

## SOLUTE EXTRACTION FROM AN INTERNALLY CIRCULATING SPHERICAL LIQUID DROP

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**Abstract**—The forced convective extraction of a solute from a steadily falling liquid drop in Hadamard or Hill flow is examined. A qualitative description of the concentration boundary layers and wakes is given and the dominance of transfer in the drop interior is demonstrated. A quantitative description of the mass transfer in the drop interior is given and the overall mass transfer from the drop is obtained.

This result is in general agreement with some published experimental results.

### NOMENCLATURE

<p><math>a</math>, radius of drop;</p> <p><math>C</math>, order of magnitude of change in concentration of solute on moving from stagnation ring to drop surface;</p> <p><math>C'</math>, order of magnitude of the difference in concentration of solute between two regions specified by the context;</p> <p><math>c</math>, normalized concentration [see equations (9) and (10)];</p> <p><math>c'</math>, concentration of solute (mass of solute per unit volume of solution);</p> <p><math>c_1</math>, value of <math>c</math> at a point inside the drop;</p> <p><math>c_0</math>, value of <math>c</math> at a point outside the drop;</p> <p><math>\bar{c}</math>, average value of <math>c</math> on a stream surface;</p> <p><math>c^*</math>, deviation of <math>c</math> from <math>\bar{c}</math>;</p> <p><math>D</math>, coefficient of diffusion;</p> <p><math>E</math>, equilibrium constant;</p> <p><math>H</math>, <math>\oint h_m h_q h \cdot dq</math>;</p> <p><math>h_m</math>, <math>\frac{1}{8p \cdot \sin \theta \cdot \Delta}</math>;</p> <p><math>h_q</math>, <math>\frac{(2p^2 - 1)^2}{4p^3 \cos^3 \theta \cdot \Delta}</math>;</p> <p><math>h_\lambda</math>, <math>p \sin \theta</math>;</p> <p><math>J</math>, <math>\oint \frac{h_q h_\lambda}{h_m} dq</math>;</p> <p><math>m</math>, <math>4p^2(1 - p^2) \sin^2 \theta</math>;</p> <p><math>Pe</math>, <math>\frac{v_0 a}{D}</math>, Peclet number;</p> <p><math>p</math>, <math>\frac{r}{a}</math>;</p>	<p><math>q</math>, <math>\frac{p^4 \cos^4 \theta}{2p^2 - 1}</math>, co-ordinate orthogonal to <math>m</math> and <math>\lambda</math>;</p> <p><math>Re</math>, <math>\frac{Ua}{\nu}</math>, Reynolds number;</p> <p><math>r</math>, distance from drop centre;</p> <p><math>s</math>, axial distance from the rear stagnation point in a direction towards the drop centre;</p> <p><math>Sc</math>, <math>\frac{\nu}{D}</math>, Schmidt number;</p> <p><math>t</math>, time;</p> <p><math>U</math>, speed of drop relative to bulk fluid at infinity;</p> <p><math>\mathbf{v}</math>, velocity;</p> <p><math>v_m, v_q, v_\lambda</math>, components of <math>\mathbf{v}</math> in <math>m, q, \lambda</math> co-ordinate system;</p> <p><math>v_0</math>, fluid speed at drop equator.</p> <p><b>Greek symbols</b></p> <p><math>\Delta</math>, <math>\Delta^2 = (1 - 2p^2)^2 \sin^2 \theta + (1 - p^2)^2 \cos^2 \theta</math>;</p> <p><math>\kappa</math>, ratio of internal fluid viscosity to external fluid viscosity;</p> <p><math>\lambda</math>, <math>\lambda =</math> constant are planes through the axis of symmetry;</p> <p><math>\nu</math>, kinematic viscosity;</p> <p><math>\psi</math>, stream function;</p> <p><math>\theta</math>, angular co-ordinate measured from upstream direction;</p> <p><math>\tau</math>, <math>t \frac{D_1}{a^2}</math>, non-dimensionalised time.</p> <p><b>Subscripts</b></p> <p>0, outside the drop;</p> <p>1, inside the drop.</p> <p><b>Superscripts</b></p> <p><math>i</math>, initial value.</p>
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## 1. INTRODUCTION

THIS paper examines the forced convective transfer of a solute from a steadily falling liquid drop under certain idealized conditions. The liquid drop moves at constant velocity through a second liquid (the two liquids being immiscible) whilst the solute, initially at higher concentration inside the liquid drop than outside, transfers from the inside to the outside by a diffusion process. The fluid inside the drop is assumed to be steadily circulating without turbulence and free convection is taken to be unimportant. Furthermore, this discussion is restricted to the case where the Peclet number of the diffusion process is large. It is also assumed that the drop is spherical and that the motion takes place either in the low Reynolds number creeping flow regime or in the high Reynolds number inertial flow regime. The principal effect considered is that of internal circulation, and it is with this in mind that these assumptions have been formulated. The cases of little or no internal circulation are not considered here.

