Mass Transfer and Wake Phenomena

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The external velocity field associated with the fall of a single drop through a quiescent liquid phase is discussed. The field responsible for the transfer of vorticity from the point of generation to the disperse phase is assumed to be almost identical with the field that transfers mass from the drop interface. The stream surfaces associated with this field are made visible by means of dye trails. Since the field is not amenable to mathematical formulation, and since many reports appear in the literature concerning correlations between measurable flow parameters and the physical properties of the system, the field configurations relative to mass transfer mechanisms are considered qualitatively. The delineation of the flow patterns behind the drop gives some indication of the manner in which the external flow contributes to the transfer coefficients.

During the past twenty years a great deal of information has been obtained concerning the motions of liquid drops through gaseous and liquid media. Investigations of meteorological phenomena have been responsible for most experimental data relating to liquid-gas systems, while, apart from the few attempts to model water-air parameters by means of liquid-liquid phases, most experiments designed and executed to determine the motions of liquid drops through a second liquid phase have been prompted by chemical engineering considerations. Information regarding drop motions is pertinent to practically all operations in which two phases are brought into contact as drops of one phase dispersed in the other. Since the mathematical complexities arising from the nonlinear terms in the Navier-Stokes equations of motion admit almost no relevant solutions to the problems associated with velocities and drop sizes normally encountered in chemical engineering operations, most of the information has been a result of empirical and semiempirical approaches to these flow problems. It would appear that the flow fields around all but the smallest drops are more or less unknown, and the experimental data specific to each problem offer little in the way of generalization. The gross and detailed motions of drops have been examined relative to pertinent engineering processes such as diffusion and mass transfer, but there appear to be few experimental results which explicitly show the effects of the disturbances created in the continuous liquid phase. Of course, the flow pattern around all but the smallest drops cannot be determined analytically, but certain reproducible and sharply defined patterns can be delineated experimentally. Since any mass transfer between a moving drop and the ambient fluid is dependent on the manner and rate at which the various sections of the drop surface are flushed by the field liquid, the mechanics by which the wakes are formed and vorticity transferred from the region of generation to the main body of the fluid help to elucidate mass transfer, diffusion, and mixing processes. Since the difference in the physical

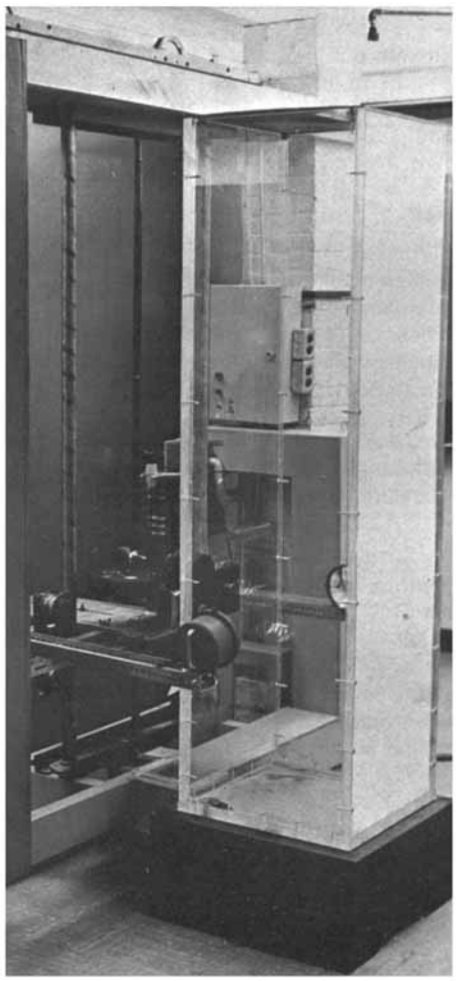

Fig. 1. Apparatus for study of wake formations behind falling drops.

properties responsible for mass transfer between the two continuous fluids is generally small, the rate at which the drop phase contacts fresh masses of the field fluid is an efficiency factor. It would appear that the peculiarities of the flow pattern around the drop must be considered as well as the gross motion of the drop through the fluid. Of course, it is recognized that in the experimental determination of transfer coefficients one is inherent in the other.

