Diffusion and Flow in a Radially Moving Film

R. L. MERSON and J. A. QUINN

University of Illinois, Urbana, Illinois

Numerous experimental studies have been reported on the application of unsteady state diffusion theory to interphase transfer in flow systems. Interpretation of these studies calls for a detailed knowledge of the flow field in the immediate vicinity of the interface and an understanding of the dynamics of the interface itself. For most contacting devices the flow field is not amenable to analysis, and the diffusion process is often obscured by hydrodynamic factors. Three of these factors, acceleration of the interface at the point of formation, stagnant end effects, and the Marangoni effect, are explored in this study of mass transfer in binary systems with solute diffusing into a radially moving interface.

In all dynamic experiments, for example a laminar jet, a falling drop, a wetted-wall column, or the present experiment, the two phases are brought into contact at a point where the absolute interfacial velocity is zero. The interface then accelerates to a fully developed flow over some finite distance. The analysis of the fluid dynamics is complex, and the effect of this acceleration region on the rate of mass transfer has not been well established. The stagnant end effect, which occurs if the two phases are separated after some time of contact, is relatively less important because the mass flux at the exit is considerably smaller than at the inlet. Nevertheless, the stagnant end effect may still be important, especially if short times are involved or if a large portion of the interface is stagnant owing to the accumulation of surface-active impurities. The Marangoni effect, or interfacial turbulence, and the extent to which it enhances the mass transfer rate cannot be predicted at the present time. Moreover, in many types of apparatus it is difficult to tell when it occurs.

This work was undertaken to investigate the various effects mentioned above as they relate to transfer through the liquid-liquid interface. Aside from the extensive literature on liquid drops, few dynamic studies on the liquid-liquid interface have been reported. Ratcliff and Reid (*9*) studied the transfer of benzene into water with a spherical-film absorber. Quinn and Jeannin (*8*) and Ward and Quinn (*12*), using laminar liquid-liquid jets, have reported experiments on two-component liquid systems. The principal feature of the present experiment is that the interface is readily observable and that all pertinent variables, such as surface age, can be measured directly.

THE EXPERIMENT

In the binary systems studied, one phase was always water, and the upper diffusing phase was a less dense organic liquid. Since small traces of surface-active contaminants can drastically influence the experiment, special precautions were observed to keep the reagents and apparatus free from impurities. Water from the building supply was deionized and degassed, and the organic liquids were reagent grade. Pertinent physical properties used in the calculations are listed in Table 1.

The main feature of the contacting apparatus was a thin layer of water which flowed radially outward in a horizontal plane from a central source. For the first portion of its traverse the water was separated from the organic phase by a thin metal disk. At the edge of the disk the two phases joined, precisely defining the initial time of contact, and flowed to the wall of the chamber where they separated by gravity. The upper phase circulated internally by virtue of the viscous drag exerted by the water layer flowing beneath it.

The mass transfer chamber, shown in Figure 1, consisted of a precision bore glass cylinder held in compression between two stainless-steel flanges. A hollow stainless-steel rod was threaded into the upper flange and extended down into the glass cylinder. This shaft served as an inlet tube for the water and as a support for the water chamber which was a closed, hollow, stainless-steel cylinder, coaxial with the glass cylinder

TABLE 1. PROPERTIES OF THE SYSTEMS STUDIED

Upper phase	Solubility* in water $\cdot 10^5$ (g.-moles/cc.)	Diffusivity* in water $\cdot 10^6$ (sq. cm./sec.)	Viscosity* (centipoise)	Interfacial† tension (dynes/cm.)	Interface motion
Isobutanol	120.0 (*3*)	9.83 (*3*)	3.27 (*3*)	2.1 (*3*) 25°	moving
Ethyl acetate	84.3 (*3*)	9.09 (*3*)	0.45 (*3*)	6.4 (*3*)	moving
Octanol	0.38 (*4*)	—	8.93 (*5*)	8.5 (*5*)	moving
Methyl isobutyl ketone	18.7 (*2*)	—	0.58 (*5*) 20°	9.8 (*1*)	partially stagnant
n-Butyl acetate	4.3 (*4*)	—	0.73 (*4*) 30°	12.5 (*1*)	stagnant
Oleic acid	insol. (*4*)	—	25.6 (*4*)	15.6 (*5*)	stagnant
Benzene	2.23 (*12*)	11.0 (*12*)	0.61 (*4*)	35.0 (*5*)	stagnant
Carbon dioxide	3.39 (*4*)	19.7 (*10*)	—	—	stagnant
Air	—	—	—	72.8 (*5*)	stagnant

* At 25°C. unless otherwise indicated.
† At 20°C. unless otherwise indicated.