This problem has been considered by a number of authors. Johns and Beckmann [8] used a numerical method. They were able to obtain solutions for a range of Peclet numbers from 0 to 80 in the case where all the resistance to diffusion was inside the drop. Analytical methods have also been used—there are two papers by Levich *et al.* [1, 2], and one by Ruckenstein [3]. These authors considered that the behaviour of the boundary layer determined the mass transfer and assumed that the solute concentration in the drop, away from the boundary layer, is unchanged. This restricts the validity of the solutions to the initial stages of the diffusion process. Kronig and Brink [4] assumed that the diffusion process in the interior of the drop was of paramount importance. Their view is adopted by this present paper, which obtains results close to theirs.

It will be assumed that the transport of a solute is governed by the diffusion equation:

$$\frac{\partial c'}{\partial t} = -\mathbf{v} \cdot \nabla c' + D \nabla^2 c' \quad (1)$$

and that the flow in and around a drop is described by the stream functions:

$$\psi_1 = \frac{U}{a^2} \frac{(a^2 r^2 - r^4)}{4(\kappa + 1)} \sin^2 \theta \quad (2)$$

and

$$\psi_0 = -\frac{U}{4} \left( 2r^2 - \frac{3\kappa + 2}{\kappa + 1} ar + \frac{\kappa}{\kappa + 1} \frac{a^3}{r} \right) \sin^2 \theta \quad (3)$$

in the low Reynolds number case, and

$$\psi_1 = \frac{3U}{4a^2} (a^2 r^2 - r^4) \sin^2 \theta \quad (4)$$

and

$$\psi_0 = -\frac{1}{2} U r^2 \left( 1 - \frac{a^3}{r^3} \right) \sin^2 \theta \quad (5)$$

in the high Reynolds number case. These flow fields are taken from Hadamard [5] and Harper and Moore [7] respectively. For practical purposes it must be emphasized that the high Reynolds number flow given here will only be realized in the complete absence of surface-active materials. Throughout the following description it will be assumed that a solute is being extracted from the drop, but the theory is clearly applicable, with the obvious minor changes, to the case of a solute being extracted from the bulk fluid by the drop. It is also applicable to the case of a pure bulk solute dissolving into a solvent drop.

## 2. THE QUALITATIVE THEORY

There is an initial period during which the motion is unsteady as the drop accelerates or decelerates towards its terminal velocity. Obviously the transfer process is occurring during the initial acceleration of the drop but, if the depth to which the effect of extraction penetrates in this time is small compared with the size of the drop, this initial period can be neglected and it may be assumed that the diffusion process commences when the drop attains terminal velocity. This penetration depth may be estimated from the equations governing the motion and the mass transfer and can be shown to be of the order of  $a \cdot Sc^{-1/2}$  where  $Sc$  is the Schmidt number. Much of the future discussion will be concerned with concentration boundary layers of thickness  $a \cdot Pe^{-1/2}$ . These will be substantially unaffected provided that  $a \cdot Pe^{-1/2} \gg a \cdot Sc^{-1/2}$ , that is if  $Re \ll 1$ , which is always true for the low Reynolds number regime. It is not true at high Reynolds number, but even in this case the penetration depth is small compared with the drop radius, provided that the Schmidt number is large.

The drop surface is the position of a discontinuity in the physical properties of the liquids. In particular the diffusion coefficient changes there. Concentration boundary layers of thickness  $a \cdot Pe^{-1/2}$  therefore form on both the inside and the outside of this surface. (There are two Peclet numbers—one for the external fluid and one for the internal fluid—but for the purposes of this order of magnitude discussion there is no point in distinguishing between them.) The fluid in the concentration boundary layers will participate in the general motion round the drop. When it reaches the neighbourhood of the rear stagnation point the fluid in the boundary layer outside the drop is convected away from the drop surface and forms a concentration wake. Similarly, the fluid in the internal boundary layer is convected away from the drop surface and

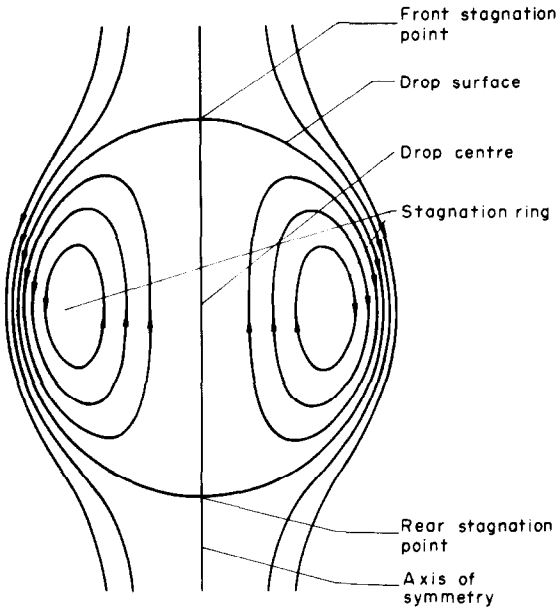


FIG. 1. The unperturbed flow pattern in and around a liquid drop at high Reynolds number.