Investigations of flow regimes associated with liquid drops have been influenced by the theoretical and experimental results obtained for solids moving relative to gaseous or liquid media. Excellent bibliographies appear in a series of papers published by Torobin and Gauvin (*22*). Liquid drops are not discussed, but the pertinent hydrodynamical phenomena are reviewed and evaluated. However, if the fields of flow associated with the motions of rigid bodies have been difficult to treat theoretically, comparable liquid drop phenomena have almost defied mathematical formulation. The intractability is attributed to the internal circulatory motions, generated by the shear forces at the interface, and the deformations and oscillations which characterize drop motions over a great part of the Reynolds number spectrum. In most reports appearing in the literature in which drop motions are discussed, the disturbances in the continuous media are either treated casually or neglected altogether. Garner (*2*) outlined the conditions which promote or inhibit mass transfer between a falling drop and a liquid medium and noted the physical properties of either phase which appear to be responsible for such a transfer. The effects of the external flow field on the transfer phenomena are assumed to be implicit in those attributed to internal circulation shape and oscillation factors.

In reports dealing with the mechanics of drops per se, the external velocity field is generally recognized as being a factor contributing to such parameters as drag coefficient, but it is not treated explicitly. Hughes and Gilliland (*13*) considered many aspects of the gross and detailed motions of single drops falling through quiescent air, but the nature of the disturbances created in the gaseous medium was given little attention. Batchelor and Davies (*1*) examined the appropriate literature on the mechanics of drops and presented their results in a form which indicates the paucity of information on the external flow. Hamielec and Johnson (*10*) used numerical methods to predict velocity profiles for fluid spheres moving through viscous media under the influence of gravity. They discussed the mutual influence of the interior and exterior flow patterns, but the predicted profiles have never been observed experimentally even for the narrow range of Reynolds numbers for which the sphericity restrictions are valid.

The problem has been examined experimentally and theoretically because of the meteorological significance of the velocity fields around small spherical water droplets falling through the atmosphere. The nature of the disturbances associated with such droplets has relevance to drop growth theories by the collision and coalescence of small droplets. The hydrodynamical equations of flow around small liquid spheres have been considered and solutions obtained from which the trajectories of two droplets falling in close proximity can be calculated. Pearcey and Hill (*19*), Hocking (*11*), and Shafrir and Nieburger (*21*) have used different approximations to obtain solutions applicable to different flow parameters. The results are reduced to collision efficiencies which have a questionable agreement with experimentally determined values. This indirect evidence suggests that a backflow may exist behind drops of a certain size, but the velocity field is not delineated.

RESULTS OF PREVIOUS EXPERIMENTS

The data appearing in the literature indicate that many investigators have designed and executed experiments and obtained time-displacement data for a large number of liquid-liquid systems. Photographic methods have been employed to determine deviations from sphericity for equilibrium drop shapes and to record data relevant to oscillating drops. Since the properties of a particular system and the measured flow data are not amenable to mathematical formulation, the information is generally presented in usable form by the establishment of correlations between measured quantities such as terminal velocities and the physical properties of the disperse and field phases. Dimensional analysis techniques have been used by most investigators to effect correlations between experimental data and the pertinent variables or groups of variables. This type of presentation is exemplified by the analysis of Hu and Kintner (*12*). These investigators used the data obtained by allowing drops of ten different organic solvents to fall through distilled water to establish a correlation between experimental results and the physical properties of the system. They presented their correlation in the form of a graph where the group $C_D N_{We} N_P^{0.15}$ is plotted against the group $N_{Re} N_p^{0.15}$. Other investigators have used

