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Scavenging of atmospheric particles by water drops

Ming-shian Wu

Department: Chemistry

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This dissertation has been prepared in the style utilized by the Journal of Colloid and Interface Science. Pages 1 - 8 contain the introduction, which has been added for the purpose usual to dissertation writing. Pages 9 - 42 contain the first manuscript and pages 42 - 61, the second manuscript submitted to that journal.

ABSTRACT

In the first paper, the mechanisms of particle capture and coalescence of aerosols by a moving water drop in the atmosphere are studied using the boundary-layer flow approximation. The particle trajectory is computed by solving the equations of motion of the particle both outside and inside the boundary layer using the Adams-Moulton method. The grazing trajectory is found by a trial-and-error technique. The collision and collection efficiencies of scavenging due to particle inertia and to the velocity gradient of the flow field are then computed for water drops ranging from 0.1 to 1.0 mm in radius and for particle of 1 - 10 μ in radius. The results obtained in this work are in good agreement with experimental data given by Walton and Woolcock.

In the second paper, the effects of intermolecular forces on the collection efficiency of submicron aerosol particles are studied. It is assumed that the intermolecular forces provide a certain region as an absorbent surface in the vicinity of the drop. Numerical results have been obtained for the cases of a water drop collecting AgCl aerosols and a water drop collecting submicron cloud droplets. It is found that the collection efficiency depends mainly on the diffusion process. Our calculations agree reasonably well with recent experimental results of Kerker for AgCl aerosols for the case of small drop.

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I. INTRODUCTION

The atmosphere is a dynamic system. It absorbs substances from both natural and man-made sources, digests them and returns them to their proper sinks. If the substances enter the atmosphere faster than they return to their sinks, the contamination of air results. The contaminated air may injure human health and damage properties or, even without producing measurable harm, constitute a nuisance and create a poor living environment. The contamination of the atmosphere has been recognized as a problem since Madame Curie warned the world that radioactive material would pollute the air. Decades have passed by, but the potential hazards remain. Precipitation provides us the only natural means to clean our atmosphere. We are well aware that natural precipitation removes the atmospheric particles in many ways, yet they can be classified into two categories: (a) aerosol particles act as condensation nuclei at the very beginning of the formation of natural precipitation, (b) aerosol particles attach themselves to the natural precipitation by inertial impact, diffusion, molecular attraction, thermophoresis, diffusiophoresis, microturbulence, and electrical interaction, etc..

This dissertation primarily deals with a water drop collecting airborne particles. In the first paper, the effects of inertial impact and velocity gradient on the collection efficiency of large aerosol particles by raindrops falling in the air are investigated. In the second paper, the effect of intermolecular forces on the capture of small aerosol particles by a water drop is studied. A brief review of literature related to the first manuscript will be presented, followed by a review of literature concerning the second manuscript.

Theoretical studies have been made of the effect of inertial impact on filtration, icing on aircrafts, separators and scavenging of airborne particles. The first two important investigations were by Sell (1) and by Albrecht (2). Sell studied the velocity profiles experimentally and calculated the particle trajectories for variously shaped bodies. He showed that the efficiency of inertial impact should be a function of inertial parameters. Using the potential flow equation, Albrecht introduced the particle at 3 radii upstream of the body and assumed the particle velocity at that point to be the same as that of the free stream. These assumptions and conclusions have been the bases for the study of the filtration of aerosol particles by fiber (3,4) and that of aircraft icing (5,6). Golovin and Putnam (7) summarized the foregoing works and concluded that the collection efficiency was dependent on both the inertial parameters and the flow field.

A study based on the inertial impact of rain scavenging is valid only for particles of radius larger than one micron. Theoretical efforts have been mainly directed to it. The result is expressed by the collision efficiency, which is defined as the fraction of particles colliding with the drop to those originally contained in the track of the drop. Because of the difficulty in characterizing the flow field, Langmuir (8) used the limiting cases of potential flow for very high Reynolds number, Re, and of viscous flow for very low Re. He derived an analytical expression for the intermediate Re by fitting his numerical results from the two above cases. Fonda and Herne (9) followed essentially the same procedure and gave the results for intermediate Re region in a chart. Since then, the viscous pattern and the potential approximation have been the methods used for the flow field (10,11).

Although the potential flow approximation provides a good description for the flow near the forward surface of a sphere in high Re region, it may dangerously underestimate the collection efficiency of large particles and overestimate that of small particles. The viscous flow pattern is good only for a very small Re region, but it is not realistic for a freely falling raindrop at its terminal velocity. A different approach is therefore required for the intermediate Re region. The most direct method to take into account both the inertial and viscous effects of the fluid flow is the boundary layer approximation. Tomotika (12) treated the boundary layer of the sphere moving in a uniform fluid by starting from the momentum integral equation and employing both the theoretical and experimental velocity distributions just outside the boundary layer. Because of his explicit and accurate expression, his model will be adopted in this work.

Due to the viscous effect, a spherical particle suspended in a uniform fluid will experience a translational force which can be expressed by Stokes' law if the particle experiences no discontinuous effect. If the particle is so small that its dimension is near the order of the mean free path of the carrying fluid, such a discontinuous effect as the velocity slip on the surface of the particle does exist . The viscous force acting on the particle is then reduced slightly and the inertial parameter of the particle becomes correspondingly larger. A correction factor has been proposed (13,14) and its final form is known as the Knudsen-Weber correction factor (15):

$$C_m = 1 + (\ell/r)\{1.257 + 0.4\exp(1.1r/\ell)\}, \qquad (1)$$

where ℓ is the mean free path of the carrying fluid and r, the radius of

the particle. Due to the velocity gradient of the fluid a particle moving in a viscous flow will also experience a rotational force which, accompanied by the translational force, yields a force orthogonal to the direction of the translational velocity. This velocity gradient effect has been studied (16) and will be considered in the first paper.

A water drop, falling at its terminal velocity, may be deformed from its spherical shape by the combined actions of surface tension, aerodynamic pressure, hydrostatic pressure, electrostatic charge and internal circulation, etc.. It is found (17) that the deformation is very small for drops of radius smaller than 500 μ and the flow pattern inside the drop caused by the viscous force of the fluid is consistent with that of the air outside the drop. Therefore, without producing significant errors, we may assume the drop to be rigid and spherical.

When the particles are within the range of 0.1 - 1.0 μ in radius, the so-called Greenfield gap, theoretical data on the collection efficiency are not readily available. Slinn and Hales (18) recently suggested that diffusiophoresis, thermophoresis and electrical effects might be the principal mechanisms responsible for the collection efficiency of particles in this region. The first two mechanisms have effects on the motion of aerosol particles when there are non-uniformities in the host gas. They may be expected to be important in the wash-out process only when either evaporation or condensation takes place on the surface of the precipitation elements. If the environment around the drop is saturated with water vapor, as in the case of rain, the evaporation and condensation processes are not remarkable. Hence, diffusiophoresis and thermophoresis are of lesser importance in rain scavenging. The electrical force that exists between the drop and the

particle may be an important factor in the actual process of rain scavenging, but further investigations are needed to estimate the magnitude and the sign of the charges of the drop and the particle, and those of electric fields between them. In conjunction with the diffusion process, Zebel (19) studied theoretically the effects of electrical forces on the capture of small particles by water drops falling in a homogeneous electric field, assuming both the drop and the particle to be hard spheres. For the uncharged case, he considered the diffusion process only by using the classical model.

The following items are the main assumptions of the classical model for the capture of the small particles by a collector: 1) The fluid motion is continuous and without slippage in the proximity of the collector. 2) The particle follows the streamlines of the undisturbed fluid around the diffusion boundary layer. 3) The hydrodynamic resistance to the approach of the particle obeys Stokes' law. In fact, there are some shortcomings in this model. As the small particle comes close to the boundary of the collector, the hydrodynamic resistance becomes large due to viscous interaction (20), the long range diffusiophoretic force (21), the slippage of the particle (22) and the difficulty for the volume of fluid between the collector and the particle to drain away (23). On the other hand, if we neglect the foregoing mentioned effects, the hydrodynamic resistance would be lower than that predicted by continuum theory when the gap between the collector and the particle becomes comparable to the mean free path of the fluid molecules. Besides this, the intermolecular forces take their actions in this small gap. Similar to the electrostatic forces, the intermolecular forces may influence the deposition process and keep

the particles from being swept away after the collision.

Molecular forces have been extensively applied to the studies of coagulation (24,25,26), adhesion (26,27) and filtration (27,28) of aerosols. They have been also considered to be important in the washout process, but neither theoretical nor experimental work has been undertaken. This is probably due to the fact that the effects of molecular forces are quite involved and further appropriate assumptions are needed. In the second paper, we shall explore the effects of the intermolecular forces on the collection process of submicron aerosol particles by a water drop. For simplicity, we shall assume that long range intermolecular forces form an absorbent surface near the boundary of the drop, influence the concentration distribution of particles near the absorbent surface and act as a compensation for the effects that increase the hydrodynamic resistance in the proximity of the absorbent surface.

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II. THE AERODYNAMIC CAPTURE OF AEROSOL PARTICLES BY WATER DROPS IN AIR[1,2]

MING-SHIAN WU and JOSEPH T. ZUNG

Department of Chemistry, University of Missouri-Rolla, Rolla, Missouri

ABSTRACT

The mechanisms of particle capture and coalescence of aerosols by a moving water drop in the atmosphere are studied using the boundary-layer flow approximation. The particle trajectory is computed by solving the equations of motion of the particle both outside and inside the boundary layer using the Adams-Moulton method. The grazing trajectory is found by a trial-and-error technique. The collision and collection efficiencies of scavenging due to the particle inertia and to the velocity gradient of the flow field are then computed for water drops ranging from 0.1 mm to 1.0 mm in radius and for particles of 1 - 10 microns in radius. The results obtained in this work are in good agreement with experimental data given by Walton and Woolcock.

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[2]Based on a dissertation submitted by M. S. Wu to the Graduate School of the University of Missouri-Rolla in partial fulfillment of the requirement for the Ph. D. degree in Chemistry.

1. INTRODUCTION

The removal of atmospheric particles by clouds and raindrops, i.e., the precipitation scavenging, plays an increasingly important role in air pollution control and environmental chemistry. It is carried out by two distinct mechanisms: (a) rainout or snowout, by which a particle attaches itself to a cloud droplet either by diffusion or by acting as a condensation nucleus (1), (b) washout, by which a particle is captured by a falling raindrop. In this paper, we are concerned only with the latter case.

The scavenging of aerosol particles, according to Slinn and Hales (2), is a complex process consisting of five microphysical events: Brownian diffusion, thermophoresis, diffusiophoresis, microturbulence, and aerodynamic capture. For particles larger than 5 μ in radius, the capture process is accomplished mainly by inertial impact of the particle on the falling drop. The other processes may become important for submicron particles.

The actual problem of washout is, by its nature, very involved. Consideration should be given to the multiple interactions between drops, between particles, and between drop and particle. To simplify the problem, we consider here the action of only <u>one</u> water drop moving through a cloud of particles. As a water drop falls through an aerosol cloud, it sweeps out a certain volume of air. This displaced mass of air tends to drag the aerosol particles with it. The particles, because of their inertia, are not immediately accelerated to the velocity of the air mass but lag behind so that a portion collide with the drop. The <u>collision efficiency</u> E_1 is then defined as the ratio of the number of particles touching the drop to those whose centers initially lay within

the path of the drop. Depending upon the nature of the surface and molecular forces of both the drop and the particle, a particle, upon hitting the drop, may either be captured or swept away. Hence, not all particles colliding with the drop will be collected. The coalescence efficiency E_2 is defined as the ratio of the number of particles sticking to the surface of the drop and captured by it to those touching the drop. Finally, the collection efficiency E is the ratio of the actual number of particles captured to those initially laying within the track of the drop (3). Obviously, $E = E_1 \times E_2$.

In this paper, we shall study the process of particle capture using the boundary-layer flow approximation, examine the mechanism of coalescence of a particle to a water drop, and propose a method for computing the collection efficiency in the washout of aerosol particles by a moving water drop.