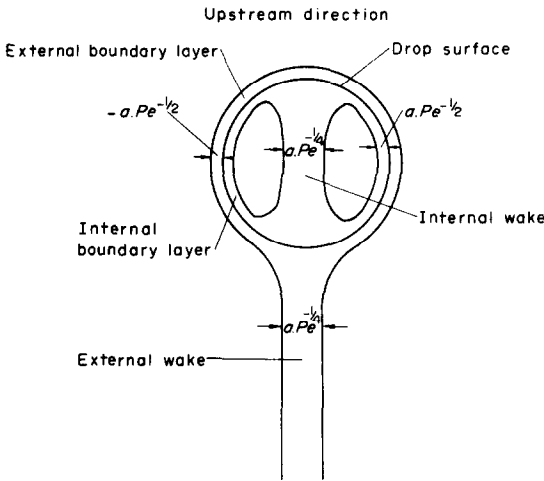


FIG. 2. The boundary layer-wake system.

forms an "internal wake". The fluid in this internal concentration wake flows along the central axis of the drop until, in the neighbourhood of the front stagnation point, it is convected by the internal circulation of the drop into the internal boundary layer. This system of boundary layers and wakes is pictured in Fig. 2.

It is possible to estimate the thickness of these concentration wakes by considering the flow, since the wake consists of fluid which has been convected from a boundary layer. At both high and low Reynolds numbers the pattern of streamsurfaces inside the drop

is the same and the stream function is of the form

$$\psi_1 = \frac{v_0}{2} r^2 \left(1 - \frac{r^2}{a^2}\right) \sin^2 \theta. \quad (6)$$

A fluid layer which has thickness of order  $a \cdot Pe^{-1/2}$  at the drop equator (as has the boundary layer) will therefore have thickness of order  $a \cdot Pe^{-1/4}$  at the drop axis. Consequently the internal wake has thickness of order  $a \cdot Pe^{-1/4}$ . The external wake also has thickness of order  $a \cdot Pe^{-1/4}$ . The two concentration wakes, therefore, are much thicker than the concentration boundary layers. The relative thicknesses of the wakes and boundary layers is of importance because their effect is to determine the relative rates of mass transfer from one streamsurface to another in the different regions. The concentration change is the same in moving from the boundary layer to the fluid outside the boundary layer as it is moving from the wake to the fluid outside the wake, and yet the thickness of a wake is much greater than the thickness of a boundary layer. Gradients of concentration are therefore much larger in a concentration boundary layer than they are in a concentration wake and the rate of mass transfer is consequently much faster. This has a particularly important consequence when the case of the internal concentration wake is considered. The thinness of a concentration boundary layer enables the fluid in it to significantly alter its concentration during its passage round the inside of the drop surface. Since a wake is so much thicker than a boundary layer, fluid in the wake can travel a similar distance without its concentration being significantly altered. This means that when the fluid in the internal wake flows along the central axis and thence back into the boundary layer in the region of the front stagnation point its concentration is relatively unaltered. Thus the concentration of a particle which is part of the boundary layer-wake system is effectively the same when it reaches the region of the front stagnation point as it was when it left the region of the rear stagnation point, in spite of having moved along the central axis of the drop in the internal wake. The error in neglecting the change in concentration as a particle flows through the internal wake is of the order of  $C' \cdot Pe^{-1/2}$ . [This value, like other orders of magnitude given in this section, is obtained by substituting approximate magnitudes for the relevant terms in the diffusion equation (1). In this case  $\partial c'/\partial t$  is represented by  $\Delta c \cdot a/U$  where  $\Delta c$  is the required change in concentration of a particle as it moves from one end of the internal wake to the other,  $a/U$  is the order of magnitude of the time taken for a particle to move a distance approximately equal to  $a$  at a speed approximately equal to  $U$ . The convection term  $v \cdot \nabla c'$  is not relevant here since it is the effect of diffusion into the wake from the rest of the drop that

is being estimated. The diffusion term  $D\nabla^2 c'$  is represented by  $DC'/(a \cdot Pe^{-1/4})^2$  where  $C'$  is the change in concentration in moving from the axis of the wake to the outside of the wake—that is a distance of order  $a \cdot Pe^{-1/4}$ . Equating these expressions for  $\partial c'/\partial t$  and  $D\nabla^2 c'$  gives the required order of magnitude.