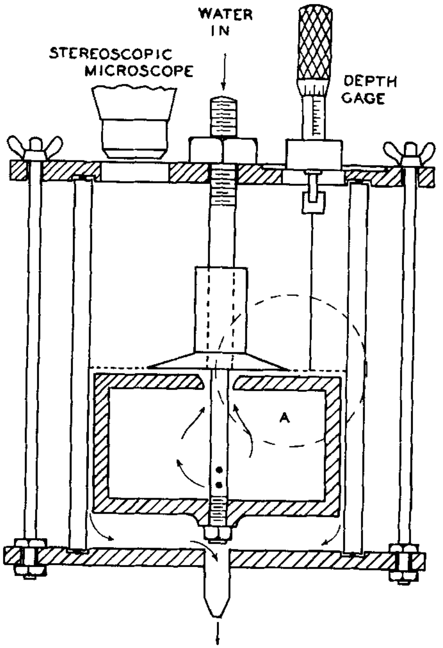

Fig. 1*a*. The mass transfer chamber.

leaving an annular space of 0.125 cm. between the steel and the glass. To maintain concentric symmetry within this annular ring, three spacers made from 18 gauge hypodermic needles were inserted into the ring near the bottom of the water chamber. Water flowed into the water chamber through holes drilled in the support rod, passed uniformly upward through an annular space formed by the rod and hole cut in the top end of the water chamber, and was deflected radially outward by the disk mounted on the support rod above the water chamber. The water then flowed as a thin sheet across the flat top of the water chamber, picking up organic solute by diffusion before moving down the annular space between the chamber and the glass wall and out a hole drilled in the bottom compression flange. The thickness of the water sheet was regulated by adjusting the height of the effluent line.

The disk was mounted on a sleeve which could be raised or lowered above the surface of the water chamber, thus controlling the thickness of the water layer at the tip of the disk. To measure the thickness at other radial positions in the region of mass transfer, a depth gauge was constructed consisting of a thin stainless-steel needle mounted on a micrometer head. As the micrometer head was turned, the needle traveled downward through the organic phase until it just met its image in the interface, and the thickness relative to the top of the water chamber was read from the micrometer. The needle was mounted on a track so that a thickness profile, such as that shown for the benzene-water system in Figure 2, could be obtained across the entire flow path. For the liquid-liquid systems, treatment of the entire surface of the glass with an insoluble polymeric silicone coating (General Electric Dri-Film SC-87) produced a downward meniscus which was undesirable. To produce a planar surface, only the portion of glass above the desired interface level was coated. Figure 2 shows the elimination of the large natural benzene meniscus, for a film of water set for 1 mm. thickness at the disk, by coating the glass wall above 1 mm.

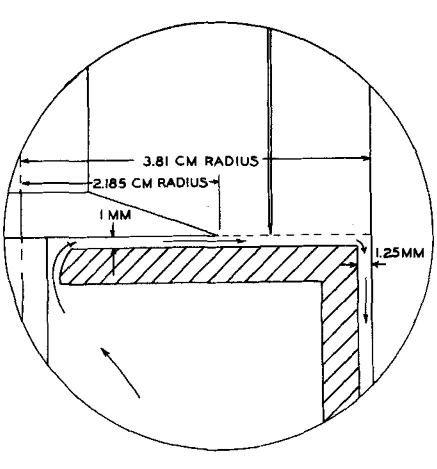

Fig. 1*b*. Enlarged view at *A* showing water flow path.

Runs were also made with water-saturated carbon dioxide as the upper phase. For the carbon dioxide system, gas flow from the supply cylinder was controlled satisfactorily by two diaphragm valves in series in conjunction with a surge tank equipped with a carbon dioxide bleed line vented to the atmosphere. This provided a constant pressure source of carbon dioxide which was connected to the gas-tight mass transfer chamber through a capillary-tube flow meter. Thus gas flowed into the chamber at the rate at which it was absorbed by the water, just as the organic phase was siphoned into the chamber at the same rate it was absorbed.