2. THE PROCESS OF PARTICLE CAPTURE

For particles larger than 5 μ in radius, collision with a moving spherical drop is due mainly to inertial impaction, which in itself is a very complex process. Theoretically, one may hope to solve the Navier-Stokes equation for the flow of a viscous fluid round a spherical body, find the velocity profiles (streamlines), and estimate the collision efficiency, etc., but in most of the cases the Navier-Stokes equation is nonlinear and cannot be treated in a rigorous manner (4). This is due to the fact that both inertial and viscous forces act on the sphere simultaneously. When a fluid flows around the drop, the flow pattern generates viscous forces on it and thereby affects its trajectory. On the other hand, the particles, due to their inertia, tend to move across the streamlines instead of being carried by the viscous forces along with the fluid elements, and consequently strike the leading surface of the oncoming drop. Our problem is to estimate the relative importance of inertial and viscous forces in a collision process between a moving drop and a particle immersed in the fluid medium.

If the drop is assumed to be much larger than the particle, the fluid flow pattern is characterized mainly by the flow round the drop and one can neglect the effect of small particles on the flow pattern. Our problem can then be reduced to considering the trajectory of the particle relative to the drop when both inertial and viscous forces act on the small particles. The two following extreme cases are self-evident: (a) if the particle is influenced only by inertia, it will collide with the drop if it were originally laying within the path of the drop, and the collision efficiency E_1 is unity, (b) if the particle is influenced only by viscous forces, it will be carried by the fluid

round the drop, and E_1 is zero. The relative importance of the inertial and viscous forces is measured by the Reynolds number, Re, for the drop, with $Re = DU/\eta$, where D and U are, respectively, the diameter and the velocity of the drop, and the kinematic viscosity η of the fluid whose density is ρ and the coefficient of viscosity is μ is equal to μ/ρ. For large Re, i.e., $Re \gg 1$, the inertial term is predominant and one has a potential flow condition. For small Re, $Re \ll 1$, the inertial forces are negligible and the flow is viscous. In practical problems, however, one has to deal with some intermediate range of Re, when the flow patterns are neither completely viscous nor totally potential. Most of the previous work on this problem was concerned with either the potential flow condition (3,5,6,7) or the viscous flow assumption (6,8, 9). Although the potential flow approximation provides a good description of the flow field near the forward surface of the drop in large Re region, it tends to underestimate the collision efficiency of highly inertial particles and overestimate that of the particles having small inertia. The viscous flow assumption, on the other hand, is not at all realistic for a freely falling raindrop at its terminal velocity (10). For intermediate Re region, one must therefore use the boundary-layer flow approximation to account for both the inertial and viscous effects of the fluid.

Consider a water drop of radius r_1 moving at its terminal velocity U_o relative to a fluid of density ρ_1 and viscosity μ containing monodispersed aerosol particles of radius r_2 and density ρ_2 moving with velocity v. For simplicity, we may assume that the water drop behaves as a rigid sphere, neglecting the deformation and internal circulation (11). Since the velocities involved are much lower than that of sound,

compressibility effects may be neglected. We further assume that the concentrations of both raindrops and aerosol particles are low enough to neglect all interactions between drops and between particles, and that the relative humidity during raining is high enough to warrant a saturated atmosphere and to neglect either condensation and evaporation at the surface of the drop. The thickness of the boundary layer (see Fig. 1) is given by (12)

$$\delta = A(2r_1 U_o \rho_1/\mu)^{-\frac{1}{2}}(\mu/U_\theta^\delta \rho_1), \tag{1}$$

where U_θ^δ is the tangential velocity of the fluid stream neighboring the boundary layer and A is a function of the angle θ between the stagnation point and the given point under consideration. For $\theta < 45°$, $A = 6\theta/\pi$, and eq. (1) becomes

$$\delta/r_1 = (12\theta/\pi)(U_o/U_\theta^\delta)(\mu/2r_1 U_o \rho_1)^{-\frac{1}{2}} = (12\theta/\pi)(U_o/U_\theta^\delta)Re^{-\frac{1}{2}}. \tag{2}$$

The potential flow velocity distribution of the tangential component is given by

$$(U_\theta^\delta/U_o) = (3/2)\sin\theta. \tag{3}$$

Substitution of eq. (3) into eq. (2) yields

$$\delta/r_1 = (8\theta/\pi\sin\theta)Re^{-\frac{1}{2}}. \tag{4}$$

Using l'Hospital rule, one finds the thickness of the boundary layer at the stagnation point

$$(\delta/r_1)_{\theta=0} = (8/\pi)(\theta/\sin\theta)_{\theta=0}Re^{-\frac{1}{2}} = 8Re^{-\frac{1}{2}}/\pi. \tag{5}$$

Velocity Profiles

If it is assumed that the fluid flow can be exhaustively characterized by the axisymmetrical two-dimensional flow field and the flow field

outside the boundary layer is potential, then the velocity components of the fluid outside the boundary layer in polar coordinates are given by

$$U_r/U_o = -\cos\theta[1 - (r_1/r)^3] \tag{6a}$$

$$U_\theta/U_o = \sin\theta[1 + \tfrac{1}{2}(r_1/r)^3], \tag{6b}$$

or, in a cylindrical coordinate system,

$$U_x/U_o = 1 - r_1^3(2x^2 - y^2)/2(x^2 + y^2)^{5/2} \tag{7a}$$

$$U_y/U_o = 3r_1^3 xy/2(x^2 + y^2)^{5/2}. \tag{7b}$$

Inside the boundary layer, we assume that the velocity profiles of the viscous flow have the same shape at different values of θ. When $\delta << r_1$, the tangential velocity component is given by

$$U_\theta = U_\theta^\delta[(3z/2\delta) - \tfrac{1}{2}(z/\delta)^3], \tag{8}$$

where $z = r - r_1$. In order to obtain the radial velocity component U_r, we integrate the continuity equation

$$r^{-2}(\partial/\partial r)(r^2 U_r) + (r\sin\theta)^{-1}(\partial/\partial\theta)(U_\theta \sin\theta) = 0, \tag{9}$$

or,

$$r^2 U_r = -\int_{r_1}^{r} (r/\sin\theta)(\partial/\partial\theta)(U_\theta \sin\theta)\,dr, \tag{10}$$

and impose the boundary conditions, $U_r = 0$ at $r = r_1$, and obtain

$$U_r/U_o = -(1/r)[B(z^4/4 + z^5/20r) + C(\tfrac{1}{2}z^2 + z^3/6r)], \tag{11}$$

where

$$B = [8(\sin\theta - \theta\cos\theta)/\pi\sin\theta Re^{\frac{1}{2}}][(3/4)\delta^{-3}(1 + \delta)^{-4} + (3/2)\delta^{-4} + (3/4)\delta^{-4}(1 + \delta)^{-3}] - (\cos\theta/\delta)[1 + \tfrac{1}{2}(1 + \delta)^{-3}], \tag{12}$$

$$C = -[8(\sin\theta - \theta\cos\theta)/\pi\sin\theta Re^{\frac{1}{2}}][(9/4)\delta^{-1}(1 + \delta)^{-4} + (3/2)\delta^{-2} + (3/4)\delta^{-2}(1 + \delta)^{-3}] + (3\cos\theta/\delta)[1 + \tfrac{1}{2}(1 + \delta)^{-3}]. \tag{13}$$

Equations (7a,b), (8), and (11) will be used to compute the velocity profiles of the fluid around the drop in Section 4.

Equation of motion of the particle

Assuming the drag experienced by the particle in the flow field around the drop to be of Stokesian form and neglecting the disturbance in the flow field caused by the particle itself, one can write down the equation of motion in the form

$$dv/dt = (9/2)[\mu(U - v)/r_2^2\rho_2 C_m] - g, \tag{14}$$

where g is the gravitational constant and C_m is the Knudsen-Weber correction term (8) to account for the discontinuous effects when r_2 is near the order of mean free path of air molecules.

By the process of separation of variables, eq. (14) may be rewritten in the form

$$dv_x/dt = K'(U_x - v_x) - g, \tag{15}$$

$$dv_y/dt = K'(U_y - v_y), \tag{16}$$

where $K' = (9/2)(\mu/r_2^2\rho_2 C_m)$, and v_x and v_y are, respectively, the x- and y- components of particle velocity.

By setting $r_1 = 1$, $U_o = 1$, and $t = r_1/U_o = 1$, eqs. (15) and (16) can be rewritten in a dimensionless form

$$dv_x/dt = K(U_x - v_x) - r_1 g/U_o^2, \tag{17}$$

$$dv_y/dt = K(U_y - v_y), \tag{18}$$

where $K = (9/2)(\mu r_1/r_2^2\rho_2 U_o C_m)$. K^{-1} is the inertial parameter or Stokes' number which gives the ratio of the stop distance that the particle travels when it is introduced into a still fluid at velocity

U_o to the radius of the drop.

Effect of the Velocity Gradient

It is well known that an aerosol particle in the boundary layer experiences a rotational motion due to the velocity gradient of the fluid field. This rotational motion, accompanied by a relative translational velocity, yields a new acceleration force orthogonal to the direction of the translational velocity. The angular velocity due to rotation is given by

$$\Omega = \tfrac{1}{2} \nabla \times U, \tag{19}$$

where only the term $\partial U_\theta / \partial r$ is significant. The acceleration force F_r orthogonal to the direction of the translational velocity $\bar{v}$, according to Rubinow and Keller (13), is

$$F_r = \pi r_2^3 \rho_1 (\Omega \times \bar{v})[1 + O(Re)]. \tag{20}$$

When $\bar{v}$ is very small, O(Re) may be neglected. Furthermore, since Ω is perpendicular to $\bar{v}$ in most practical cases, eq. (20) can be simplified to

$$F_r = \pi r_2^3 \rho_1 \Omega \bar{v}. \tag{21}$$

Using $\bar{v}$ as the relative velocity between the particle and the fluid, i.e., $\bar{v} = v - U$, and substituting eq. (21), eqs. (17) and (18) become

$$dv_x/dt = K(U_x - v_x) - r_1 g/U_o^2 + (3/4) r_1 \rho_1 \Omega (v_y - U_y)/U_o^2 \rho_2, \tag{22}$$

$$dv_y/dt = K(U_y - v_y) - (3/4) r_1 \rho_1 \Omega (v_x - U_x)/U_o^2 \rho_2. \tag{23}$$

These are the equations of motion where the effect of velocity gradient has been taken into account.

3. THE MECHANISM OF COALESCENCE

So far we have derived the equations of motion of particle around the drop. These equations will be solved to yield the collision efficiency, yet, not all collisions result in effective collection by the drop. The second mechanism, that of coalescence , determines whether a colliding particle will effectively stick to the drop or be swept away or, in some instances, go through the drop, dependent on the nature of the surface forces and the other characteristics, such as, wettability and impact velocity, etc., of the particle.

After Pemberton (14) had studied the penetration process for the rain scavenging of nonwettable particles, McDonald (15) extended the theory of penetration to the partial wettable particles. He considered the particle to be collected only when it penetrated completely into the water drop. As shown in Fig. 2, the work W done against the surface tension T by the particle with the depth of penetration h and the contact angle β can be expressed by

$$W = \int_0^h 2\pi r_2 T \sin\alpha \sin(\beta + \alpha) dh, \tag{24}$$

or, in terms of the penetration angle α,

$$W = \int_0^\alpha 2\pi r_2^2 T \sin^2\alpha \sin(\beta + \alpha) d\alpha. \tag{25}$$

The work of full penetration W_T is obtained by integrating eq. (25) from 0 to π, the result is

$$W_T = (8/3)\pi r_2^2 T \cos(\pi - \beta). \tag{26}$$

If the particle of mass m possesses a radial kinetic energy, $\frac{1}{2} m v_r^2$, just before it collides with the drop, the remaining energy after collision and full penetration is

$$E = \tfrac{1}{2}mv_r^2 - (1 + m/M)W_T, \tag{27}$$

where M is the mass of the drop. The factor (1 + m/M) takes into account the momentum loss to the drop during collision. The work W_T is positive when $\beta > 90°$ (the particle loses energy after full penetration), yet W_T is negative when $\beta < 90°$ (the particle gains energy from the penetration process). Since in most cases $m << M$, the particle velocity after full penetration becomes

$$v_r^* = (v_r^2 - 2W_T/m)^{\frac{1}{2}}. \tag{28}$$

There is no net tangential force during the penetration, therefore, the tangential velocity v_θ of the particle after penetration remains unchanged. We now assume that inside the drop a particle moves according to Stokes' law and that the effects of gravity and internal circulation can be neglected. The equations of motion of the particle inside a steady state flow of the drop are

$$mdv_x/dt = - 6\pi\mu_w r_2 v_x, \tag{29}$$

$$mdv_y/dt = - 6\pi\mu_w r_2 v_y, \tag{30}$$

where μ_w is the viscosity of water inside the drop. Since the trajectory of the particle inside the drop is linear if $r_2 << r_1$, eqs. (29) and (30) can be simplified to

$$mdv_x/dx = - 6\pi\mu_w r_2, \tag{31}$$

$$mdv_y/dy = - 6\pi\mu_w r_2. \tag{32}$$

Inside the drop a particle has a chance to escape only if it fulfills one of the two following conditions: (a) $\beta > 90°$ and the particle reaches the other end of the drop surface, (b) $\beta \leq 90°$, the particle

reaches the other end of the drop surface and still possesses a radial kinetic energy equal to or greater than the minimum energy required for the particle to overcome the surface tension, $-W_T$. Hence, the following boundary conditions for eqs. (31) and (32) are used.