Because the concentration boundary layer is very thin the fluid from it passes close to the drop rear stagnation point. Just how close may be estimated by determining the maximum distance of the rear stagnation point from the streamsurface which bounds the boundary layer and wake system, and is of the order  $a \cdot Pe^{-1/6}$ . It follows that  $\nabla^2 c'$  in this region is of the order of  $DC'/(a \cdot Pe^{-1/6})^2$ . A particle takes time of order  $a/v_0[\log s/a]$  to travel through the region near the stagnation point, so the concentration change during this passage is of order  $C' \cdot Pe^{-2/3}[\log s/a]$ . In the case of a particle that moves from the boundary layer (where  $s$  is of order  $a \cdot Pe^{-1/2}$ ) to the internal wake (where  $s$  is of order  $a$ ) the change in concentration is therefore of the order of  $C' \cdot Pe^{-2/3} \cdot \log Pe$ , and a similar change occurs in moving from the internal wake through the region near the front stagnation point and into the internal boundary layer. These changes are smaller than the change that occurs in moving along the internal wake (although the difference is small), so that the change in concentration in a particle as it moves from the internal concentration boundary layer in the region of the rear stagnation point to the same boundary layer in the region of the front stagnation point is determined by the change in the wake and is of order  $C' \cdot Pe^{-1/2}$ . Thus the statement that the concentration in the internal wake is the same function of the stream function in the region of the rear stagnation point as it is in the region of the front stagnation point, is in error only by a term of order  $Pe^{-1/2}$  of the concentration change in moving from the drop interior to the wake. The external wake, like the internal wake, has thickness of order  $a \cdot Pe^{-1/4}$ . Its length is therefore of order  $a \cdot Pe^{1/2}$ . The external wake is thus long and thin compared with the drop radius.

It is now possible to describe the overall mechanism of the transfer process. The essential point is that the boundary layers are a fast and efficient mechanism for mass transfer, the wakes are less efficient, and the drop interior less efficient still. (Here and subsequently the word "interior" refers to everything enclosed by the drop surface, excepting the internal boundary layer-wake system.)

To make this argument more specific suppose that the fall in concentration across a boundary layer is of order  $C'$ , so that the concentration gradient is of the order of  $C' \cdot Pe^{1/2}/a$ , then the order of magnitude of the change in concentration of a particle moving through the boundary layer is  $C'$  in time  $a/U$ . Compare

this with the rate of diffusion in the interior; here the concentration gradient is of order  $C/a$  (where  $C$  is the order of the change in concentration on moving from the stagnation ring to the drop surface), so that in time of order  $a/U$  the concentration of a particle changes, by molecular diffusion, by an amount of order  $C \cdot Pe^{-1}$ . Clearly if  $C$  and  $C'$  are of the same order of magnitude the boundary layers will be transferring solute at a much faster rate than the drop interior. In the low Reynolds number case there is a short initial period during which this does happen. The result is that the solute in the internal boundary layer is transferred to the outside of the drop and, since the rate of transfer from the interior is far too slow to replenish this loss, the concentration in the internal boundary layer falls to a low value. Since the internal wake consists of fluid which has passed through the internal boundary layer it also loses most of the solute initially dissolved inside it in the short initial stage of the transfer process. This initial stage lasts only as long as it takes for the fluid to be transferred out of the boundary layer-wake system. This is the time taken for the fluid within the boundary layer-wake system to circulate once—that is for a time of order  $a/U \log Pe$ . In the high Reynolds number case the initial concentration in the internal boundary layer is much smaller than that in the interior because the solute is removed during the period of unsteady flow. In both high and low Reynolds number cases therefore the concentration in the internal boundary layer quickly falls to a low value. From this moment on the rate governing process at all points inside the drop is the diffusion of solute in the interior, for changes in the boundary layer are determined only by transfer of solute to it from the interior. Effectively therefore the boundary layer is at steady state: its own time scale is completely suppressed and time affects it only indirectly through the time-dependent transfer from the interior.

The internal wake plays practically no part in the transfer. Transfer into it from the interior occurs in the same manner as transfer into the internal boundary layer, but its surface area is too small (it is of order  $a^2 \cdot Pe^{-1/4}$ , compared with the boundary layer's area of order  $a^2$ ) for any significant quantity of solute to be involved. And, as previously described, the wake is too thick for its own structure to be significantly altered.

The role of the external boundary layer is simply to transfer the solute from the drop surface to the external wake. This boundary layer is also an effective mechanism for the transfer and can remove the solute from the surface at a rate comparable with that with which it is transferred from the internal boundary layer to the surface.