Procedure

Before mass transfer measurements were made, the disk spacing was set for the desired thickness of the water layer, glass parts were cleaned in hot chromic acid cleaning solution, and metal parts were washed in organic solvents and rinsed with water. At a given water flow rate and disk spacing the experimental problem was to establish and maintain a water layer of the desired thickness δ with a flat interface. By making fine adjustments on the water outlet level the interface could be maintained for several hours to within 0.05 mm. Other experimental variables were the water flow rate, measured by collecting the effluent in a graduated cylinder for a timed interval, and the temperature of the water, measured at the inlet and outlet of the chamber. The majority of the experimental runs were carried out in a temperature range of 22° to 26°C. (7).

At the beginning of a run the interface was swept clean of possible contaminants by gently sucking up liquid from both phases very near the interface. This was done for several minutes until all of the talcum powder which previously had been spread on the surface had been swept up. Steady conditions were then rapidly established. At this point, fine particles of talc, previously ignited to remove organic impurities, were sprinkled lightly on the upper surface of the organic phase. The talc fell through the lighter organic phase, providing a striking manifestation of the flow in this region, but it was not wetted by the water and thus floated in the interface with the velocity of the interface. This motion was observed from directly above through a three-dimensional microscope of 10x magnification. About forty individual particles were timed with a stop watch as they traveled from the tip of the disk out to various radial positions on a calibrated scale in the eyepiece of the microscope. This procedure gave an unequivocal experimental value for the time of contact of the two phases.

Simultaneous to the talc measurements, samples of the organic solutions were taken periodically at the water outlet in tightly sealed dram vials, filled full to prevent loss of the organic by evaporation into the gas space. The samples were analyzed for organic content on a gas chromatograph equipped with a hydrogen flame detector. For the carbon dioxide system, the gas consumption rate was observed directly with the capillary-tube flow meter, and representative samples were titrated for carbon dioxide content. These samples were carefully transferred with a syringe from the outlet line to a solution of

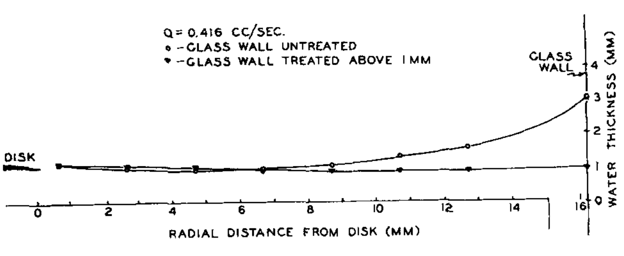

Fig. 2. Water thickness profile for the benzene-water system showing elimination of the natural meniscus.

sodium hydroxide and barium chloride under an atmosphere of nitrogen. In addition, the outlet concentration of carbon dioxide was continuously monitored with a flow-through conductivity cell mounted in the outlet line.

ANALYSIS

For the relatively dilute solutions employed in the experiment the equation of continuity for the solute may be expressed as

$$\frac{\partial C}{\partial t} + (V \cdot \nabla)\, C = D \nabla^2 C \qquad (1)$$

with the diffusivity assumed constant. When one neglects radial diffusion and considers that the flow is directed radially with no component normal to the interface, Equation (1) reduces to

$$u \frac{\partial C}{\partial r} = D \frac{\partial^2 C}{\partial y^2} \qquad (2)$$

at steady state. For small contact times, the diffusing solute will not penetrate far from the interface, and the velocity in Equation (2) can be replaced by the interface velocity u_i, a function of r alone. With the surface age defined as

$$\tau = \int_{r_o}^{r} \frac{dr}{u_i} \qquad (3)$$

Equation (2) becomes

$$\frac{\partial C}{\partial \tau} = D \frac{\partial^2 C}{\partial y^2} \qquad (4)$$

On the assumption of interfacial equilibrium and a film thickness large compared with the depth of penetration, appropriate boundary conditions are

$$C(0, \tau) = C^*, \; C(y, 0) = C(\infty, \tau) = 0 \qquad (5)$$

The familiar solution to Equations (4) and (5) yields the local mass transfer rate

$$N = -D \frac{\partial C}{\partial y}\bigg|_{y=0} = C^* \sqrt{\frac{D}{\pi \tau}} \qquad (6)$$

which can be integrated over the area of transfer to give the total mass transfer rate

$$\phi = \int_{r_o}^{r_1} 2\pi r N dr = \overline{C} Q \qquad (7)$$

Evaluation of the integral in Equation (7) requires an expression for the surface age. The surface age can be calculated from an analysis of the fluid dynamics of the film, or it can be measured directly as described in the previous section.