(A) Initial Conditions: $x_i = -r_1$, $y_i = 0$; $v_{xi} = v_r^*$, $v_{yi} = v_\theta$;

(B) Final Conditions:

case (a) $x_f < 2r_1 v_r^{*2}/(v_r^{*2} + v_\theta^2) - r_1$; $y_f < 2r_1 v_r^* v_\theta/(v_r^{*2} + v_\theta^2)$;

$$v_{xf} = 0, \quad v_{yf} = 0; \tag{33}$$

case (b) $x_f = 2r_1 v_r^{*2}/(v_r^{*2} + v_\theta^2) - r_1$; $y_f = 2r_1 v_r^* v_\theta/(v_r^{*2} + v_\theta^2)$;

$$v_{xf} < (-2W_T/m)^{\frac{1}{2}}; \quad v_{yf} < (-2W_T/m)^{\frac{1}{2}} v_\theta/v_r^*,$$

where, as shown in Fig. 3, the x- coordinate is the line connecting the point of impact on the drop surface and the center of the drop. Integrating eqs. (31) and (32), we obtain the criteria for preventing the particle from escaping after full penetration into the drop:

$$\text{case (a)} \quad v_\theta^2 < 12\pi r_1 r_2 \mu_w v_r^*/m - v_r^{*2}; \tag{34}$$

$$\text{case (b)} \quad v_\theta^2 < \{v_r^{*2}[12\pi r_1 r_2 \mu_w/m + (-2W_T/m)^{\frac{1}{2}}] - v_r^{*3}\}/[v_r^* - (-2W_T/m)^{\frac{1}{2}}]. \tag{35}$$

4. METHODS OF CALCULATION OF THE COLLECTION EFFICIENCY

To calculate the collection efficiency, we must compute the particle trajectory in the fluid field. We first examine the trajectory of the particle far upstream. By series expansion, eqs. (7a) and (7b) may be rewritten in a dimensionless form,

$$U_x = 1 + x^{-3} - 3y^2x^{-5} + (45/8)y^4x^{-7} - (35/4)y^6x^{-9} + O(y^8x^{-11}); \quad (36)$$

$$U_y = (3/2)yx^{-4} - (15/4)y^3x^{-6} + (105/16)y^5x^{-8} - (16540/1680)y^7x^{-10} + O(y^9x^{-12}). \quad (37)$$

The equations of motion far upstream can be expressed by

$$v_x dv_x/dx = K(U_x - v_x - \varepsilon), \quad (38)$$

$$v_x dv_y/dx = K(U_y - v_y), \quad (39)$$

provided that the particle motion is in steady state and the terms $\partial v_x/\partial y$ and $\partial v_y/\partial y$ are very small. ε is equal to $(2/9)gr_2^2\rho_2C_m/\mu U_o$. The solutions of eqs. (36-39) are

$$v_x = 1 - \varepsilon + x^{-3} + 3(1 - \varepsilon)x^{-4}K^{-1} + 3x^{-5}[4(1 - \varepsilon)^2K^{-2} - y^2] + 15x^{-6}K^{-1}[4(1 - \varepsilon)^2K^{-2} - y^2](1 - \varepsilon) + 3x^{-7}K^{-1}\ 1 + 30(1 - \varepsilon)^2[4(1 - \varepsilon)^2K^{-2} - y^2] + (45/8)y^4x^{-7} + O(x^{-8}), \quad (40)$$

$$v_y = (3/2)yx^{-4} + 6yx^{-5}K^{-1}(1 - \varepsilon) + 15yx^{-6}[2K^{-2}(1 - \varepsilon)^2 - y^2/4] + 90yx^{-7}K^{-1}(1 - \varepsilon)[2K^{-2}(1 - \varepsilon)^2 - y^2/4] + O(x^{-8}), \quad (41)$$

$$y = \exp\{\ln y_o - \tfrac{1}{2}x^{-3} - (3/2)x^{-4}K^{-1}(1 - \varepsilon) - 3x^{-5}[2K^{-2}(1 - \varepsilon)^2 - y_o^2/4] - 15x^{-6}K^{-1}(1 - \varepsilon)[2K^{-2}(1 - \varepsilon)^2 - y_o^2/4] - \exp(v_x) + O(x^{-7}), \quad (42)$$

where y_o is the y- coordinate of the particle in the undisburbed region (i.e., at very far upstream).

The above predictions of particle trajectories and velocities at far upstream converge very well. However, for particles with large inertial parameters, we fail to obtain convergence at x = -6 region where we start our numerical calculations. By investigating the physical significance of the inertial parameter K^{-1} in the y- direction, one finds: $v_y = U_y$ when $K^{-1} \to 0$, and $v_y = 0$ when $K^{-1} \to \infty$. These limiting cases suggest that the initial conditions for the region outside the boundary layer should be of the form

$$v_x = 1 - \varepsilon - (U_x - 1)\exp(-1/2K) \tag{43a}$$

$$v_y = U_y \exp(-1/2K); \tag{43b}$$

$$y = \exp[\ell n\, y_o - \tfrac{1}{2}x^{-3}\exp(-1/2K)] \tag{44a}$$

$$x = -6, \tag{44b}$$

where the factor $\frac{1}{2}$ in the exponential terms is obtained by comparison with the values calculated from eqs. (40-42) for particles with small inertial parameters.

The initial conditions for the region inside the boundary layer are furnished by the results obtained from solutions of eqs. (17-18) for the region outside the boundary layer, and the initial conditions for the region inside the drop are obtained from solutions of eqs. (22-23) for inside the boundary layer.

The boundary conditions for the particle to enter the boundary layer are

$$x = -(1 + \delta)\cos\theta \tag{45a}$$

$$y \leq (1 + \delta)\sin\theta, \tag{45b}$$

and to penetrate the drop,

$$r \leq 1. \tag{46}$$

The equations of motion of the particle outside the drop are solved by a computer program using the Adams-Moulton method (16). This method requires four consecutive sets of variables, of which the first is given by eqs. (43-44), and the other three are obtained from the preceding set by Taylor's series expansion. Computation is carried out step-by-step until the particle reaches the boundary conditions of eqs. (45a,b). If the interception point does not coincide with the boundary line, a new set of x, y, v_x and v_y is computed by a method of backward interpolation satisfying the conditions

$$(x_n - x)/(y_n - y) = (x_n - x_{n-1})/(y_n - y_{n-1}), \tag{47}$$

$$x^2 + y^2 = (1 + \delta)^2. \tag{48}$$

The same method of interpolation is used to obtain the exact point of interception on the drop surface.

During the trajectory calculation, if y becomes greater than $(1 + r_2/r_1)$, then no impact takes place and the particle is assumed to escape. The value of y_o of the escaping trajectory provides the upper-bound y_u for the y_o of the grazing trajectory, while the y_o of the impact trajectory provides the lowerbound y_ℓ. The true y_o of the grazing trajectory is then calculated by iteration procedure using the values of y_u and y_ℓ, and with the condition that the quantities $(y_o - y_\ell)$ and $(y_u - y_o)$ reach an acceptable error. The obtained value is accepted as the radius of collision cross-section Y_o. The collision efficiency E_1 is then calculated by the equation

$$E_1 = [Y_o/(1 + r_2/r_1)]^2. \tag{49}$$

In the case of $\beta > 90°$, the minimum impact velocity $v_{r,min}$ required for the partilce to fully penetrate into the drop is, as mentioned in the last section,

$$v_{r,min} = (2W_T/m)^{\frac{1}{2}}, \tag{50}$$

where W_T is given by eq. (26). Hence, the particles with v_r less than $v_{r,min}$ will be swept away and escape after collision. When $\beta = 90°$ and W_T is negative, the particle will then enter the drop upon impact without requiring any minimum impact velocity.

Inside the drop, we assume that the fluid field velocity is zero. The conditions given by eqs. (34) and (35) are used to determine whether the particle will stay inside the drop or go through the drop and escape.

In order to compute the collection efficiency E, we plot the impact velocity v_r against y_o at a given contact angle, and select the value of y_o that corresponds to the impact velocity that equals $v_{r,min}$. The selected y_o is then used as the radius of the collection cross-section in the computation of the collection efficiency.

5. RESULTS AND DISCUSSION

Comparing eqs. (6a) and (11), we find that there is a discontinuity in radial velocity of flow between the potential flow field and the boundary layer. However, it is unlikely that such a discontinuity can exist in the neighborhood of the stagnation point. Furthermore, the tangential velocity near the boundary layer obtained from eq. (6b) is too small compared with the experimental results, i.e., around $\theta = 70°$ the tangential velocity just outside the boundary layer should be about $1.1 \sim 1.25\ U_o$ (12). This deviation becomes larger for smaller drops. In order to correct these shortcomings, a model is adopted (shown in Fig. 1), where, instead of the radius of the drop, the radius r_3 of an imaginary region is used to compute the potential flow field. The value of r_3 is obtained by matching the results calculated from eq. (11) and eq. (6a) at $\theta = 0.01$ and $r = 1 + \delta$.

The radius of collision cross-section Y_o has been computed from the trajectory of particles of 1 - 10 μ and for drops of 0.01 - 0.1 cm in radius. Some representative curves of Y_o vs. r_2 are shown in Fig. 4. Table I shows the collision efficiency E_1 obtained from the values of Y_o for particles ranging from 2 μ to 10 μ in radius. These values of E_1 are plotted against the drop radius r_1 in Fig. 5.

Compared with the potential flow model (3, 6, 7, 9), we find two distinct features of the fluid field that strongly influence the particle trajectories inside the boundary layer: (a) the tangential velocity that tends to sweep the particle away from the drop is smaller inside the boundary layer than in the potential flow field, thereby, increasing the possibility for a particle to reach the drop surface, (b) the radial velocity that tends to drive the particle to the drop is also smaller,

the particle then takes a longer time to reach the drop surface and experiences the sweeping force of the tangential fluid velocity for a longer duration, hence, the particle is less likely to be captured. Our results show that the first factor is more important for the larger particles while the second factor is predominant for the smaller ones. In the viscous flow approximation (7,8,9), the streamlines are too spread out around the front face of the drop, making it more difficult for the particle to reach the drop. Thus, the collision efficiency obtained by the viscous flow method is much smaller than our results.

Table II shows the contribution of velocity gradient effect to the collision efficiency. The magnitude of this contribution is proportional to three competitive factors: angular velocity, relative velocity and duration time of the particle. Because of the short duration, the velocity gradient effect in this work does not show much significance. However, it may be important to wake capture on the backside of the drop.

In the case of nonwettable or partially wettable particles, the roles of the contact angle and surface tension become important. Such particles must possess a minimum impact velocity to be effectively captured by the water drop. Figure 6 shows the minimum impact velocity for full penetration as a function of contact angle. To take into account the effect of contact angle on the collection efficiency, we plot in Fig. 7 the impact velocity against y_o, representing the y-coordinate of the particle very far upstream, and by graphical methods select the value of y_o corresponding to minimum impact velocity. Since in our calculations the conditions given by equations (34) and (35) are always satisfied, the particle after entering the drop will not have enough energy to go through the drop and escape. The selected

value of y_o may be used directly to compute the collection efficiency and the results are given in Table III. Compared with the previous calculations (15), our results must be more reliable for they are computed from more accurate impact velocities.