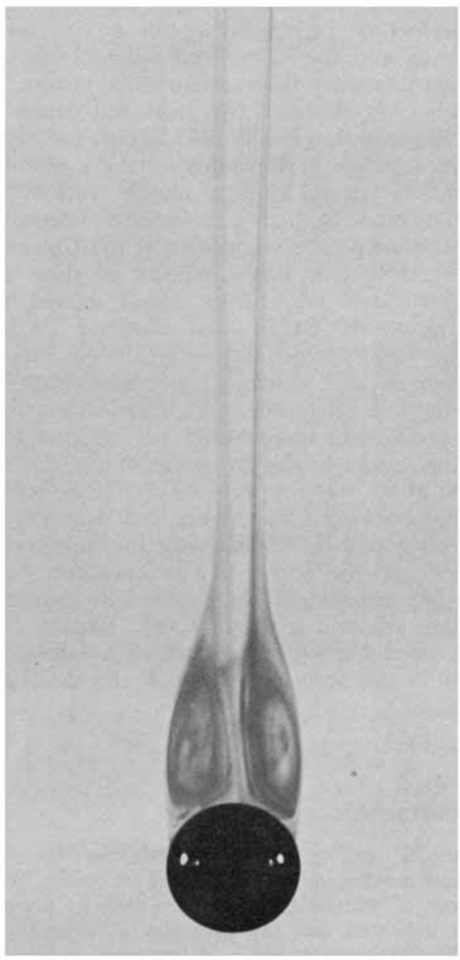

Fig. 2. The near wake region of a Class II wake. Reynolds number 220.

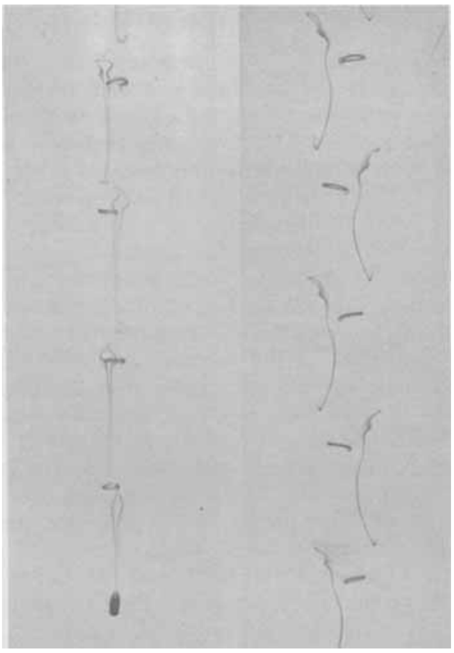

Fig. 3. Several cycles of the loop of Class IV wake. Reynolds number 370.

different physical property groups to correlate the terminal velocity and drop diameter to the physical characteristics of the system. Correlations were made by Johnson and Braida (*14*) and Licht and Narasimhamurty (*15*) for seven and six systems, respectively. The former effected a usable correlation by modifying the groups used by Hu and Kintner, and the latter used different physical property groups to present their experimental results. Satapathy and Smith (*20*) observed the gross and detailed motions of drops through immiscible field liquids for flows characterized by a range of Reynolds numbers extending from 0.01 to 1,500. Several kinds of motion were distinguished and the corresponding drag coefficients determined.

In a series of papers originating at the University, Edgbaston, Birmingham, many aspects of drop mechanics were investigated relative to actual mixing and mass transfer processes. Garner and Skelland (*4* to *7*) examined the factors which influence fall velocities in liquid-liquid systems and discussed the mechanics of solute transfer from drops to the field liquid. They determined transfer coefficients theoretically and experimentally and related the values to the dynamics of the drop, physical properties of the system, presence of surface active agents, and solute concentrations. Garner and Hale (*8*) discussed the dependence of the transfer rate on the temperature of the system and the degree of contamination. Garner and Grafton (*9*) considered the process by considering the flow pattern around a solid sphere. Garner (*2*) photographed the disturbed region behind a falling drop, but the detail is not sufficient to show the structure of the flow patterns.

EXPERIMENTATION

Experiments were carried out to determine the nature of the disturbances created in the quiescent liquid phase by the fall of single drops of various sizes and densities. As preliminary experiments indicated that the presence of impurities in either phase influences the external field in degree only, few precautions were taken to assess or eliminate these effects. Drops of different organic liquids containing finely powdered, water soluble, aniline dye were allowed to fall through quiescent water in a large tank. Dye traces, dissolved in the water of the boundary layer as it moved relative to the drop, marked the stream surfaces of the flow as the dye was carried into the wake region. The photographs appearing in this paper were of drops formed of carbon tetrachloride or some proportion of this liquid and kerosene. The lighter kerosene added to the heavier liquid afforded all densities between the value of 1 g./cc. for water and the value of 1.58 for carbon tetrachloride. For photographic considerations a small amount of K7014, D&C violet No. 2, carbon tetrachloride soluble dye was added to the drop liquid.