A complete solution to the flow problem properly allowing for acceleration at r_o and for the effect of the wall at r_1 is, obviously, prohibitively complex. However, a first approximation to the surface velocity can be obtained by assuming a one-dimensional creeping flow in the film and in the organic layer above the film. These assumptions yield the velocity profiles

$$U_w \equiv \frac{u_w}{\overline{u}_w} = a_w + b_w Y + c_w Y^2, \quad 0 \leqq Y \leqq 1 \qquad (8)$$

$$U_o \equiv \frac{u_o}{\overline{u}_w} = a_o + b_o Y + c_o Y^2, \quad 1 \leqq Y \leqq \frac{H}{\delta} \qquad (9)$$

Here the dimensionless velocity U is based on the average velocity of the water phase, $\overline{u}_w$, and the dimensionless coordinate Y is based on the thickness of the water layer δ. The six arbitrary constants can be eliminated with the following boundary conditions:

at

$$Y = 0, \; U_w = 0$$

$$Y = 1, \; U_w = U_o, \; \mu_w \frac{dU_w}{dY} = \mu_o \frac{dU_o}{dY}$$

$$Y = \frac{H}{\delta}, \; U_o = 0 \text{ or } \frac{dU_o}{dY} = 0,$$

the average velocity for the internally circulating organic phase is zero

$$\overline{u}_o = 0$$

and the average velocity in the water phase is known from the volumetric flow rate

$$\overline{u}_w = Q/2\pi r \delta$$

For the case in which the upper surface is free, the latter of the conditions at $Y = H/\delta$, the interfacial velocity is given by

$$u_i = \beta \overline{u}_w \qquad (10)$$

where

$$\beta = \frac{3}{2} \frac{\left(\frac{H}{\delta} - 1\right)}{\left(\frac{H}{\delta} - 1\right) + \frac{3}{4} \frac{\mu_o}{\mu_w}}$$

The surface age is then given by

$$\tau = \frac{\pi \delta \,(r_1^2 - r_o^2)}{\beta Q} \qquad (11)$$

Substituting Equation (11) into Equation (6) and integrating over the mass transfer surface one gets

$$\phi = 2C^* \sqrt{\frac{D \beta Q \,(r_1^2 - r_o^2)}{\delta}} \qquad (12)$$

Stagnant Interface

The development above applies to a freely moving interface. Suppose, however, that there is present in the system a surface-active substance whose sole effect on the mass transfer process is to eliminate motion in the interface. That is the effect is purely hydrodynamic, and the surface-active material offers no resistance to diffusion through the interface. For this case, the surface age has no meaning, but the mass transfer can be estimated directly from Equation (2).

An asymptotic solution valid for small depths of penetration is obtained by linearizing the velocity profile of the stagnant case. The diffusion equation then becomes

$$\frac{3Q}{\pi \delta^2} \left(\frac{y}{r}\right) \frac{\partial C}{\partial r} = D \frac{\partial^2 C}{\partial y^2} \qquad (13)$$

with the result that

$$\phi = \frac{C^*}{\Gamma\left(\frac{4}{3}\right)} \left\{ \frac{3 D \sqrt{Q}}{2\pi \delta} \right\}^{2/3} [r_1^{4/3} - r_o^{4/3}] \qquad (14)$$

for the stagnant interface.

RESULTS AND OBSERVATIONS

A series of runs was made with isobutanol as the organic phase to check the consistency of the method. Experiments were carried out at four water film thicknesses between 0.26 and 2.0 mm. over a range of the volumetric flow rate from 4 to 380 cc./min. giving a theoretical range of surface age, from Equation (11), between 0.3 and 34 sec. Total mass transfer rates were measured as the product of the volumetric flow rate and the average concentration. The results obtained were lower than those predicted from Equation (12) with the theoretical approximation of the true surface age. The agreement improved as the thickness of water film was decreased, suggesting that these conditions more closely met the assumptions of Equation (12). In addition, runs with a larger disk, which decreased the interfacial area available for mass transfer and intensified end effects, showed a marked increase in the deviation from the approximate theory.