In Fig. 8 - 10, the collision efficiencies are compared with the experimental results of Walton and Woolcock (17) along with the theoretical predictions given by Langmuir (18) and by Fonda and Herne (17). By fitting the results obtained from limiting cases of potential flow for very high Re region and viscous flow for very low Re region, Langmuir derived an analytical expression of collision efficiency for the intermediate Re region. Fonda and Herne followed essentially the same procedure, but instead of giving an analytical expression, they presented their calculations graphically. Compared with both predicted values, our calculations show a better agreement with experimental data, particularly for smaller drops. The improvement is believed to derive from our more direct, accurate and realistic method.

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TABLE I

CALCULATED VALUES OF COLLISION EFFICIENCIES

Drop Radius r_1 (cm)	Particle Radius r_2 (μ) 2	3	4	5	7	10
0.01	0	0.094	0.330	0.494	0.694	0.836
0.02	0.014	0.317	0.515	0.646	0.796	0.893
0.03	0.084	0.360	0.548	0.671	0.811	0.902
0.04	0.105	0.370	0.553	0.675	0.813	0.902
0.05	0.111	0.368	0.550	0.672	0.811	0.901
0.08	0.103	0.348	0.530	0.654	0.797	0.893
0.10	0.094	0.332	0.514	0.625	0.788	0.887

TABLE II

THE CONTRIBUTION OF VELOCITY GRADIENT

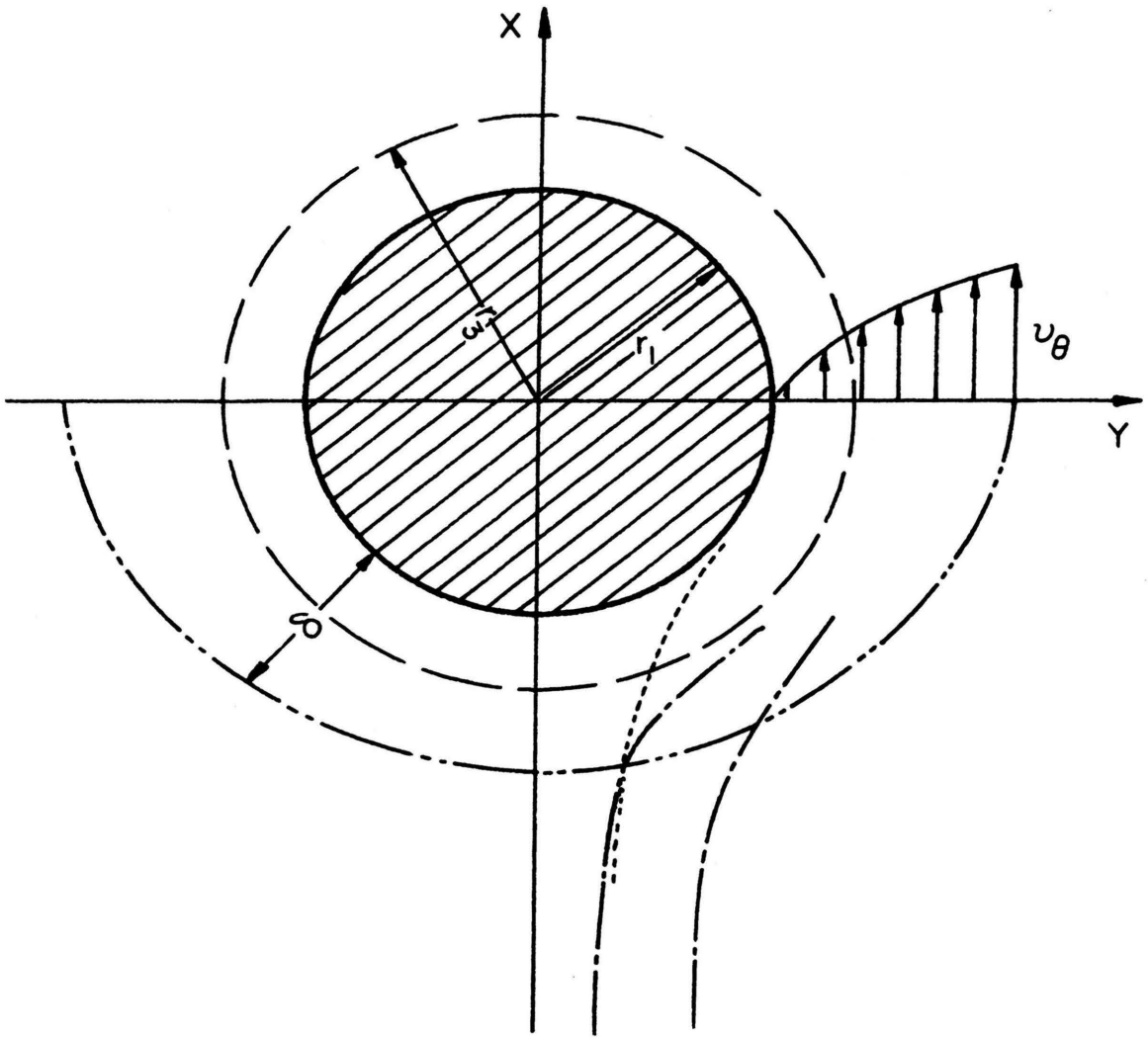
TO COLLISION EFFICIENCY ($\Delta E_1 \times 10^4$)

Drop Radius r_1 (cm)	Particle Radius r_2 (μ)					
	2	3	4	5	7	10
0.01	0	0.79	2.52	3.07	3.63	3.73
0.05	2.14	3.00	3.05	3.17	2.47	1.57
0.10	2.42	2.72	3.00	3.25	3.18	2.78

TABLE III

COLLECTION EFFICIENCIES E FOR PARTICLE OF RADIUS 10 μ

Drop Radius r_1 (cm)	Contact Angle β				
	108°	126°	144°	162°	180°
0.03	0.190	0	0	0	0
0.04	0.343	0.002	0	0	0
0.05	0.464	0.132	0	0	0
0.06	0.559	0.242	0.031	0	0
0.07	0.607	0.321	0.133	0.032	0
0.08	0.632	0.393	0.244	0.120	0.061
0.09	0.653	0.446	0.296	0.194	0.134
0.10	0.671	0.487	0.336	0.245	0.203