Figure 1 shows the apparatus used to obtain photographic data concerning the moving drop and the concomitant flow patterns as the drop moved through the water under the influence of gravity. The camera was mounted on a movable carriage that could be raised or lowered by a power driven screw device controlled by General Radio Type 1702-A motor control unit. The tracking was accomplished manually by the operator who observed the drop by means of a mirror periscope system. The reflected light from the illuminated drop was incident on a mirror attached to the moving carriage as shown in the photograph. After reflections from several mirrors enclosed in a light-tight tube, the final image was viewed by the operator in a mirror mounted just above the motor control unit. The camera was operated by means of a remote switch located on the side of the control panel. The drop and flow pattern was illuminated by two Type 1532-B General Radio strobolumes synchronized to the camera shutter. One strobolume was situated on either side of the tank. Single photographs were taken with a 2¼ × 3¼ Graphic, and the movie sequences were taken with a calibrated 35-mm. Arrflex camera. The tank of dimensions 6 ft. × 1½ ft. × 1½ ft. was constructed of 1 in. Perspex.

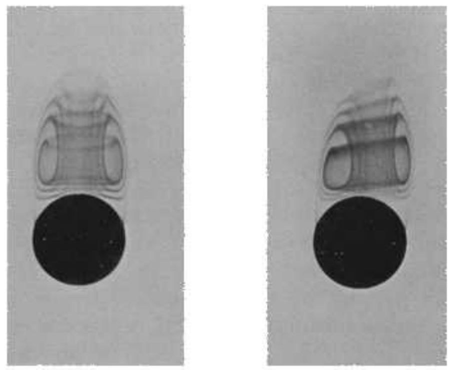

Fig. 4*a*. Class IV wake showing the central tube through which the backflow reaches the downstream surface of the drop. The two photographs were taken at right angles. Reynolds number 340.

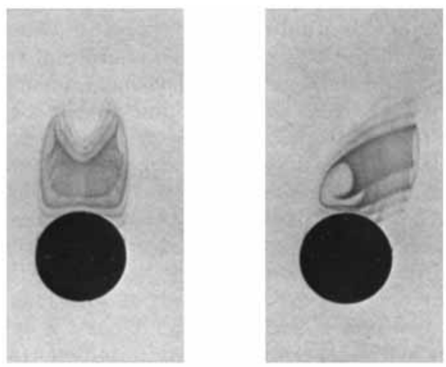

Fig. 4*b*. The change in geometry of the inflow tube is evident in this photographic pair. Reynolds number 340.

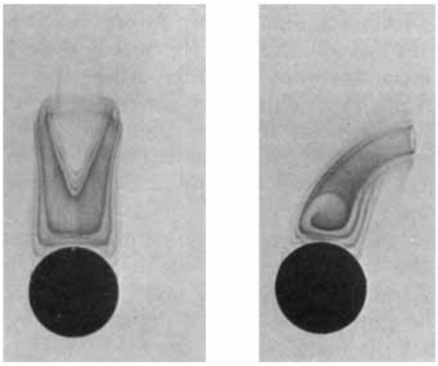

Fig. 4c. The identical drop as in the previous photographs. The backflow has all but ceased. Reynolds number 340.

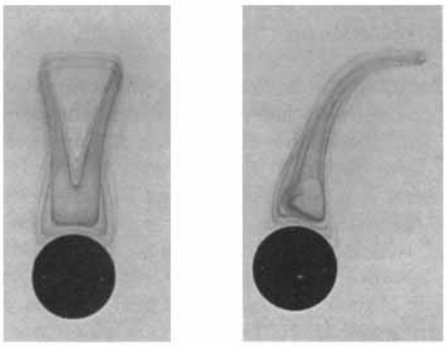

Fig. 4d. The loop is in the process of being disengaged.