On the other hand, internal agreement was excellent. Identical experiments with isobutanol and ethyl acetate gave equivalent mass transfer rates as shown in Figure 3. Here a grouping of physicochemical factors is plotted as a function of the hydrodynamic variables as suggested by Equation (12); essentially Figure 3 is a plot of molar flux vs. water flow rate. A single line can be drawn through the data of both systems, but the line has a slope only 60% of that predicted from the approximate theory. This disagreement can be explained by using the true surface age in place of Equation (11) in predicting the transfer rate.

Typical results of the talc measurements are presented in Figure 4 as a plot of the surface age vs. the square of the radial distance from the center of the chamber. These data are for the isobutanol-water system. Each point on the plot represents the time required for a talc particle to float from the disk out to a given radial position. The dashed line represents the ideal surface age calculated from Equation (11), which assumes fully developed flow. The deviation from linearity of the curves defined by the experimental points is indicative of the departure of the surface velocity from fully developed flow. Near the disk, $r^2 = 4.77$ sq. cm., acceleration of the interface can readily be seen.

To predict mass transfer rates from these data the experimental values of τ were substituted into Equation (6) which was then integrated numerically over the transfer area. To facilitate the computation the data were first fitted to appropriate algebraic equations. In the region of acceleration a quadratic equation was used, and the remainder of the points fell on a straight line, except near the wall where the talc particles were retarded by the wall. Representative values of ϕ calculated from the measured surface age are shown in Table 2 along with experimentally measured transfer rates. The agreement is excellent, within 2%, except at the highest flow rates where human reaction time becomes important in making the talc measurements and where small errors in the measured concentration are amplified when the mass transfer is expressed as a flux. Of the two systems shown in Figure 3, surface age measurements were made only on the isobutanol system. Since the two sets of data show almost exact agreement on Figure 3, it can be assumed that the ethyl acetate data would show comparable agreement when measured contact times were used. The important conclusion is that the standard diffusion equation is sufficient to predict the mass transfer results precisely, provided only that the true surface age is used in the calculations.

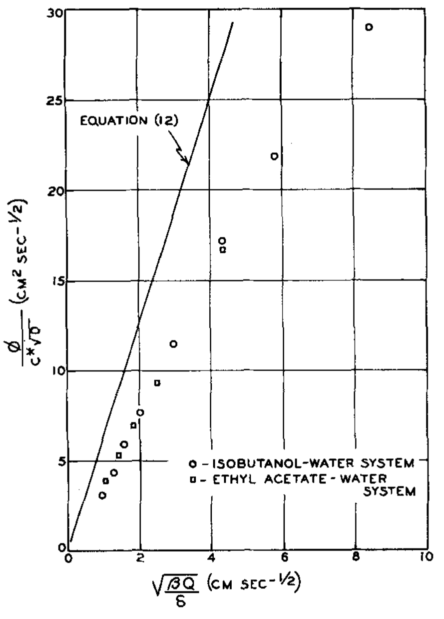

Fig. 3. Rate of mass transfer into water for isobutanol and ethyl acetate.

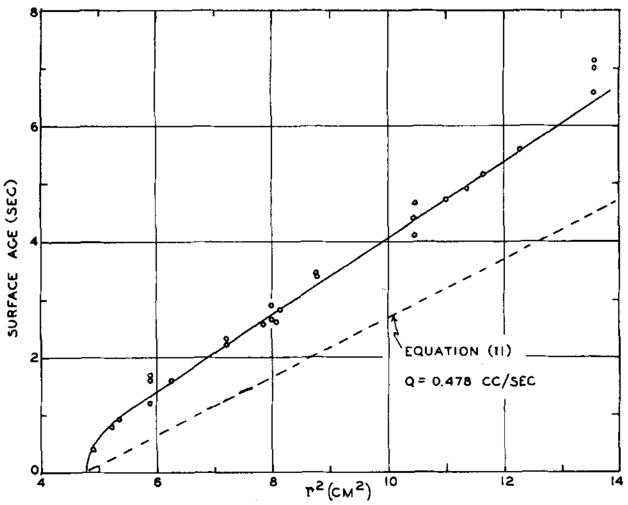

Fig. 4. Experimental determination of surface age.