Finally, it is possible to estimate the magnitude of the concentration of solute in the boundary layer—

wake system by considering the mass fluxes into and out of the system. The order of the mass flux from the internal boundary layer-wake system is  $D(C/a)a^2$  and the mass flux from the internal boundary layer to the surface and thence to the external boundary layer is of the order  $D(C/a)Pe^{1/2} \cdot a^2$ . There being no build up of solute in the internal boundary layer these two fluxes must be equal, so  $C'$  is of order  $C \cdot Pe^{-1/2}$ . The internal boundary layer-wake system therefore has a low concentration compared with the concentration near the stagnation ring. In cases where the equilibrium constant  $E$  is of order one the concentration in the external boundary layer will be of the same order of magnitude as that in the internal boundary layer, and the change in concentration in moving from the external boundary layer to the bulk fluid will also be of order  $C'$ , since this concentration difference is maintained by transfer from inside the drop. Compared with the order of  $C$  this difference is small and the statement that the external boundary layer is at bulk fluid concentration is therefore in error only by a term of order  $C \cdot Pe^{-1/2}$ . Consequently the concentration in the internal boundary layer is also near the value which would be in equilibrium with the bulk fluid, again with an error of order  $C \cdot Pe^{-1/2}$ .  $C$  is effectively the difference between the concentration at the stagnation ring and the concentration that would be in equilibrium with the bulk fluid. This result means that there can be no steep gradient of concentration in the boundary layers. The interior and the internal boundary layer merge smoothly into one another and the gradient of concentration is of order  $C/a$  in both regions. Of particular importance is the result that solute is carried along the internal wake without significant diffusion. This contradicts one of the assumptions made by Levich *et al.* [1, 2] and by Ruckenstein [3], namely that the internal boundary layer is constantly fed with fresh solution in the region of the front stagnation point. Such an assumption implies that the highly efficient boundary-layer transfer is the rate governing process, instead of the much slower interior transfer that the present theory emphasises.

### 3. MASS TRANSFER IN THE DROP INTERIOR

Since the boundary layer is thin and transfer in the interior is the rate governing process, a first approximation to the mass transfer from the drop may be obtained by neglecting the existence of the boundary layer and solving the diffusion equation as though the drop consisted only of the interior. By using the properties of transfer at high Peclet number it is possible to eliminate one of the space variables in the unsteady state diffusion equation (1), whereupon the equation may be reduced to a recognizable type that can be solved by fairly simple numerical means.

It can be shown that everywhere in the interior the mean concentration for a stream surface is a valid approximation to the concentration at any point on that stream surface. Thus it is convenient to express the diffusion equation in terms of a co-ordinate system in which one of the co-ordinates is at least approximately constant on a stream surface. The system used here (the same as that used by Kronig and Brink [4]) is

$$m = 4p^2(1-p^2)\sin^2\theta \quad (7)$$

$$q = \frac{p^4 \cos^4\theta}{2p^2-1} \quad (8)$$

$m$  is a constant along the zero Reynolds number stream surfaces.

It is also convenient to normalize the concentrations:

$$c_1 = \frac{c' - Ec'_0}{c'_1 - Ec'_0} \quad (9)$$

and

$$c_0 = \frac{c' - c'_0}{c'_1 - Ec'_0} \quad (10)$$

In terms of these variables the diffusion equation is:

$$\begin{aligned} \frac{\partial c}{\partial t} + \frac{v_q}{h_q} \frac{\partial c}{\partial q} + \frac{v_m}{h_m} \frac{\partial c}{\partial m} \\ = \frac{D_1}{a^2 h_m h_q h_\lambda} \left[ \frac{\partial}{\partial m} \left( \frac{h_q h_\lambda}{h_m} \frac{\partial c}{\partial m} \right) \right. \\ \left. + \frac{\partial}{\partial q} \left( \frac{h_\lambda h_m}{h_q} \frac{\partial c}{\partial q} \right) + \frac{\partial}{\partial \lambda} \left( \frac{h_m h_q}{h_\lambda} \frac{\partial c}{\partial \lambda} \right) \right]. \quad (11) \end{aligned}$$

In Hadamard ( $Re = 0$ ) flow  $v_m$  is zero. At high Reynolds number  $v_m$  represents the perturbation from the inviscid flow and it is small (of the order of  $v_q \cdot Re^{-1/2}$ ) compared with  $v_q$ . Also, let

$$c(m, q, \lambda) = \bar{c}(m, \lambda) + Pe^{-1} c^*(m, q, \lambda) \quad (12)$$

with

$$\oint h_m h_q h_\lambda c^* dq = 0 \quad (13)$$

$\bar{c}$  is an average (normalized) concentration on a stream surface, and  $c^*$  is the deviation from this average at any point on the same stream surface. Now it may be shown, from axial symmetry, the continuity equation, etc., that (11) reduces to

$$\begin{aligned} \frac{\partial \bar{c}}{\partial t} \oint h_m h_q h_\lambda dq \\ = \frac{D_1}{a^2} \oint \left[ \frac{\partial}{\partial m} \left( \frac{h_q h_\lambda}{h_m} \frac{\partial \bar{c}}{\partial m} \right) + Pe^{-1} \cdot \frac{\partial}{\partial m} \left( \frac{h_q h_\lambda}{h_m} \frac{\partial c^*}{\partial m} \right) \right] dq \\ - Pe^{-1} \oint \frac{\partial}{\partial m} (v_m h_q h_\lambda c^*) dq. \quad (14) \end{aligned}$$