DROP

BOUNDARY LAYER PROFILE

FLOW STREAMLINE

PARTICLE TRAJECTORY

IMAGINARY BODY PROFILE FOR POTENTIAL FLOW FIELD

Fig. 1

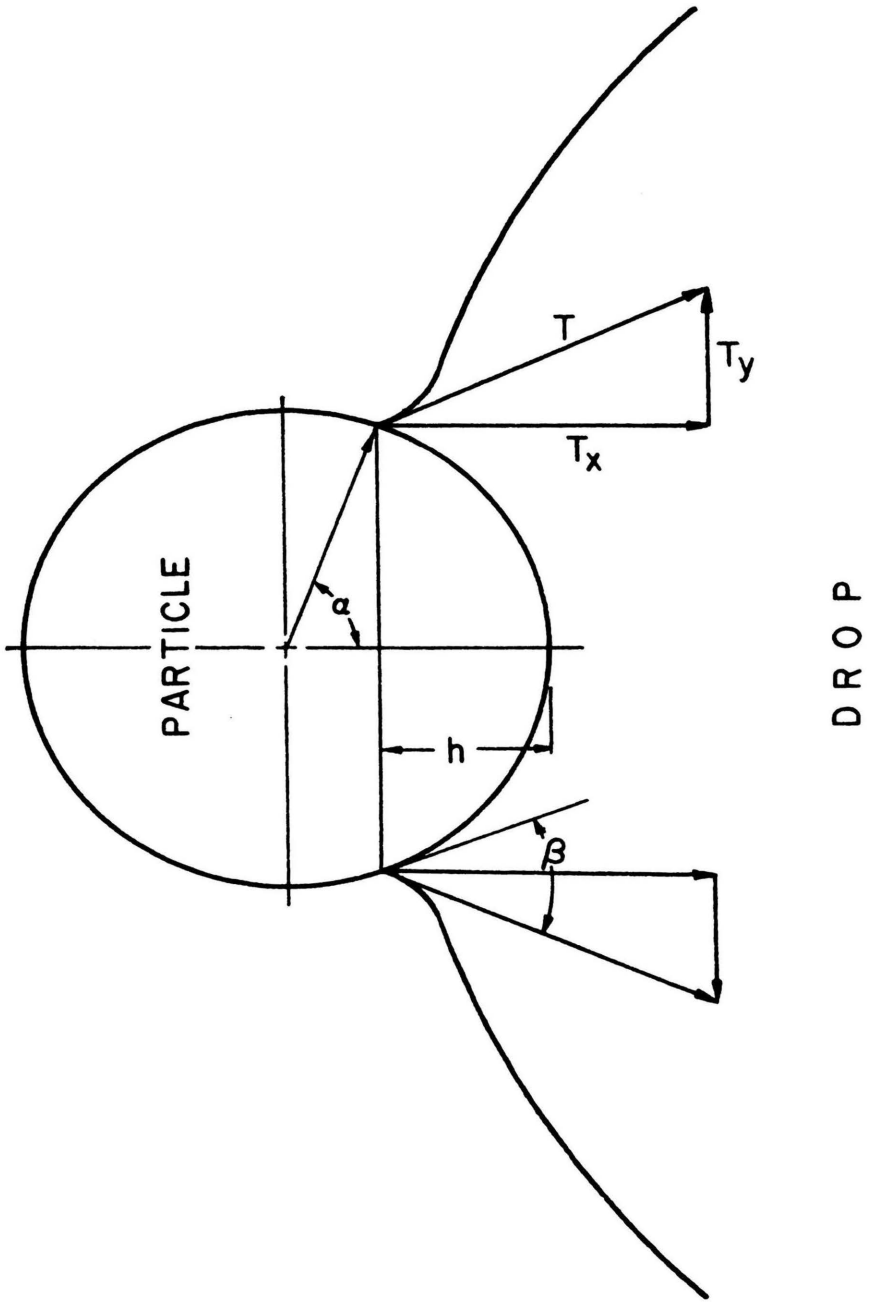


Fig. 2

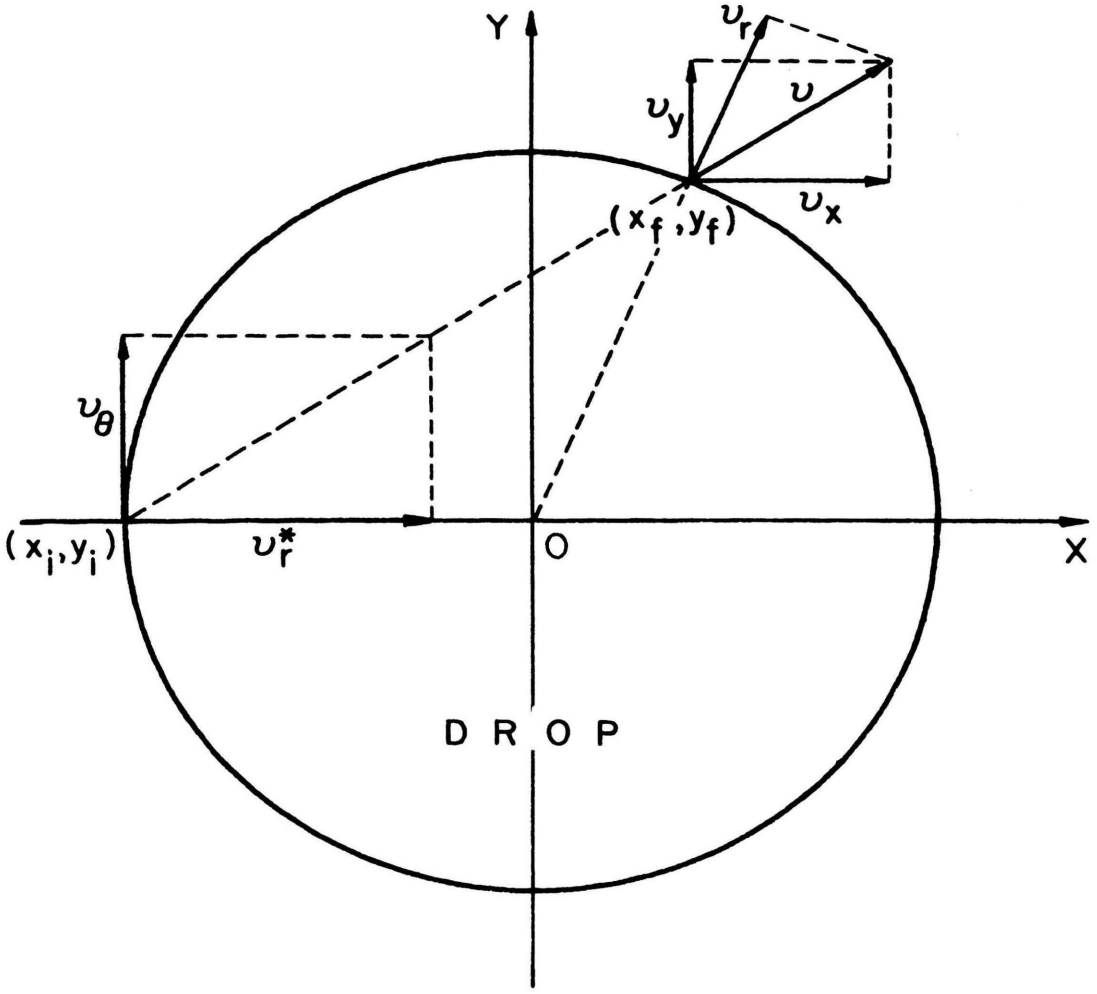


Fig. 3

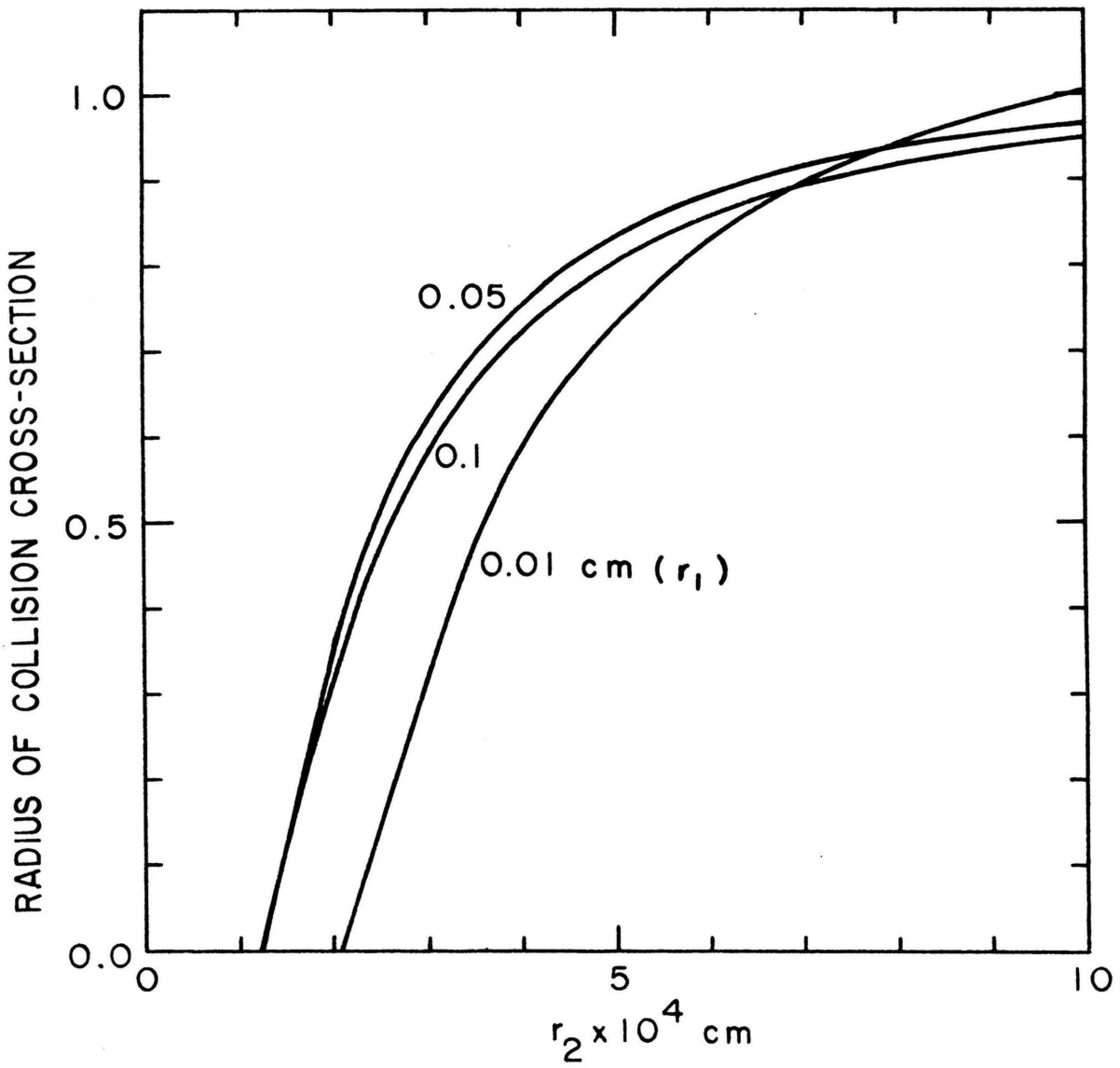

RADIUS OF COLLISION CROSS-SECTION
1.0
0.5
0.0
0
5
10
$r_2 \times 10^4$ cm
0.05
0.1
0.01 cm (r_1)

Fig. 4

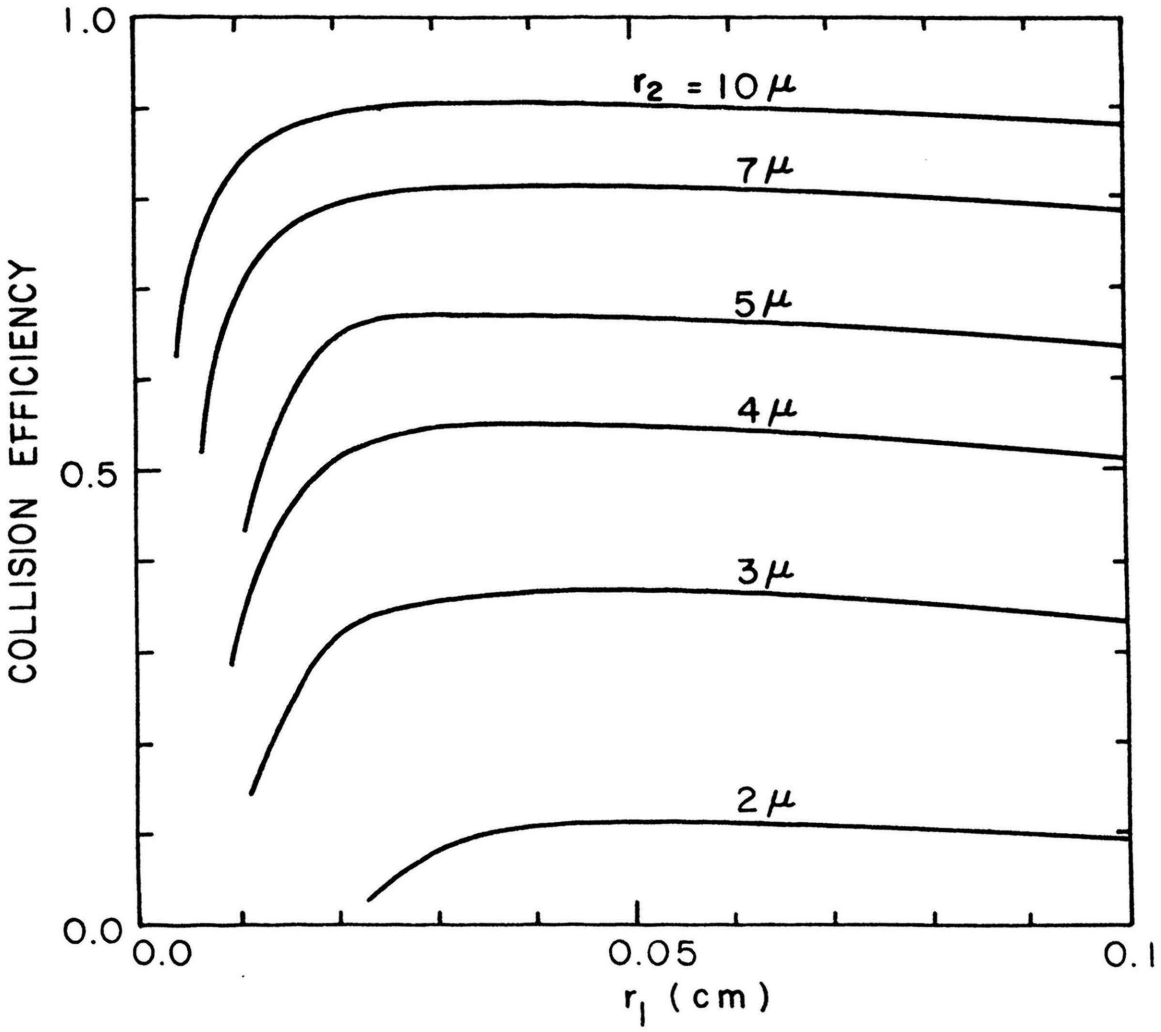

1.0
0.5
0.0
COLLISION EFFICIENCY
$r_2 = 10\mu$
7μ
5μ
4μ
3μ
2μ
0.0
0.05
0.1
r_1 (cm)

Fig. 5

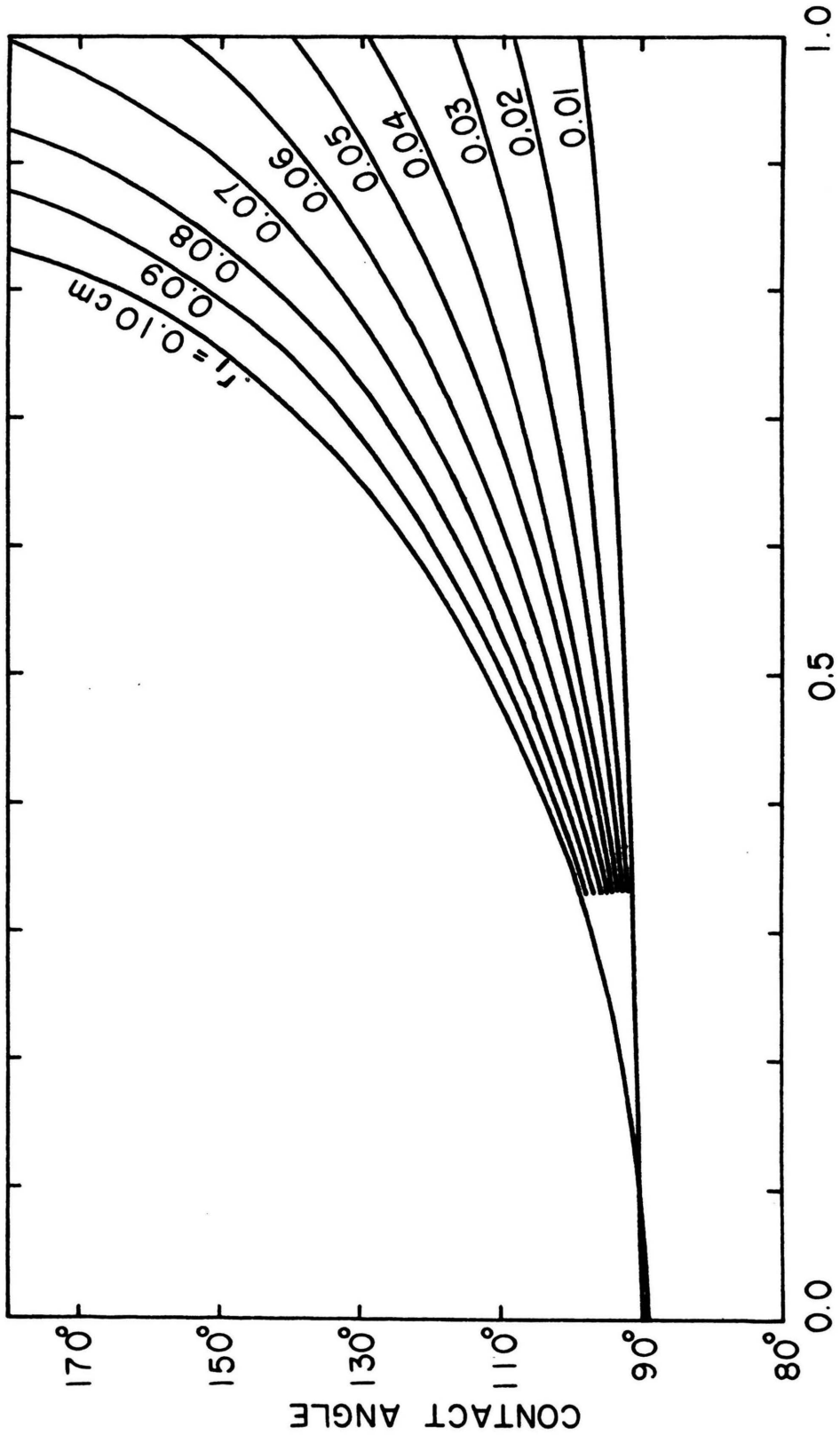

CONTACT ANGLE
170°
150°
130°
110°
90°
80°
0.0
0.5
1.0
v_r / U_o
$r_1 = 0.10$ cm
0.09
0.08
0.07
0.06
0.05
0.04
0.03
0.02
0.01