Stagnant End Effects

Experiments with the carbon dioxide-water system produced the somewhat surprising result that the interface was completely stagnant. The fact that the chamber was sealed for this system prevented talc measurements, but later experiments with an air-water interface confirmed the conclusion that the interface was stagnant. Separate studies of a benzene-water system exhibited the same behavior and will be discussed along with the carbon dioxide data.

The mass transfer results are plotted in Figure 5 as a function of $(Q/\delta)^{1/2}$. The approximate equations for the moving interface, Equation (12), and stagnant interface, Equation (14), are plotted for comparison; the latter predicts values of the flux which are 10 to 30% lower than the measured values. In a similar study of absorption of carbon dioxide in a wetted-wall column, Lynn, Straatemeier, and Kramers (6) operated with a completely stagnant surface. Their results were "15 to 20% above the curve" of an equation for absorption through a stagnant interface derived similarly to Equation (14), and they suggest that the deviation is due to the assumptions in-

TABLE 2. COMPARISON OF EXPERIMENTALLY MEASURED RATE OF MASS TRANSFER WITH RATE PREDICTED FROM MEASURED SURFACE AGE

Q, cc./sec.	$\phi \times 10^5$ (predicted), moles/sec.	$\phi \times 10^5$ (experimental), moles/sec.
0.0772	1.55	1·57
0.250	3.04	3.06
0.660	4.57	4.60
1.92	6.41	6.65

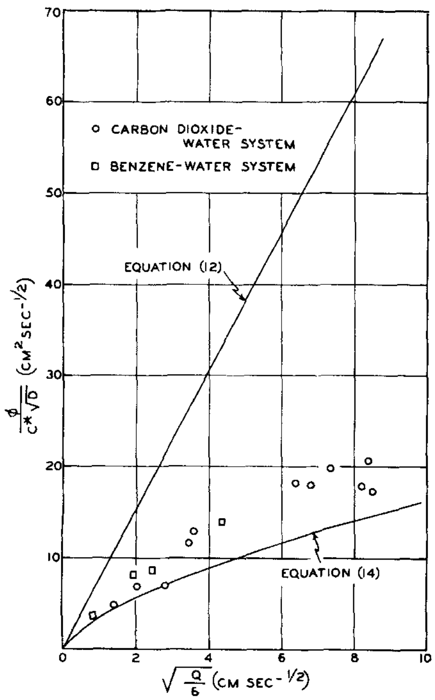

Fig. 5. Rate of mass transfer into water for carbon dioxide and benzene.

volved in the derivation. While this is true to some extent, talc measurements indicate a considerable amount of random motion in the otherwise stagnant interface, a motion which would enhance the rate of mass transfer.

The totally stagnant interface is another type of stagnant end effect, quite different from the minor effect alluded to in the freely moving isobutanol and ethyl acetate systems. For the latter case, the moving interface decelerates over a short distance near the wall stagnation point in a region where the concentration boundary layer is relatively thick and the local rate of mass transfer is small. Thus the decrease in the total or average flux is on the order of 1 to 2%. For the totally stagnant interface, however, the decrease in mass transfer is drastic.

Experiments are currently being carried out to determine the mechanism of the stagnation. Tentatively, the explanation is the same as that proposed by several investigators, namely that a surface-active contaminant present in minute quantities in one phase or both diffuses to the interface and is swept downstream like so much molecular flotsam to accumulate at the wall at the natural stagnation point in the flow. As the driftage stacks up at the wall, it forms a semirigid skin capable of supporting a shear stress and eventually, rapidly or slowly, covers the entire surface, perhaps as a monomolecular film.

A monolayer of surface-active impurities might be expected to impede the rate of mass transfer through interaction with the diffusing component. This does not appear to be the case in the systems studied here; the reduced rate of transfer with the stagnant interface may be adequately accounted for through hydrodynamic factors alone.