The second term on the r.h.s. is clearly negligible compared with the first, and (since  $v_m$  is small compared with  $U$ ) the third term may also be neglected, leaving

$$\frac{\partial \bar{c}}{\partial t} \oint h_m h_q h_\lambda dq = \frac{D_1}{a^2} \oint \frac{\partial}{\partial m} \left( \frac{h_q h_\lambda}{h_m} \frac{\partial \bar{c}}{\partial m} \right) dq \quad (15)$$

and this may be reduced to

$$H \frac{\partial \bar{c}}{\partial \tau} = \frac{\partial}{\partial m} \left( J \frac{\partial \bar{c}}{\partial m} \right). \quad (16)$$

Near the surface and axis, where  $m$  is of the order of  $Pe^{-1/2}$ , the interior merges into the boundary layer-wake system and the concentration is of the order of  $C.Pe^{-1/2}$  greater than at the drop surface. Thus imposing the boundary condition

$$\bar{c} = 0 \quad \text{at} \quad m = 0 \quad (17)$$

will introduce an error of order only  $Pe^{-1/2}$ . The boundary condition at the stagnation ring is obtained by assuming that  $\bar{c}$  is a regular function of  $m$  at this point. The required condition is

$$\frac{\partial \bar{c}}{\partial \tau} = -20 \frac{\partial \bar{c}}{\partial m} \quad \text{at} \quad m = 1. \quad (18)$$

The initial condition is the state of the concentration profile at  $\tau = 0$ . A valid approximation is

$$\bar{c} = 1 \quad \text{at} \quad \tau = 0, \quad m > 0. \quad (19)$$

The solution of this set of equations is shown graphically in Fig. 5. This solution is a first approximation to the concentration at all points within the drop, with an error of the order of  $C.Pe^{-1/2}$  or less everywhere.  $c = 0$  is a first approximation to the concentration at all points outside the drop, also with an error of order  $C.Pe^{-1/2}$  or less everywhere. The overall rate of extraction of solute is found by integrating the local mass-transfer rate over the "surface" that divides the boundary layer and the interior—to the accuracy required here that is equivalent to integrating over the surface  $m = 0$ . The local mass-transfer rate per unit area is  $-D(\partial \bar{c}/\partial r)$  so that the total mass transfer from the drop in unit time is

$$\frac{64}{3} \pi Da \left( \frac{\partial \bar{c}}{\partial m} \right)_{m=0} \quad (20)$$

and  $(\partial \bar{c}/\partial m)_{m=0}$  is shown as a function of  $\tau$  in Fig. 4.

#### 4. COMPARISON WITH EXPERIMENTAL RESULTS

Heertjes *et al.* [9], Johnson and Hamielec [10], and Skelland and Wellek [11] have measured the mass transfer in systems where a pure bulk solute dissolves into a liquid drop. This is a special case of the theory considered here: mathematically the situations are identical. In these experiments it was found that some systems (e.g. water drops in cyclohexanol, isobutanol

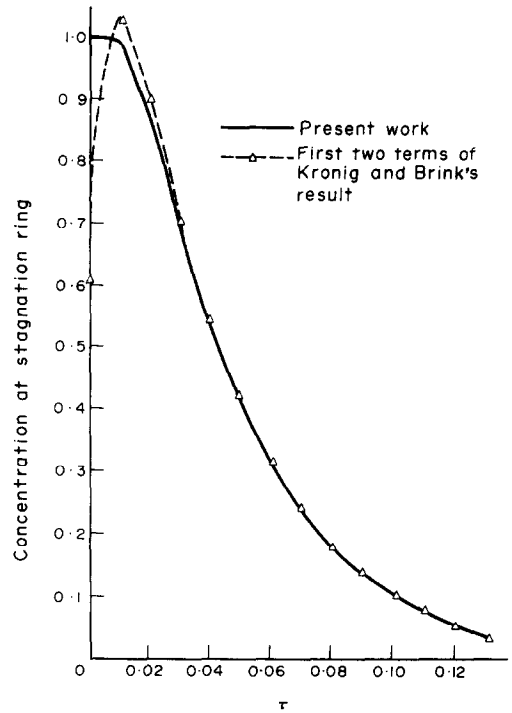


FIG. 3. Concentration at stagnation ring vs time.

