section is shown in Figure 5, where the actual experimental realization of stagnation flow has been clearly shown.

Apparatus

The apparatus built for this investigation comprised a cylindrical galvanized iron duct (inside diameter 5.25 cm) and had a total length of 1.85 m. A schematic diagram is shown in Figure 6. The calming section consisted of five screens made up of two 40 mesh screens followed by three 100 mesh screens. The purpose of the screens was to smoothen out the flow. The entrance section was 1.05 m in length (L = 20 D), and a further downstream distance of 0.8 m (L = 16 D) was kept. This ensured the development of a perfect laminar flow, which gave the assumed velocity distribution. The test section had a flange welded to which a cylinder (diameter = 1.25 cm) was fixed. The center of the cylinder was in perfect alignment with the center of the flange. Extreme care was taken to ensure that the stagnation point (ϕ = 0) was exactly at the axis of the cylindrical duct. Various sections were connected by mild steel flanges to facilitate easy dismantling.

TABLE 1. SUMMARY OF PREVIOUS KINETIC EXPERIMENTS

			Experimental details				
No.	Investigators	Type of reactor	Temperature range °K	Hydrogen concentration max. mole %	Type of catalyst	Order of reaction with respect to hydrogen	Activation energy kJ (k mole)$^{-1}$ × 10^{-3}
1	2	3	4	5	6	7	8
1.	Boroskov (1954)	Differential bed recycle reactor	373-453	1.0	Platinum 99.8% pure	First order	41.87 ± 8.37
2.	Gidaspow and Ellington (1964)	Vitreous alumina reactor tube	366-1143	3.0	Platinum coated on vitreous alumina tube	First order	71.17 (below 504°K)
3.	Kulacki and Gidaspow (1967)	Parallel plate duct with catalytic wall in laminar flow	348-638	1.5	Platinum coated on alumina plates	First order	40.19 (between 348°-473°K)
4.	Leder and Butt (1966)	Fixed bed reactor packed with catalyst	373-398	3.3	Platinum supported on alumina	Second order	41.87 (at 373°K)
5.	Gentry et al. (1974)	Coil type of reactor	298-473 and 318-373	1.0	Platinum in the form of coil (*Oxidized and reduced surfaces)	First order	64 ± 12 (between 331°-467°K) 83 ± 9 (between 318°-371°K)
6.	This work	Stagnation flow of a circular cylinder	295-333	3.0	Platinum coated on stainless steel cylinder	First order	48.06

Mass Transfer Experiments

The validity of the convective diffusion equation developed for the stagnation region was checked by undertaking studies on vaporization of naphthalene. Naphthalene vaporization has been extensively used in the literature for mass transfer studies (for example, see Goldstein et al., 1973; Honaker and Tao, 1970).

Chemically pure naphthalene was dissolved in rectified spirit and recrystallized. The recrystallized sample was then sublimed and very pure naphthalene free from other impurities was obtained. The purified naphthalene was filled up in a big test tube and slowly heated in a water bath. When a clear solution was obtained, the clean stainless steel cylinder was slowly dipped and taken out. A thin uniform coating of naphthalene was obtained on the cylinder. It was then carefully cut with a sharp knife, and the coating on specific parts of the cylinder was removed, leaving the required area (see Figure 5) in the stagnation region.

Air from the cylinder (dried over anhydrous magnesium perchlorate) was fed to the reactor through calibrated capillary flowmeters. The flow rate was adjusted. After a steady state was reached, the outlet of the duct was connected to a series of absorbers filled with spectroscopic grade rectified spirit. Air was blown for a known interval of time, and the rectified spirit was made up to known volume. The concentration of naphthalene was determined by measuring the absorbance in a UV spectrophotometer.

Studies on Mass Transfer with Catalytic Reaction

The catalytic reaction between hydrogen and oxygen was chosen as a model system for kinetic studies. The main reasons for choosing this system were as follows.