The explanation that surface-active agents cause the stagnation of the interface is supported by data for the methyl isobutyl ketone-water system. For example, the graph shown as Figure 6 was obtained in the following way. Normal operation of the mass transfer chamber at a water flow rate of 8 cc./min. produced conditions such that the organic-water interface was completely stagnant. Periodically, samples were taken and analyzed. Seven minutes before a given time, arbitrarily marked zero in the plot, the interface was swept for 7 min. This disturbance caused a slight drop in the concentration of the outlet solution but removed the invisible stagnant film so that the interface moved freely over about 80% of its radial length. At this flow rate the holdup time of the water in the chamber was approximately 9 min., so that 9 min. after the sweeping was stopped, a dramatic rise in the outlet concentration reflected the effect of the moving interface. The concentration then gradually dropped as the interfacial film grew, and, as observed in the microscope, the interface again reached complete stagnation. At higher flow rates the stagnant film did not cover the entire surface but grew back from the wall to an equilibrium position intermediate between the disk and wall.

Further investigation showed that the ability of the interface to resist stagnation depends upon the interfacial tension. Interfaces for systems with low interfacial tension moved freely, whereas those with high interfacial tension were invariably stagnant. The methyl isobutyl ketone system was intermediate, being stagnant for low flow rates but mobile at higher rates. The systems tested are listed in Table 1 with the literature values of some physical properties and the type of interfacial motion obtained. The interfaces marked stagnant probably do move for a very short time after sweeping if the shear stress caused by the flow is high enough. For example, the air-water interface was partly mobile for several minutes at thin water films and high flow rates. Under normal conditions, however, the interface was stagnant by the time the first talc measurements were made.

Free energy considerations indicate that for a constant bulk concentration of surface-active impurities the greater the interfacial tension, the greater the surface concentration of adsorbed molecules. The chemical nature and the absolute concentration of the contaminants present in these experiments is unknown. For this reason, it is not possible to explain why an interfacial tension of approximately 10 dynes/cm., for the pure binary, separates the stagnant and moving cases. Here it should be reiterated that it is extremely unlikely that these surface effects are due to inadvertent contamination. All due precautions were taken to insure that the apparatus and reagents were free from surface-active impurities. It is reasonable to assume that the concentration of impurities encountered here is comparable to that of similar studies in other laboratories and is certainly smaller than concentrations present in industrial equipment.

Interfacial Turbulence

With the contacting arrangement used here the interface was everywhere visible, and motion in the interface was readily observable. In several of the binary systems a highly structured interfacial turbulence was detected. The flow pattern, which could best be seen in light reflected from the interface, often appeared as radial streamers at or near the disk; that is it appeared as if closely spaced

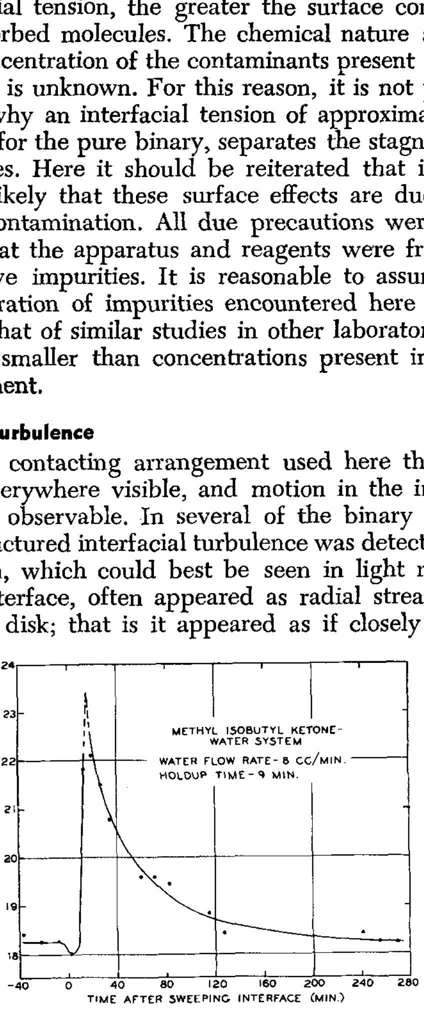

Fig. 6. Effect of interface stagnation on mass transfer.

radial lines had been drawn on the interface. At some distance out from the disk, at a surface age of 2 to 3 sec., tiny cells appeared between these streamers and grew as they traveled outward, looking somewhat like small ripples. The depth of these ripples, or indentations, was less than 30 μ, the smallest value detectable with the variable-needle depth gauge. At low flow rates it was sometimes possible to see remarkably regular hexagonal cells in the interface. At other times the agitation was more chaotic with no discernable structural pattern.