WAKE PHENOMENA

Although it has long been recognized that interfacial conditions are not identical at liquid-liquid and solid-liquid interfaces, there are certain similarities between the flow geometries of the liquid drop and the rigid sphere. In fact, the data of Hu and Kintner (*12*) admit no distinction between the motions of liquid drops and rigid spheres for Reynolds numbers less than 300. However, an examination of the flow geometry indicates a difference in the external velocity fields even when the Reynolds numbers are identical. In the case of equilibrium drop shapes, it is difficult to determine whether the observed differences in wake configurations are due to differences in interfacial conditions, deviations from sphericity, or are a result of dissimilar observational techniques. The dye streaks resulting from the addition of finely powdered dye to the drop solution make it easier to observe the disturbances behind liquid drops than those behind comparable rigid spheres. It would appear that most observations of flow behind solid spheres have been made by rigidly securing the sphere and allowing the field fluid to flow by it at a velocity corresponding to some predetermined Reynolds number. Of course, this technique is not possible with deformable liquid drops, and the flow pattern must be observed by allowing them to move through the continuous phase under the influence of the force of gravity. The two techniques are comparable only for motions over a very limited range of small Reynolds numbers. For disturbances which result from a cyclic buildup and release of vorticity, the methods are not identical. The asymmetrical, reactive forces associated with the buildup and detachment of vortex elements combine with the dynamic forces over the leading surface to give the freely falling drop a wobbling or rocking motion. Since the frequency of this motion is identical with the frequency at which vortex elements are shed, it must be assumed that the rocking motion is at least controlled by events taking place near the downstream surface of the deformed drop. The absence of this oscillatory motion in the case of a rigidly held sphere would suggest that minor configuration differences must appear in the wake. The disturbances created on the downstream side of an oscillating drop appear to be geometrically similar to those behind drops of equilibrium shape, but the interdependence of shape changes and shedding mechanisms is difficult to deduce from the photographic evidence.

The dye is scrubbed from the drop by the contiguous layer of water and maps the stream surfaces as it is dispensed into the main stream. The mechanics of these boundary layers has been formulated mathematically, and excellent texts have been published on the subject. At some point on the downstream side of the drop equator the boundary layer separates from the drop surface and continues as a free vortex sheet. The mechanism by which vorticity is transferred from the region of generation to

Fig. 5. The vortex rings formed behind the drop in a Class VI wake are in evidence. Reynolds number 1010.

the field fluid can be inferred from the motions and geometrical changes in this free sheet. Since all mass transferred to the water phase from parts of the drop surface in contact with the boundary layer is carried into the main stream by the free vortex sheet, the flow field which transfers the vorticity also dispenses the mass. The portion of the mass which finds its way into the continuous phase from the section of the drop surface demarcated by the vortex sheet is transported to the main stream by the layer of ambient liquid forming part of the shed vortex element.

WAKE CLASSIFICATIONS

Magarvey and Bishop (*16*) observed and classified wakes behind liquid drops falling through quiescent water and noted the approximate ranges of Reynolds numbers corresponding to the transition from one wake configuration to the next. As identical Reynolds numbers can be attributed to deformed and spherical drops, depending on the physical properties of the systems, the transition ranges are only approximate. The wake classification and corresponding range of Reynolds numbers are given in Table 1.

TABLE 1

Class	Range of Reynolds numbers	Nature of trail
I	0 to 210	Single trail
II	210 to 270	Double trail
III	270 to 290	Double trail with waves
IV	290 to 410	Procession of vortex loops
V	290 to 700	Double row of vortex rings
VI	700 to 2,500	Irregular wake

The Class I wake appears to be the only one in which the configurations behind freely falling drops and rigidly held spheres are identical. In the steady state the boundary layer separates from the drop surface and continues as a free vortex sheet. This sheet converges to a point and dispenses the vorticity into the main stream by means of a single vortex trail. This converging sheet is a stream surface, and the liquid trapped within is dragged along by the drop. The geometry suggests that any mass transferred from the drop to the ambient liquid must find its way into the main stream by means of the vortex sheet and single trail. Little, if any, mass that leaves the drop through the part of the surface demarcated by the sheet ever reaches the main body of the field liquid.