1. The reaction has been studied in the literature, and the detailed kinetics are available (see Table 1). Furthermore, the reaction between hydrogen and oxygen is known to be very fast, and in the region to be investigated here, calculations indicated that significant diffusional effects are likely to be present.
2. Platinum, which is widely used as catalyst for this reaction, is very active, and its deposition techniques on metallic surfaces are well known.
3. The concentration of hydrogen in the circulating reactants can be easily determined by a thermal conductivity detector by taking advantage of the large difference between the thermal conductivity of hydrogen and air.

Platinum was deposited by evaporated metal film technique by applying a vacuum of the order of 10^{-4} N/m^2. The stainless steel cylinder was first cleaned by trichloroethylene, followed by methanol and acetone in the ultrasonic cleaner. The cylinder was covered with aluminium foil, and only the required area was exposed. The question of catalyst activity is of prime consideration, and the surface activity was maintained by pretreating the platinum surface before every run in dry air for 1 hr at 423°K. The composition of hydrogen in air was determined by the specially built thermal conductivity detector.

The experimental setup for kinetic studies is shown in Figure 6. Air and hydrogen from the cylinder (dried over anhydrous magnesium perchlorate) were fed to the reactor through separate metering systems and mixed at the entrance to the reactor. The reactants were circulated by a diaphragm pump in conjunction with a surge tank. The gases were trapped in the circulation line by suitable adjustments of the valves shown in the diagram. The flow in the circulation line was adjusted by

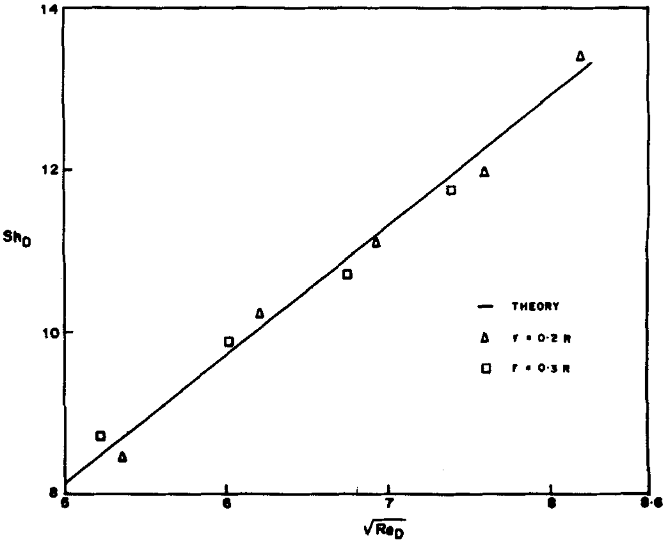

Figure 7. Mass transfer around the stagnation region, theory and experiments.

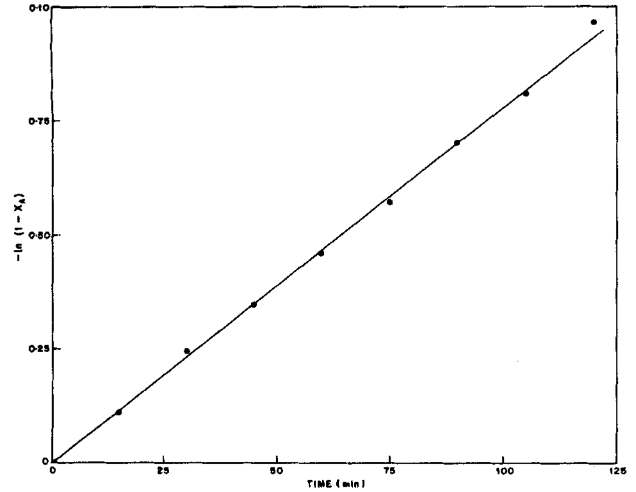

Figure 8. First-order representation of the reaction with respect to hydrogen.

means of a needle valve provided in the suction side of the pump, and the flow rate was accurately measured by means of a rotameter. The initial concentration of hydrogen in the circulation line was adjusted to around 3%. The temperature of the reaction was controlled by three separate heating sections and measured by copper constantan thermocouple inserted in the cylinder and located exactly above the stagnation region. When the flow and temperature became steady, samples were withdrawn from the circulation line at known intervals by a hypodermic gas tight syringe. In every case, 1.5 ml sample was used with air as the carrier gas. The mole percent of hydrogen present in the sample was indicated by the deflection (peak height) measured in the strip chart recorder. Prior to the commencement of every run, the absence of the homogeneous reaction between hydrogen and oxygen was confirmed by circulating the reactants without the catalyst at the same conditions of reaction.