The Marangoni effect arises spontaneously at an interface from ever present, localized fluctuations in interfacial concentration or temperature. These small fluctuations cause local variations in the interfacial tension which in turn produce motion in and around the interface. If the system is stable, small disturbances will be damped out; if it is unstable, they will be amplified and grow into finite, three-dimensional, convection patterns. Using hydrodynamic stability theory, Sternling and Scriven (*11*) have presented a theoretical analysis for ternary systems where a solute is transferring between two noninteracting phases.

Apparently, Marangoni effects have not been reported previously for binary systems. Moreover, the mechanism by which such motion occurs is not readily apparent. For an isothermal binary system, with interfacial equilibrium assumed, the concentration at the interface, and hence the interfacial tension, is constant. To explain the motion observed in this work, radial variations in interfacial tension must have existed. Possible explanations are that the interface was not at equilibrium, or, more likely, temperature gradients existed near the interface. Temperature variations could be brought about by the heat of solution accompanying the mass transfer. A temperature gradient would produce an interfacial tension gradient directly as the interfacial tension is a function of temperature and indirectly because the interfacial concentration is temperature dependent. A still further explanation of the interfacial turbulence is that impurities were transferring between the phases. This last effect seems unlikely in light of the purity of the reagents used.

When unsaturated isobutanol was placed upon the flowing water, structured turbulence was observed in the interface. Presaturating the water with isobutanol had no effect on the turbulence, but saturation of the alcohol terminated the activity. Similarly, turbulence was noted for transfer of water into unsaturated ethyl acetate and into *n*-butyl acetate. Again, saturation of the organic phase terminated the turbulence either by gradual diminution as diffusion proceeded or by presaturation which precluded transfer altogether. Octanol behaved differently, exhibiting turbulence which was not affected by saturating the alcohol with water, an indication that transfer out of the organic phase was responsible. Benzene and methyl isobutyl ketone showed no spontaneous disturbances.

It must be emphasized that whereas the positive indications of interfacial turbulence are unmistakable, the statement that there was no turbulence means that none was seen. Under the proper conditions of lighting, chamber size, or interfacial cleanliness, turbulence might be observed beyond what is reported here. For example high rates of mass transfer appear to promote turbulence. Benzene, which has a very small solubility in water, also produced a stagnant interface which lowered the rate of mass transfer still further. It is possible that if the benzene-water interface could be viewed shortly after formation, before diffusion rates diminished, spontaneous motion in the interface could be seen.

In ternary systems interfacial turbulence unquestionably enhances the rate of mass transfer. In the present experiments mass transfer measurements were carried out with the isobutanol-water system in which interfacial turbulence was present and with the ethyl acetate-water system in which it was absent. Results for isobutanol identical to those shown in Figure 3 were obtained. Evidently, the scale of the motion, though clearly visible, is sufficiently small that it has very little effect on the rate of mass transfer.

ACKNOWLEDGMENT

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NOTATION

a,b,c = constants defined by Equations (8) and (9)
C = solute concentration in water phase, g.-moles/cc.
C^* = equilibrium concentration, g.-moles/cc.
$\bar{C}$ = average concentration, g.-moles/cc.
D = diffusivity, sq. cm./sec.
H = depth of the two fluid phases, cm.
N = local mass transfer rate, g.-moles/sq. cm. sec.
Q = volumetric water flow rate, cc./sec.
r = radial distance measured from the axis of the cylinder, cm.
r_o = radius of the disk, cm.
r_1 = radius of the glass cylinder, cm.
t = time, sec.
u = velocity in the radial direction, cm./sec.
u_i = interface velocity, cm./sec.
$\bar{u}$ = average radial velocity of a phase, cm./sec.
U = dimensionless velocity = $u/\bar{u}_w$
V = velocity, cm./sec.
y = coordinate normal to interface, cm.
Y = dimensionless coordinate = y/δ

Greek Letters

β = ratio of interface velocity to average water velocity, dimensionless
Γ = gamma function
δ = thickness of the water film, cm.
μ = viscosity, g./cm. sec.
τ = surface age, sec.
ϕ = total mass transfer rate, moles/sec.

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