The mechanism by which vorticity is transferred to the main stream by the Class II wake is somewhat different and more complicated than that described above. In the steady state the configuration is characterized by double vortex trails and an intricate spiralling structure immediately behind the drop. Magarvey and MacLatchy (*18*) have considered this type of wake in discussing the disturbances created on the downstream side of liquid spheres. There is a continuous backflow which flushes a restricted area of the back surface of the drop and escapes as one of the spiraled layers which form the double vortex trails. In addition to the mass which is transported by the boundary-layer liquid in contact with the leading surface of the sphere, it is possible for mass to be carried continuously into the main stream by the portion of the backflowing liquid which contacts the back surface of the drop before it escapes the near wake region by way of the vortex trails. Asymmetry as to the trail sizes or differences in their distances from the upstream-downstream axis of the drop results in force components that are responsible for the spiralling trajectory characteristic of drops in this narrow range of Reynolds numbers. The Class III wake is

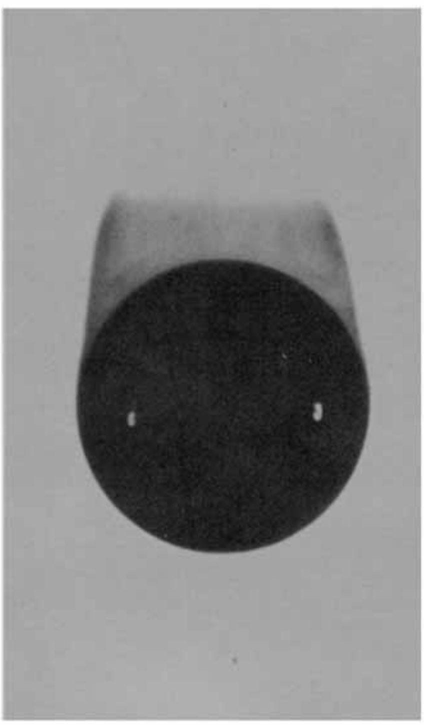

Fig. 6a. Photographs selected from a sequence showing the initial stages in the formation of a wake behind a falling drop. Reynolds number 400.

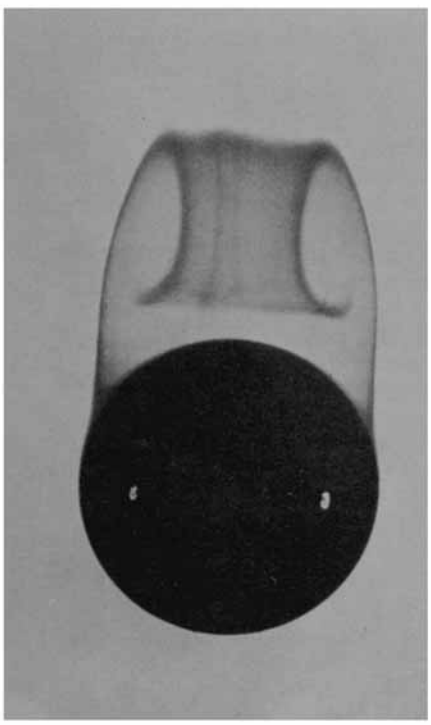

Fig. 6b. Photographs selected from a sequence showing the initial stages in the formation of a wake behind a falling drop. Reynolds number 400.

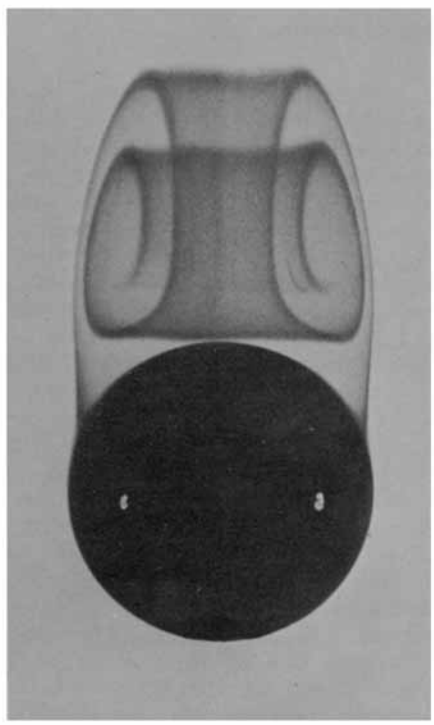

Fig. 6c. Photographs selected from a sequence showing the initial stages in the formation of a wake behind a falling drop. Reynolds number 400.