RESULTS

Mass Transfer Experiments

Mass transfer experiments were carried out over the naphthalene coated surface by varying the Reynolds number around the cylinder. The rate of mass transfer was measured and compared with the theoretical predictions [Equation (13)]. The Sherwood number for mass transfer calculated theoretically and measured experimentally are plotted against the Reynolds number around the cylinder. The plot is shown in Figure 7. The deviation between the theory and experiments (even in the case of $r/R = 0.3$) were well within 5%, thus confirming the soundness of the experimental procedure.

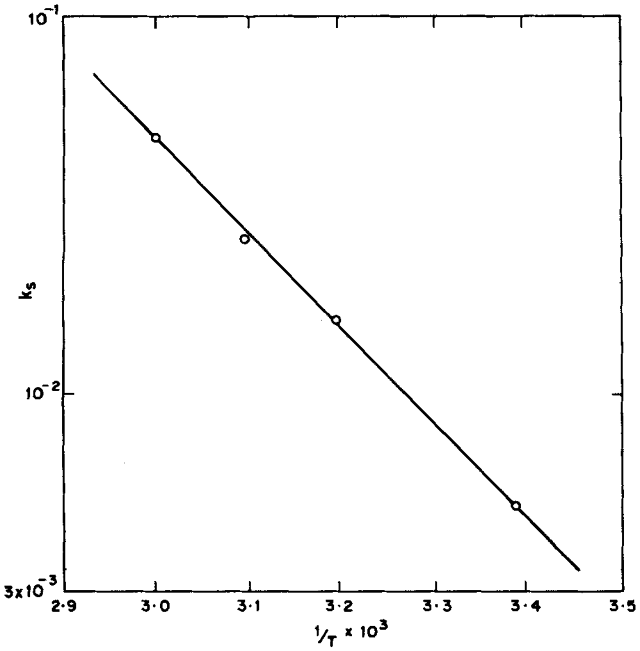

Figure 10. True reaction rate constant vs. temperature.

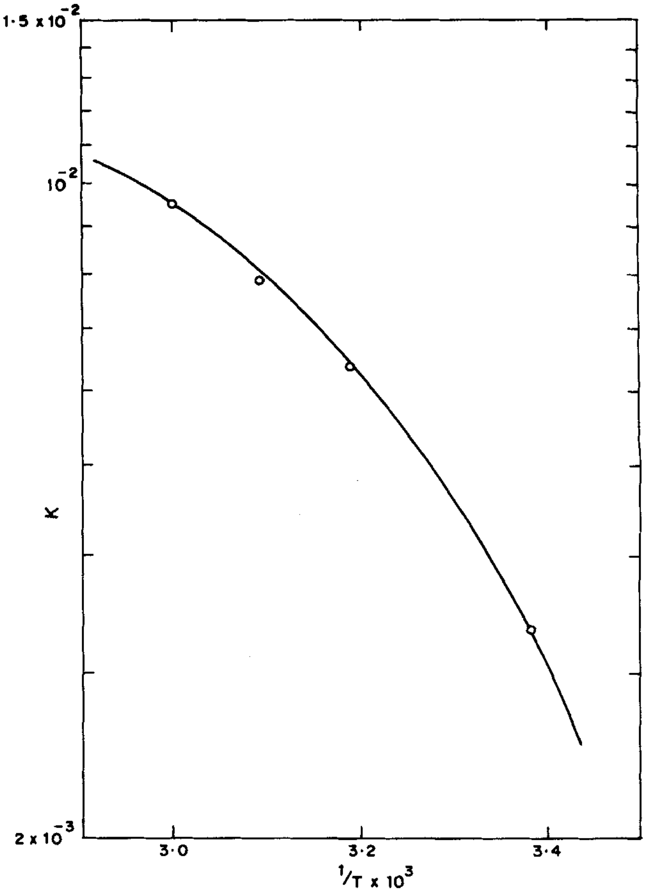

Figure 9. Apparent reaction rate constant vs. temperature.