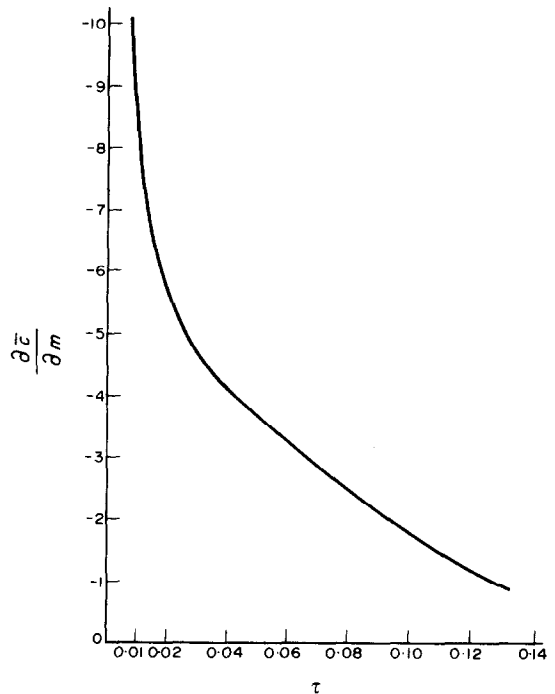


FIG. 4.  $\left( \frac{\partial \bar{c}}{\partial m} \right)_{m=0}$  as a function of  $\tau$ .

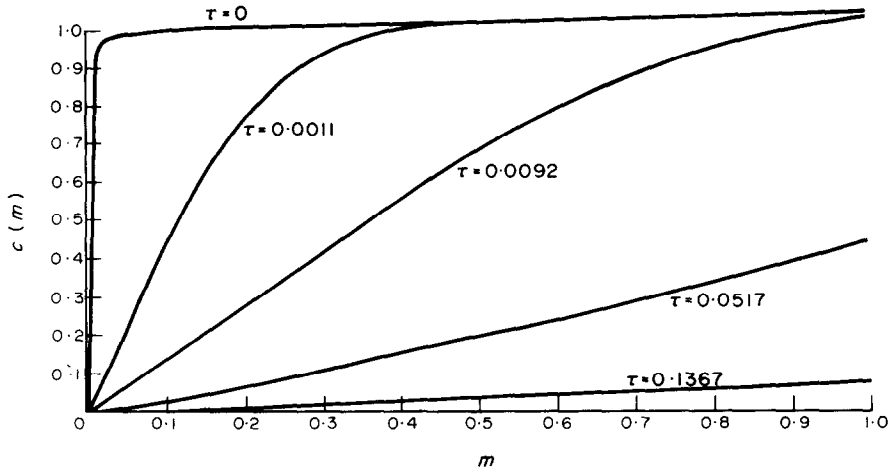


FIG. 5. Concentration profiles at various times.

drops in water) were in general agreement with Kronig and Brink's results (and hence with the results of the present paper) but this agreement is not sufficiently close to support a claim that the results of the theory are an accurate prediction of the rate of mass transfer. The observed rates of mass transfer are higher than those predicted by the theory. In other systems there was no agreement between theory and experiment. The reasons for these discrepancies is not always clear. Kadenskaya *et al.* [12] measured the rate of mass transfer when acetic acid was extracted from water by single drops of ethyl acetate. They conclude that there is good agreement with the results predicted by Kronig and Brink for drops smaller than 2.7 mm dia. These experiments were made at Reynolds numbers of the order of 100 to 500.

It can be seen therefore that the experimental results are insufficient to provide verification of the theory, although they do indicate general agreement.

##### 5. REMARKS

From a practical point of view the most important result of the qualitative description is its emphasis of the role of the internal wake in convecting solute from the region of the rear stagnation point to the region of the front stagnation point with negligible diffusion, and the consequent dominance of diffusion in the interior. If molecular diffusion played an important role in the internal wake then fresh solute would be continuously fed into the internal boundary layer with the result that the relatively fast boundary-layer diffusion would determine the time scale of the process, instead of the relatively slow diffusion in the interior which this theory has shown to be of paramount importance. A second consequence of the dominance

of the interior is that the coefficient of diffusion in the bulk fluid ( $D_0$ ) is of little importance. To a first approximation the process is governed by the coefficient of diffusion in the dispersed phase ( $D_1$ ).

Mathematically Kronig and Brink's problem [4] is identical to the solution of equations (16)–(19), but with the boundary condition at the stagnation ring (18) omitted and an assumption that  $\bar{c}$  may be approximated by a quadratic function of  $m$  inserted. This present paper has shown that Kronig and Brink's results can be extended to the case of high Reynolds number with the error remaining of order  $Pe^{-1/2}$ . The principal distinction between the high and low Reynolds number transfer is in the initial stage of unsteady motion—at high Reynolds number solute from a region deeper than the internal boundary layer is transferred out of the drop in the unsteady motion period. In the high Reynolds number case, therefore, the concentration boundary layers are effectively steady state throughout the period of steady motion, whereas in the low Reynolds number case there is a short initial period during which the boundary layers are unsteady.