Fig. 6

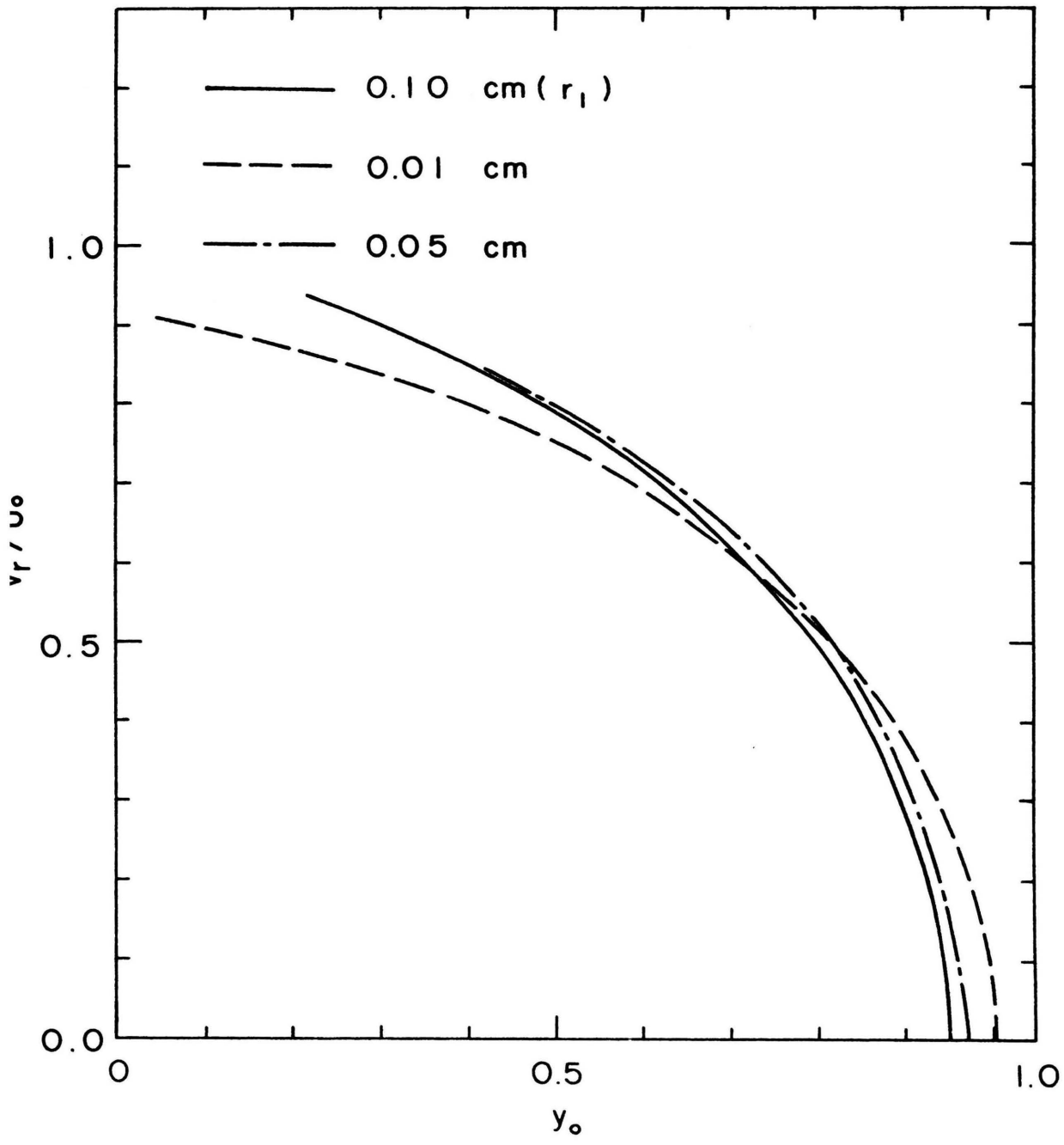


Fig. 7

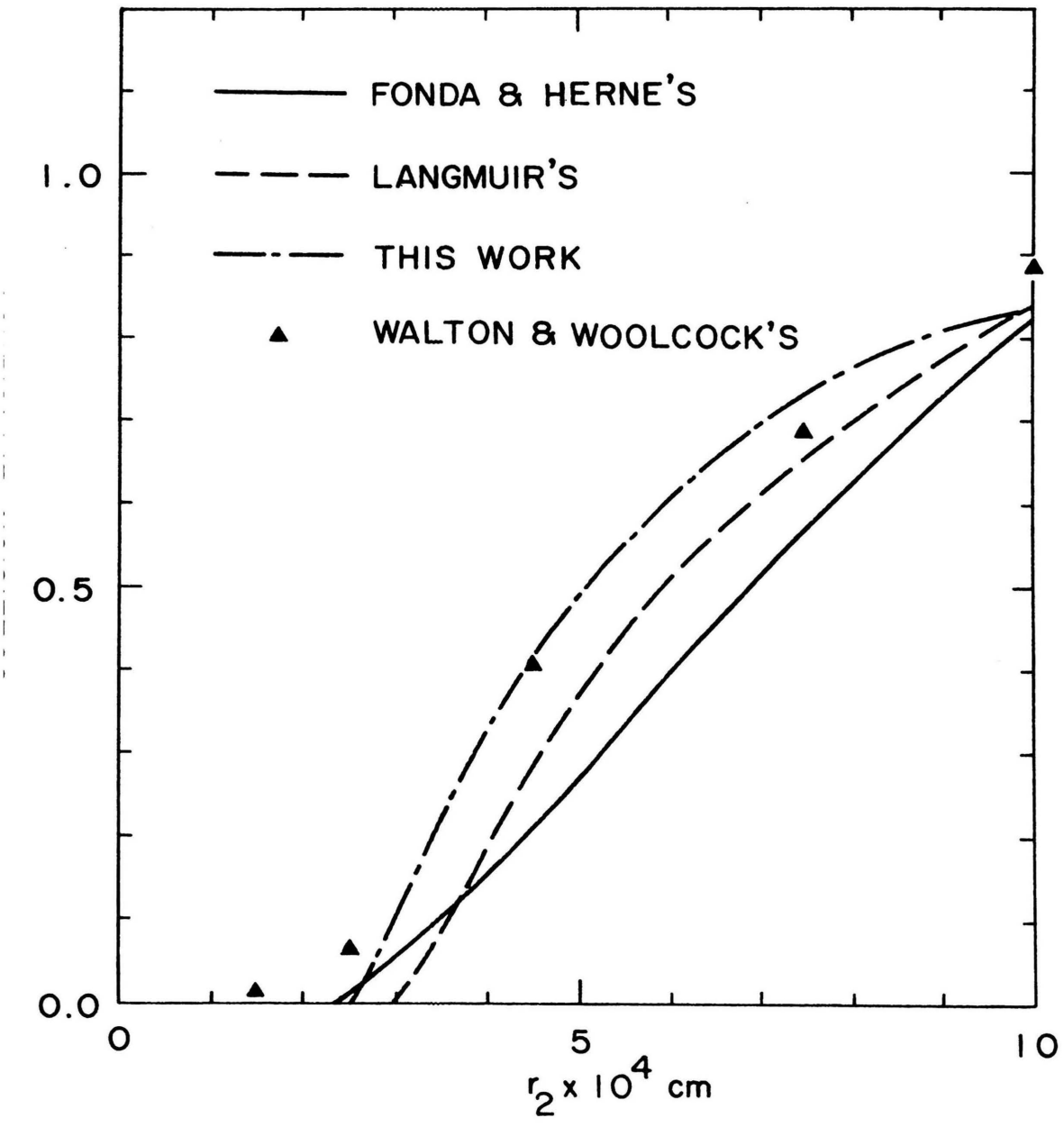

FONDA & HERNE'S
LANGMUIR'S
THIS WORK
WALTON & WOOLCOCK'S
1.0
0.5
0.0
0
5
10
$r_2 \times 10^4$ cm

Fig. 8

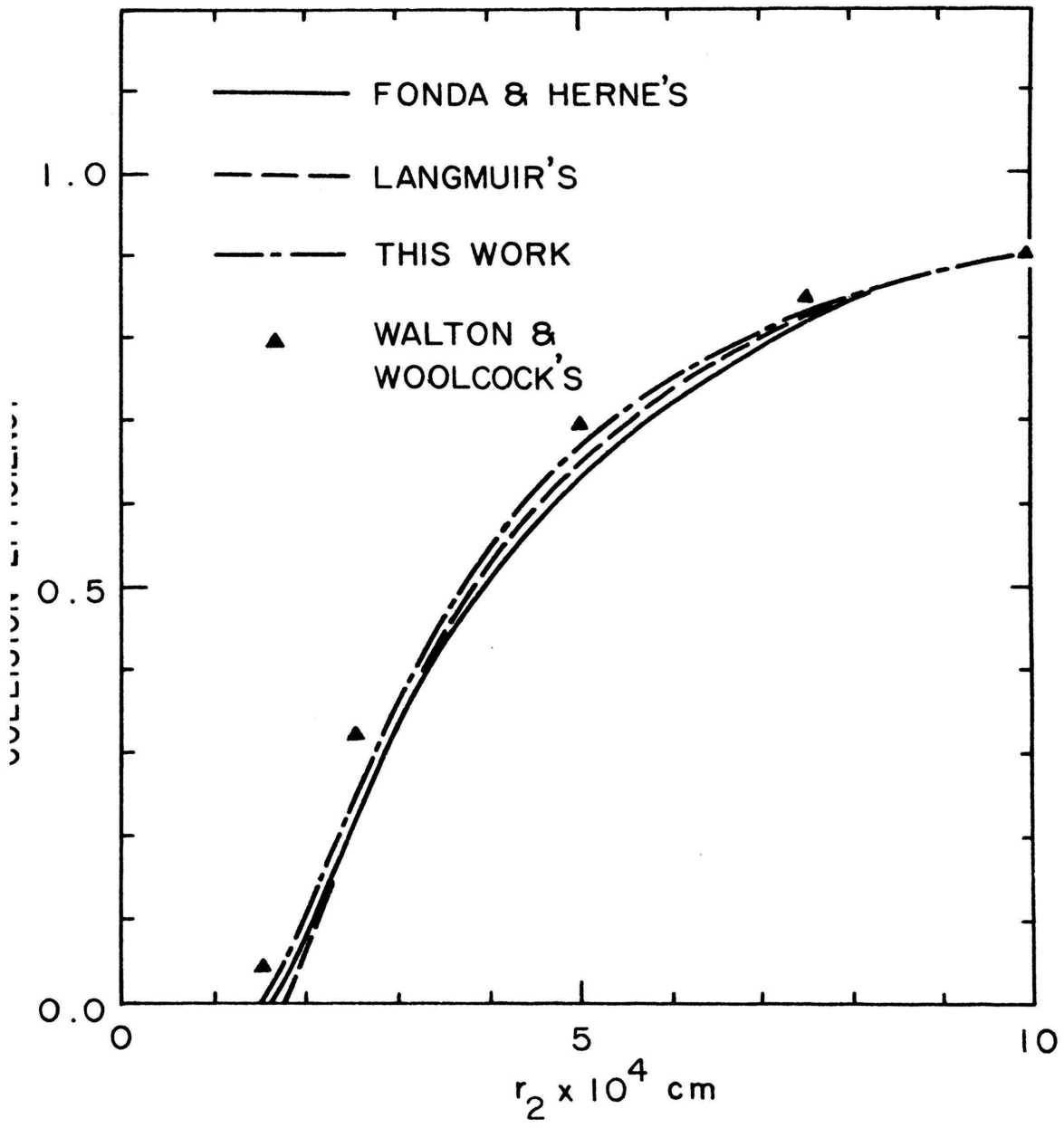

FONDA & HERNE'S
LANGMUIR'S
THIS WORK
WALTON & WOOLCOCK'S
1.0
0.5
0.0
0
5
10
$r_2 \times 10^4$ cm

Fig. 9

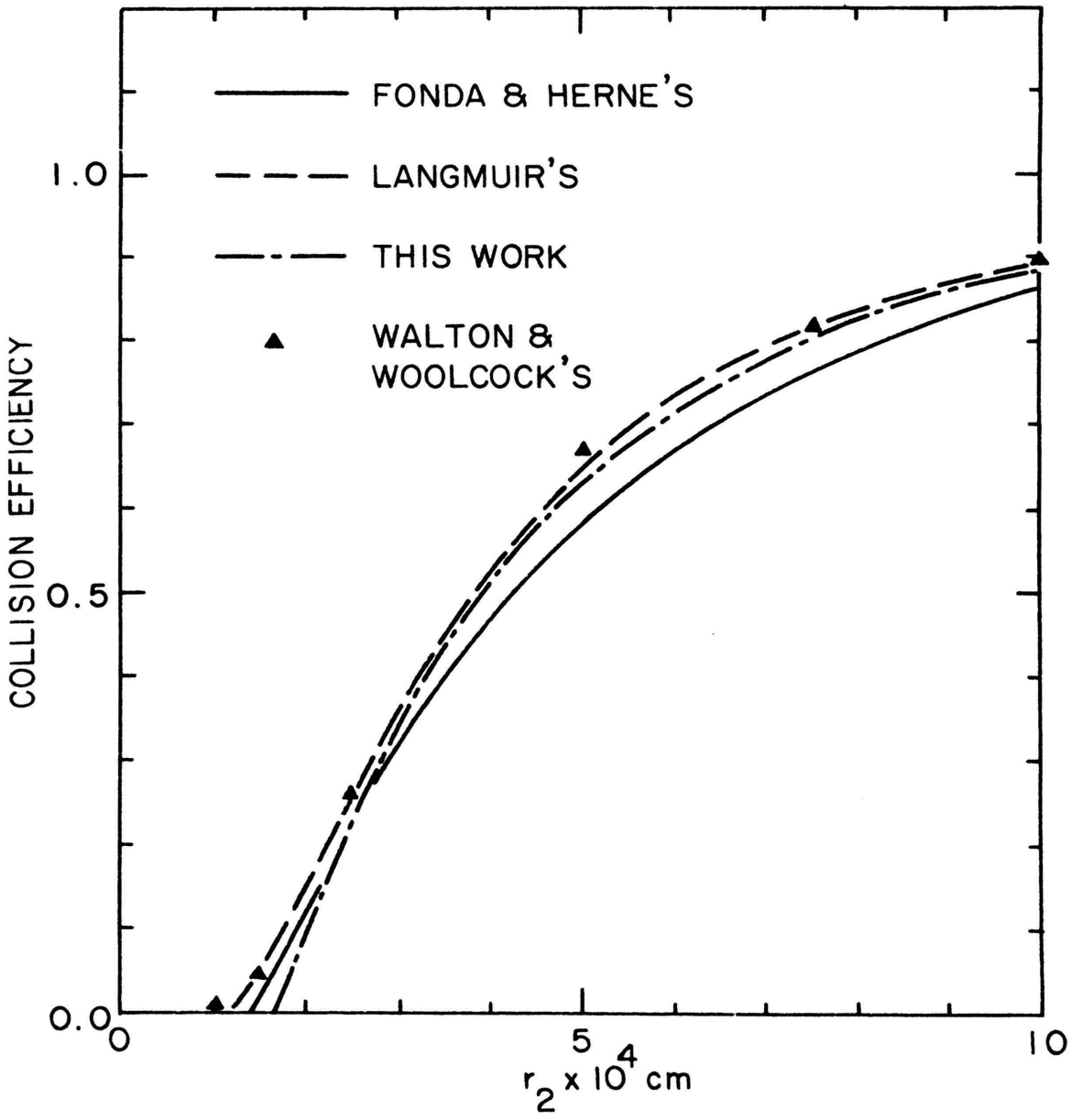

FONDA & HERNE'S
LANGMUIR'S
THIS WORK
WALTON & WOOLCOCK'S
COLLISION EFFICIENCY
1.0
0.5
0.0
0
5
10
r2 x 10^4 cm

Fig. 10

III. THE ROLE OF MOLECULAR FORCES IN THE SCAVENGING OF AEROSOL PARTICLES[1,2]

MING-SHIAN WU and JOSEPH T. ZUNG

Department of Chemistry, University of Missouri-Rolla, Rolla, Missouri

ABSTRACT

The effects of intermolecular forces on the collection efficiency of submicron aerosol particles are studied. It is assumed that the intermolecular forces provide a certain region as an absorbent surface in the vicinity of the drop. Numerical results have been obtained for the cases of a water drop collecting AgCl aerosols and a water drop collecting submicron cloud droplets. It is found that the collection efficiency depends mainly on the diffusion process. Our calculations agree reasonably well with recent experimental results of Kerker for AgCl aerosols for the case of small drops.

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[2]Based on a dissertation submitted by M. S. Wu to the Graduate School of the University of Missouri-Rolla in partial fulfillment of the requirement for the Ph. D. degree in Chemistry.

1. INTRODUCTION

In our previous analysis (1) the contribution to the collection efficiency of submicron particles due solely to their inertial impact on the water drop was shown to be essentially zero. Moreover, the contribution to the collection efficiency attributed to the Brownian diffusion process becomes important only when the particle radius is less than 0.1 μ. For particles within the range 0.1 - 1.0 μ in radius, the so-called Greenfield gap, there is no sound theoretical method for computing the collection efficiency. Slinn and Hales (2) recently studied the effects of diffusiophoresis on the scavenging of particles in this region. Zebel (3) considered the diffusion process and electrical effect as the principal mechanisms responsible for the collection efficiency of charged particles by a drop falling in an electric field. For uncharged particle he considered the diffusion process only by using the classical model.

It is believed, however, that the classical model has some shortcomings. On the one hand, as the small particle comes close to the boundary of the drop, the hydrodynamic resistance becomes large due to the viscous interaction (4), the long range diffusiophoretic force (5), the slippage of the particle (5), and the difficulty for the volume of fluid between the drop and the particle to drain away (6). On the other hand, if we neglect the foregoing mentioned effects, the hydrodynamic resistance would be lower than that predicted by continuum theory when the gap between the drop and the particle becomes comparable to the mean free path of the fluid molecules. Besides this, the intermolecular forces take their actions in this small gap.

The effects of the intermolecular forces have been extensively studied in connection with such important environmental problems as

coagulation of aerosol particles (7-9), adhesion of dusts and powders (9-10), and air filtration (10-11). It is also known that intermolecular forces play an important role in the scavenging of submicron aerosol particles by water drops. Despite the importance of the problem, neither theoretical nor experimental data are at present readily available.