Kinetic Experiments

The reaction between hydrogen and oxygen catalyzed by platinum was studied over the temperature range 295° to 333°K and feed concentration of hydrogen in the range 0 to 3% in air. The studies were confined to only the stagnation region (that is, $r/R = 0.2$ and extending to 30°ϕ on either side from the stagnation point). The concentration of hydrogen (and hence the fractional conversion X_A) in the circulating reactants was measured at known intervals, and a plot of $-\ln(1-X_A)$ vs. time was made. A typical plot is shown in Figure 8. It gave a straight line passing through the origin, indicating that the reaction was first order with respect to hydrogen. The slope of this plot gave the apparent or observed rate constant K. This was plotted against temperature as shown in Figure 9. The plot indicated that the reaction was affected by both diffusion and reaction. The slope at each point gave the apparent activation energy E_a. Since k_g was accurately determined by the mass transfer expression, the true or intrinsic reaction rate constant k_s was extracted by the additivity relation. Figure 10 shows the characteristic plot of the true reaction rate constant k_s vs. temperature. The shape of this plot gave the true activation energy E, and the intercept gave the Arrhenius parameter. The catalytic parameter $\overline{C}_n$ and the integrated diffusion factor $\overline{\eta}$ were calculated. The experimental values agreed well with the characteristic plots of Rosner (1964) as shown in Figure 11.

The above results of the kinetic experiments are compared with the reported experiments in the literature on the same system as shown in Table 1. The value of true activation energy is well within the range of reported values.

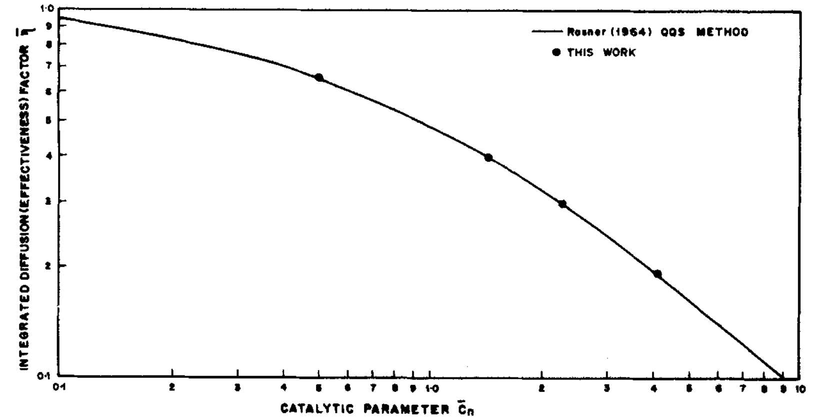

Figure 11. Dependence of integrated diffusion (effectiveness) factor on catalytic parameter.

The present study indicates that stagnation flow experiments can serve as a useful and powerful technique for the study of heterogeneously catalyzed reactions. Coating of a catalyst should be no limitation, since it is always conceivable to have downward flow of a gas past a cylinder, in the stagnation region of which is embedded an active catalyst by cutting out a groove. Although the case considered here refers to first-order kinetics, the technique can be used in the case of nonlinear kinetics also. The results presented by Levich (1962) would be useful for such cases.

The present study is helpful in defining the limits in which meaningful experiments on heterogeneously catalyzed reactions can be carried out in a stagnation flow setup.