not essentially different than the Class II in formation and structure. However, the rate at which vorticity is generated and transferred to the near wake region by the involving vortex sheet is such that the two smooth vortex trail mechanism of dispensing it into the main stream is not adequate. Consequently, the balance between generation and dispersion rates is maintained by the periodic escape of small vorticity bursts. These appear as nodules or waves on the double trail wake. These two classes of wakes correspond to part of the range of Reynolds numbers over which Hu and Kintner (*12*) could make no distinction between the drag coefficients of solid spheres and liquid drops. Figure 2 shows the region immediately behind a drop characterized by a Class III wake.

Figure 3 shows several cycles of the Class IV loop wake behind a drop of Reynolds number 350. The vorticity is generated and transferred to the region behind the drop at a much faster rate than it can be carried away by uniform or pulsating trails. The region behind the drop demarcated by the vortex sheet increases in size until it becomes unstable, and the spiraling structure is discharged as a loop. Figures 4*a*, 4*b*, 4*c* and 4*d* are photographs selected from a moving-picture sequence showing different stages in the loop discharge process. It is difficult to assess the efficiency of the mechanism in transferring mass from the drop to the main stream by way of the liquid which makes contact on the downstream surface. An examination of many photographic sequences indicates that backflowing ambient fluid flushes the downstream surfaces only during part of the cycle. Figures 4*a*, 4*b*, 4*c*, and 4*d* show the central tube through which the backflow enters the wake region. As this flow impinges on an element of the area of the drop removed some distance from the axis, the reactive force causes the drop to move sideways. As the elements are built up and shed alternately from diametrically opposite sides of the axis, a slight zigzagging motion is superimposed on the drop trajectory. The Class V wake is more or less a modification of the previous class as far as the formation mechanism is concerned. However, the exact vorticity buildup and release mechanism cannot be inferred from the photographic evidence obtained in these laboratories.

Class VI wakes are not characterized by a regular vorticity buildup and release cycle. It would appear, however, that this is a very efficient means of transferring mass from the drop to the field liquid. The stream surfaces generated by contact with the drop are soon destroyed, and the mass transferred is promptly dispersed over a considerable region of the field liquid. Figure 5 shows a Class VI wake with the many vortex rings which are associated with the irregular wakes.

MASS TRANSFER DURING WAKE FORMATION PROCESS

From an examination of many photographic sequences it is inferred that the rate at which mass is transferred from the drop to the ambient fluid is not very great during the period in which the wake is being established behind the drop. Magarvey and Blackford (*17*) examined the initial motions of a liquid sphere falling from rest through a continuous liquid phase and noted the wake formation process leading to a shedding wake. Figures 6*a*, 6*b*, and 6*c* are photographs from a sequence showing the involution of the vortex sheet immediately preceding the detachment of the first vortex element. It would appear that most mass transferred from the drop during these initial stages of wake formation is carried by the involving vortex sheet. From a consideration of the dye concentration in the wake, it must be concluded that little or no dye is scrubbed from the downstream surface during the period when velocities associated with the backflow are small.

The above discussion of wake processes relative to mass transfer has not included effects of the motions within the drop. This aspect of the problem has been treated by several workers, and its effects as far as terminal velocities and drag coefficients are concerned are implicit in the Reynolds number values to which the wake configurations are correlated. The assumption has been made that the mass to be transferred is present in some quantity at the interface, and no mention is made of the internal circulation as a factor in transporting the mass from within the drop to the interface. Of course, all considerations are for single drops falling through a quiescent phase. For drops falling in close proximity the above wakes would exist in a greatly distorted form or not at all.

ACKNOWLEDGMENT

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NOTATION

C_D = drag coefficient
g = gravitational acceleration
N_p = physical property group
N_{Re} = Reynolds number
N_{We} = Weber number
U = terminal velocity
ρ_d = density of drop phase
ρ = density of continuous phase
σ_i = interfacial tension
$\Delta\rho$ = density difference between drop and field phases

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