In this paper, we shall explore the effects of the intermolecular forces on the collection process of submicron aerosol particle by a water drop. For simplicity, we shall consider that the long range intermolecular forces form an absorbent surface near the boundary of the drop and act as a compensation for the effects that increase the hydrodynamic resistance in the proximity of the absorbent surface.

2. THE NATURE OF INTERMOLECULAR FORCES

The long range intermolecular forces are of the following types (12): induction, orientation, and dispersion. For two macrosystems of condensed bodies, calculations (10,12) showed that induction and orientation forces were of little significance, while dispersion forces were important.

Assuming that the special molecular arrangement on the surface can be ignored, and the attraction forces between two molecules separated by a distance r is of the Lennard-Jones type, the potential energy U_E is given by

$$U_E = - \lambda / r^6, \tag{1}$$

where λ is a constant dependent upon the characteristics of the two interacting molecules. If only the dispersion force is significant, λ is of the form

$$\lambda = (3/2) h \nu_{o1} \nu_{o2} \alpha_1 \alpha_2 / (\nu_{o1} + \nu_{o2}), \tag{2}$$

where ν_{oi} is the characteristic frequency of species i in its unperturbed state, α_i is its polarizability, and h is Planck's constant. The potential energy between two spheres of radii r_1 and r_2 can be derived from eq. (1) as (13,14):

$$U_t = - \frac{\Pi^2 q_1 q_2 \lambda}{6} \{ \frac{2 r_1 r_2}{d^2 - (r_1 + r_2)^2} + \frac{2 r_1 r_2}{d^2 - (r_1 - r_2)^2} + \ell n [\frac{d^2 - (r_1 + r_2)^2}{d^2 - (r_1 - r_2)^2}] \}, \tag{3}$$

where q_i is the molecular density of species i, and d is the distance between the centers of the spheres. The attraction force between two spheres is then obtained as:

$$F_t = - \frac{32\Pi^2 q_1 q_2 \lambda d r_1^3 r_2^3}{3[d^2-(r_1+r_2)^2]^2[d^2-(r_1-r_2)^2]^2}. \tag{4}$$

Upon considering the retardation effect on the dispersion force (14), the potential between two molecules is inversely proportional to r^7 instead of r^6, and the proportionality constant λ becomes

$$\lambda = (23/8)hc\alpha_1\alpha_2/\Pi^2, \tag{5}$$

where c is the velocity of light in the air. The potential energy and the attraction force between two spheres become,

$$\begin{aligned} U_t = - \frac{\Pi^2 q_1 q_2 \lambda}{30}\{ & \frac{-1}{(d+r_1+r_2)} + \frac{1}{(d-r_1+r_2)} + \frac{1}{(d+r_1-r_2)} + \frac{1}{(d-r_1-r_2)} \\ & + \frac{r_1 r_2}{d}\left[\frac{1}{(d+r_1+r_2)^2} + \frac{1}{(d-r_1+r_2)^2} + \frac{1}{(d+r_1-r_2)^2} + \frac{1}{(d-r_1-r_2)^2}\right] \\ & - \ell n\left[\frac{d^2-(r_1+r_2)^2}{d^2-(r_1-r_2)^2}\right]/d\}, \end{aligned} \tag{6}$$

$$\begin{aligned} F_t = - \frac{\Pi^2 q_1 q_2 \lambda}{30}\{ & \frac{1}{(d+r_1+r_2)^2} + \frac{1}{(d-r_1+r_2)^2} + \frac{1}{(d+r_1-r_2)^2} + \frac{1}{(d-r_1-r_2)^2} \\ & + \frac{r_1 r_2}{d^2}\left[\frac{1}{(d+r_1+r_2)^2} + \frac{1}{(d-r_1+r_2)^2} + \frac{1}{(d+r_1-r_2)^2} + \frac{1}{(d-r_1-r_2)^2}\right] \\ & + \frac{r_1 r_2}{d}\left[\frac{2}{(d+r_1+r_2)^3} + \frac{2}{(d-r_1+r_2)^3} + \frac{2}{(d+r_1-r_2)^3} + \frac{2}{(d-r_1-r_2)^3}\right] \\ & - \ell n\left[\frac{d^2-(r_1+r_2)^2}{d^2-(r_1-r_2)^2}\right]/d^2 + \frac{8r_1 r_2}{[d^2-(r_1+r_2)^2][d^2-(r_1-r_2)^2]}\}. \end{aligned} \tag{7}$$

If r_1 and r_2 are, respectively, the radius of the drop and that of the particle, then eqs. (6) and (7) represent the intermolecular potential and the attraction force between them. U_t and F_t are significant only

in the proximity of the drop surface, and their magnitudes increase sharply as $d \rightarrow (r_1+r_2)$. Consequently, the intermolecular forces do not affect the mass transfer of the uncharged particles as much as suggested by Zebel (3) for charged particles in electric fields.