NOTATION

C = concentration in fluid phase
C_1 = surface concentration of the fluid
$\underline{C}_0$ = bulk concentration of the fluid
$\bar{C}_n$ = catalytic parameter for n^{th}-order surface reaction, Equation (25)
D = diameter of the cylinder
D_c = diffusion coefficient
E = true activation energy
E_a = apparent activation energy
f = nondimensional parameter, Equation (7)
$F(Sc)$ = function of Schmidt number, Equation (13)
K = apparent or observed reaction rate constant
k_g = mass transfer coefficient
k_s = true or intrinsic reaction rate constant
r = radial distance from the center of the duct ($2r$ = length of the coated surface)
$\underline{R}$ = radius of the duct
$\bar{R}$ = rate of reaction
R_1 = radius of the cylinder
Re = Reynolds number
Sc = Schmidt number
Sh = Sherwood number
t = time
T = absolute temperature
u = velocity in the boundary layer in x direction
$\underline{U}$ = velocity in the potential flow [$= 2U_\infty \sin(x/R_1)$]
$\bar{U}$ = average velocity in the duct
U_∞ = velocity in the mainstream or approach velocity to the cylinder
v = velocity in the boundary layer in y direction
x = axial coordinate
X_A = fractional conversion of hydrogen
y = transverse coordinate

Greek Letters

δ = concentration boundary layer thickness
δ_1 = displacement thickness
δ_2 = momentum thickness
η = nondimensional parameter, Equation (9)
$\bar{\eta}$ = integrated diffusion effectiveness factor, Equation (24)
θ = dimensionless concentration, Equation (8)
ν = kinematic viscosity of the fluid
ϕ = angle around the cylinder measured from the stagnation region, deg.

Subscripts

D = based on diameter
x = local value based on characteristic length

LITERATURE CITED

Boreskov, G. K., "Activite Catalytique Des Metaux En Rapport Avec La Reaction D'oxydation De L'Hydrogene," *J. Chim. Phys.*, **51**, 759 (1954).

Gentry, S. J., J. G. Firth and A. Jones, "Catalytic oxidation of hydrogen over platinum," *J. Chem. Soc. Faraday Trans.*, **1**, 3, 600 (1974).

Gidaspow, D., and R. T. Ellington, "Surface Combustion of Hydrogen: Part 1. On Platinum Coated Alumina," *AIChE J.*, **10**, 707 (1964).

Goldstein, R. J., E. M. Sparrow and D. C. Jones, "Natural Convection Mass Transfer Adjacent to Horizontal Plates," *Int. J. Heat Mass Transfer*, **16**, 1025 (1973).

Heymach, G. J., "Study of the Kinetics of Diffusionally limited Heterogeneous Reactions on a Rotating Disc," Ph.D. dissertation, Univ. Pa. (1969).

Honaker, D. E., and L. C. Tao, "Effect of Sonic Pulsations on Mass Transfer Rate of Naphthalene from a Plate into Air," *Ind. Eng. Chem. Fundamentals*, **9**, 325 (1970).

Joksimovic-Tjapkin, S. M., and D. Delic, "Kinetics of Concentrated Hydrogen Peroxide Decomposition on a Rotating Disc," *ibid.*, **12**, 33 (1973).

Kulacki, F. A., and D. Gidaspow, "Convective Diffusion in a Parallel Plate Duct with One Catalytic Wall-Laminar Flow—First Order Reaction, Part II: Experimental," *Can. J. Chem. Eng.*, **45**, 72 (1967).

Leder, F., and J. B. Butt, "Surface Catalysis of the Hydrogen-Oxygen Reaction on Platinum at Low Temperatures," *AIChE J.*, **12**, 718 (1966).

Levich, V. G., *Physicochemical Hydrodynamics*, 2 ed., Prentice Hall, Englewood Cliffs, N.J. (1962).

Rosner, D. E., "Convective Diffusion as an Intruder in Kinetic Studies of Surface Catalyzed Reactions," *AIAA J.*, **2**, 593 (1964).

Schlichting, H., *Boundary Layer Theory*, 6 ed., McGraw Hill, New York (1968).

Serad, G., "Chemical Hydrodynamics with a Rotating Disc," Ph.D. dissertation, Univ. Pa. (1964).

White, D. E., "Diffusion-limited Heterogeneous Catalytic Reactions on a Rotating Disc," Ph.D. dissertation, Univ. Pa. (1972).

Manuscript received September 7, 1979; revision received February 12, and accepted February 29, 1980.