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##### REFERENCES

1. V. G. Levich, V. S. Krylov and V. P. Vorotilin, Theory of non-stationary diffusion from a moving droplet, *Dokl. Akad. Nauk. SSSR* **161**, 648 (1965).
2. V. G. Levich, V. S. Krylov and V. P. Vorotilin, Theory of extraction from a falling drop, *Dokl. Akad. Nauk. SSSR* **160**, 1358 (1965).

3. E. Ruckenstein, Mass transfer between a single drop and a continuous phase, *Int. J. Heat Mass Transfer* **10**, 1785 (1967).
4. R. Kronig and J. C. Brink, On the theory of extraction from falling droplets, *Appl. Sci. Res.* **A2**, 142 (1951).
5. J. Hadamard, Mouvement permanent lent d'une sphère liquide et visqueuse dans un liquide visqueux, *C.R. Hebd. Séanc. Acad. Sci., Paris* **152**, 1735–1738 (1911).
6. W. Rybczynski, Über die fortschreitende Bewegung einer flüssigen Kugel in einem zähen Medium, *Bull. Acad. Sci. Cracovie (Series A)* 1911, 40–46.
7. J. F. Harper and D. W. Moore, The motion of a spherical liquid drop at high Reynolds number, *J. Fluid Mech.* **32**, 367 (1968).
8. L. E. Johns, Jr. and R. B. Beckmann, Mechanism of dispersed phase mass transfer in viscous, single drop extraction systems, *A.I.Ch.E. Jl* **12**, 10 (1966).
9. P. M. Heertjes, W. A. Holve and H. Talsma, Mass transfer between isobutanol and water in a spray column, *Chem. Engng Sci.* **3**, 122 (1954).
10. A. I. Johnson and A. E. Hamielec, Mass transfer inside drops, *A.I.Ch.E. Jl* **6**, 145 (1960).
11. A. H. P. Skelland and R. M. Wellek, Resistance to mass transfer inside droplets, *A.I.Ch.E. Jl* **10**, 491–496 (1964).
12. N. I. Kadenskaya, A. S. Zhelezynyak and B. I. Brounshtein, Mass transfer in extraction of acetic acid by single drops of ethyl acetate, *Zh. Prikl. Khim.* **38**, 1156 (1965).
13. A. E. Handlos and T. Baron, Mass and heat transfer from drops in liquid-liquid extraction, *A.I.Ch.E. Jl* **3**, 127 (1957).
14. F. Y. Pan and A. Acrivos, Heat transfer at high Peclet number in regions of closed streamlines, *Int. J. Heat Mass Transfer* **11**, 439–444 (1968).

### EXTRACTION DE SOLUTE D'UNE GOUTTE LIQUIDE AVEC CIRCULATION INTERNE

**Résumé**—On examine l'extraction en convection forcée d'un soluté contenu dans une goutte liquide tombant en écoulement de Hadamard ou de Hill. On donne une description qualitative des couches limites de concentration et des sillages et on démontre l'importance du transfert à l'intérieur de la goutte. Une description quantitative du transfert massique à l'intérieur de la goutte est présentée et on obtient le transfert massique global pour la goutte. Ce résultat s'accorde généralement avec les résultats expérimentaux publiés.

### EXTRAKTION EINES GELÖSTEN STOFFES AUS EINEM FLÜSSIGKEITSTROPFEN MIT INNERER ZIRKULATION

**Zusammenfassung**—Bei erzwungener Konvektion wird die Extraktion eines gelösten Stoffes aus einem stetig fallenden Flüssigkeitstropfen in einer Hadamard- oder Hill-Strömung untersucht. Eine qualitative Beschreibung der Konzentrationsgrenzschichten wird gegeben und die Dominanz des Übergangs im Tropfeninnern gezeigt. Es wird eine quantitative Beschreibung des Stoffübergangs im Innern des Tropfens und der Gesamtstoffübergangskoeffizient des Tropfens angegeben. Dieses Resultat stimmt im allgemeinen mit eigenen veröffentlichten experimentellen Ergebnissen überein.

### ЭКСТРАГИРОВАНИЕ РАСТВОРИМОГО ВЕЩЕСТВА ИЗ СФЕРИЧЕСКОЙ ЖИДКОЙ КАПЛИ С ВНУТРЕННЕЙ ЦИРКУЛЯЦИЕЙ

**Аннотация**—Исследуется экстрагирование путём вынужденной конвекции растворенного вещества из равномерно падающей жидкой капли в потоке Адамара или Хилла. Качественно описываются концентрационные пограничные слои и волны и демонстрируется преобладание переноса внутри капли. Количественно описывается массоперенос внутри капли, и получены данные по общему массопереносу от капли. Этот результат в основном согласуется с некоторыми опубликованными экспериментальными данными.