3. PARTICLE CAPTURE BY MOLECULAR FORCES

We assume that the drop falls steadily at its terminal velocity, U_o, and that there is a thin diffusion boundary layer of thickness $\delta << r_1$ around the drop surface. We further assume that for the low Reynolds number cases the flow field near the diffusion boundary layer follows the Stokes' solution (7). This necessarily restricts r_1 to smaller sizes($< 50\ \mu$) than previously contemplated by us (1). Neglecting the internal circulation, the fluid velocity can be expressed by

$$U_r/U_o = -[1 - 1.5r_1/r + 0.5(r_1/r)^3]\cos\theta; \tag{8}$$

$$U_\theta/U_o = [2 - 1.5r_1/r - 0.5(r_1/r)^3]\sin\theta, \tag{9}$$

where U_r and U_o are, respectively, the r- and θ- components of the fluid velocity at point (r,θ). Equations (8) and (9) imply that $r_2 << r_1$, i.e., the particle to be captured is much smaller than the drop and the existence of the particle will not greatly influence the flow pattern.

When no external force exists and the particle is small enough to follow the streamlines, one may assume that the particle has the same velocity as that of the fluid. Hence

$$v_r/U_o = -[1 - 1.5r_1/r + 0.5(r_1/r)^3]\cos\theta; \tag{10}$$

$$v_\theta/U_o = [2 - 1.5r_1/r - 0.5(r_1/r)^3]\sin\theta. \tag{11}$$

Further, transformation of the last two equations from r-θ coordinates to x-y coordinates (see Fig. 1), followed by conversion to the dimensionless forms (with r_1 and U_o the units of distance and velocity) will yield upon series expansion and neglect of higher power terms

$$v_x = (3/2)y\sin x; \tag{12}$$

$$v_y = -(3/2)y^2\cos x, \tag{13}$$

which are valid only when $y << 1$.

Let N be the number concentration of particles and v be the velocity of the particle. The particle flux J is Nv if only convective transport is significant. When there exists also a concentration gradient, the particle flux is related to the diffusion process by

$$J = - D\nabla N + Nv, \tag{14}$$

where D is the diffusion coefficient. It is assumed that when the particle reaches the absorbent surface, it is effectively collected by the drop at once. This results in a concentration gradient near the absorbent surface. Considering only the steady state and assuming the particle flow to be incompressible, the continuity equation for the particle flux is

$$\nabla \cdot J = 0. \tag{15}$$

Let N_o be the number concentration far from the absorbent surface (where only the convective transport process is significant) and let $n = N/N_o$, $j = J/(N_o U_o)$. These values of j and n are introduced into eqs. (14) and (15), and the modified form of eq. (14) is substituted into the modified eq. (15). If one ignores the small terms, one obtains the boundary layer equation

$$v_x(\partial n/\partial x) + v_y(\partial n/\partial y) = Pe^{-1}(\partial^2 n/\partial y^2) \tag{16}$$

with the Péclet number $Pe = r_1 U_o/D$. Equation (16) shows that the particle will diffuse toward the absorbent surface if there exists a concentration gradient in a thin layer and the convective transport is large enough to maintain this concentration gradient. This thin layer, with a thickness $\delta << 1$, is called the diffusion boundary layer. In this layer $\partial n/\partial x$ is very small compared with $\partial n/\partial y$. If one neglects the $\partial n/\partial x$ term integrates with respect to y from 0 to y, one obtains

$$(\partial n/\partial y)_y = (\partial n/\partial y)_{y=0}\exp(-Pe\int_0^y v_y dy). \tag{17}$$

If one substitutes eq.(13) into eq. (17), follows this by series expansion of the exponential term, neglects the higher power terms, and imposes the boundary condition $(\partial n/\partial y)_\delta = 0$, he obtains

$$1 - \tfrac{1}{2}Pe\delta^3\cos x + (1/8)Pe^2\delta^6\cos^2 x - (1/48)Pe^3\delta^9\cos^3 x = 0. \tag{18}$$

Equation (18) expresses the relationship between the boundary layer thickness δ and the values of x. Considering $(\partial n/\partial y)_{y=0}$ as a constant for a given value of x, integration of eq. (17) after series expansion and neglect of the higher power terms, yields

$$n = (\partial n/\partial y)_{y=0}[y - (1/8)Pey^4\cos x + (1/56)Pe^2y^7\cos^2 x - (1/480)Pe^3y^{10}\cos^3 x]. \tag{19}$$

Equation (19) gives the concentration distribution inside the diffusion boundary layer. The term $(\partial n/\partial y)_{y=0}$ may be obtained by using eq. (19) and the boundary condition n = 1 at $y = \delta$. One has

$$(\partial n/\partial y)_{y=0} = [\delta - (1/8)Pe\delta^4\cos x + (1/56)Pe^2\delta^7\cos^2 x - (1/480)Pe^3\delta^{10}\cos^3 x]^{-1}. \tag{20}$$

The particle density at the surface of the drop is inferred from

$$J|_{y=0} = N_o D(\partial n/\partial y)_{y=0}/r_1. \tag{21}$$

If one assumes that all collisions with the drop surface lead to capture, then the collection efficiency E can be expressed as

$$E = (2/Pe)\int_0^{x_u} (\partial n/\partial y)_{y=0}\sin x dx, \tag{22}$$

where x_u is the upper limit at which the diffusion boundary layer exists.

As mentioned before, the magnitude of intermolecular forces diminishes sharply as y increases in the vicinity of the drop. Hence the intermolecular forces cannot be expected to create a strong force field and to build up a diffusion boundary layer. They can only provide a certain region as an absorbent surface for the collection, influence the nearby concentration distribution, and act as a compensation for the effects that increase the hydrodynamic resistance in the proximity of the absorbent surface. When the particle reaches the absorbent surface, it will sooner or later be captured by the drop. If the distance between the absorbent surface and the drop surface is I and one assumes that the particle will be driven rapidly to the drop surface as soon as it reaches this region, equation (20) becomes

$$(\partial n/\partial y)_I = [\delta - I - (1/8)Pe(\delta^4 - I^4)\cos x + (1/56)Pe^2(\delta^7 - I^7)\cos^2 x - (1/480)Pe^3(\delta^{10} - I^{10})\cos^3 x]^{-1}. \quad (23)$$

Since the thickness δ should not be greatly influenced by the intermolecular forces, the relationship between δ and x as shown in eq. (18) remains unchanged.

Upon examination of eq. (7), one finds that, in the region $(r_1+r_2) \leq d \leq d_o$, with d_o being the position where

$$(\partial F_t/\partial d) = 1, \quad (24)$$

the magnitude of F_t increases sharply as $d \to (r_1+r_2)$. If the particle reaches this region, it may be warranted to be collected. Letting d_o be the radius of the absorbent surface and $I = d_o - (r_1+r_2)$, the collection efficiency E becomes

$$E = \frac{2d_o^2}{Pe(r_1+r_2)^2}\int_0^{x_u} (\partial n/\partial y)_{y=I} \sin x dx. \tag{25}$$

Use of eq. (25) with the constant value $(\partial n/\partial y)_{y=I}$ at the forward stagnation point gives the simple formulas for the collection efficiency:

1) For $x_u = \Pi/2$,

$$E = \frac{2d_o^2}{Pe(r_1+r_2)^2}(\partial n/\partial y)\Big|_{y=I,x=0}. \tag{26}$$

2) For $x_u = \Pi$,

$$E = \frac{4d_o^2}{Pe(r_1+r_2)^2}(\partial n/\partial y)\Big|_{y=I,x=0}. \tag{27}$$

For a large drop, δ is very small compared with r_1, and the collection on the backside of the drop due to wake effect may be as important as that on the front side, consequently x_u should be extended to Π.

4. RESULTS AND DISCUSSION

The particle diffusion coefficient D can be estimated from the Stokes-Einstein relation (15):

$$D = kTB, \tag{28}$$

where k is the Boltzman constant and T is the absolute temperature. The mobility B can be calculated for a spherical particle of radius r_2 in a fluid of viscosity μ from the equation (15):

$$B = \{1 + (\ell/r_2)[1.257 + 0.4\exp(r_2/\ell)]\}/(6\pi\mu r_2). \tag{29}$$

The mean free path ℓ is given to a close approximation by:

$$\ell = (\mu/\rho)\ (\frac{\pi M}{2RT})^{\frac{1}{2}}, \tag{30}$$

where ρ is the density of carrying gas, M is its molecular weight and R is the gas constant. For air at 20°C. and atmospheric pressure, ℓ is approximately 6.53×10^{-5} mm.

The terminal velocity U_o of the drop may be computed from the empirical formula proposed by Best (16):

$$U = 943\{1 - \exp[-(2r_1/1.77)^{1.147}]\}. \tag{31}$$

Using eqs. (28-31) and the physical constants given in Table I, we have computed the collection efficiency, E, for the case of a water drop collecting AgCl aerosols and that of a water drop collecting submicron cloud droplets. Some representative results are shown in Table II and Table III. In general, E decreases as the particle size increases for a given drop size, and decreases as the drop size increases for a given particle size. In order to explain the general trends observed, we examine in more detail two principal mechanisms that are responsible for the effective capture of submicron particles, namely, the diffusion

process and the intermolecular attraction.

On the one hand, the diffusion process is influenced mainly by the convective transport and the mobility of the particle. The relative importance of these two factors is represented by the Péclet number,which is the ratio of convective transport, NU_o, to diffusive transport, ND/d_o. Although a large value of Pe may warrant a comparatively large amount of particle flux to the drop upstream, it also provides a much larger sweeping force that sweeps the particle around the drop. Consequently, a larger Pe gives rise to a smaller collection efficiency.

On the other hand, the intermolecular attraction in our case extends over a region of several microns in the vicinity of the drop surface. It is assumed that the intermolecular forces provide a certain region as an absorbent surface in the proximity of the drop. Some of the representative values of the radius of the absorbent surface d_o are given in Table IV. Both d_o and the magnitude of the intermolecular forces depend mainly on the sizes of the interacting objects and the interaction constant λ. A larger value of d_o yields a larger value of E for a given size of the collecting drop.

From the data obtained, the collection efficiency E depends mainly on the diffusion process if the coalescence efficiency is 1. Since the intermolecular forces extend over a region of only a few microns, they cannot be expected to influence greatly the mass transfer process. Thus the determining step in an effective collection is for the particle to fall by diffusion within the range of the intermolecular forces.

The calculation has been extended to the case of large drops. For the large drop, the validity of the Stokes' solution is in doubt and both the internal circulation and wake effect become important. Therefore,the

data of the large drop showed in Table II and Table III are only for the purpose of comparison.

In our consideration of the flow field the internal circulation of the drop has been neglected. However, since the internal circulation may reduce to some extent the shear flow, it may also reduce the viscous interaction. Hence, the influence of the intermolecular forces on the particle could be larger. Consequently, by neglecting the internal circulation we may underestimate the collection efficiency of the large collecting drops.

We have considered the collection on the backside of the drop by extending x_u to π. However, the flow field on the backside of the drop is quite different from that of the front side, since on the backside there are eddies. Due to their inertia the particles may lag behind the fluid when they follow the streamlines and at the same time they may experience a rotational force arising from the high velocity gradient in the eddies. Consequently, the particle flux in the diffusion boundary layer could be larger and the collection efficiency may increase.

Compared with the experimental data given by Kerker (17) for AgCl aerosols, our results are in good agreement (the differences are within 10%) with the values obtained from

$$E = 1.68\ Pe^{-2/3}, \tag{32}$$

which is proposed from his experimental results in the case of the drop radius smaller than 1.0 mm. For the large drop, however, our results are much smaller.

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TABLE I. PHYSICAL CONSTANTS OF Ag^+Cl^- AND H_2O

	Ag^+ *	Cl^- *	H_2O **
$\alpha(cm^3)$	1.9×10^{-24}	2.98×10^{-24}	1.68×10^{-24}
$q(cm^{-3})$	2.337×10^{22}	2.337×10^{22}	3.346×10^{22}
Ionization Potential (Kcal/mole)	494	86.5	415

*From Phillips, C. S. G., and William, R. J. P., "Inorganic Chemistry," Oxford University Press, New York, 1965.

**see reference 12.

TABLE II. THE COLLECTION EFFICIENCY OF AgCl AEROSOLS BY A WATER DROP FOR THE CASE OF $x_u = \pi/2$. ($E \times 10^5$)

r_1(mm)	$r_2(\mu)$ 0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0
0.01	1645.0	873.8	629.1	505.4	429.3	377.2	339.0	309.5	286.1	266.8
0.02	608.3	322.1	231.4	185.6	157.4	138.1	123.9	113.0	104.3	97.2
0.03	340.5	180.1	129.2	103.5	87.7	76.9	69.0	62.9	58.0	54.0
0.05	164.4	86.8	62.2	49.8	42.2	36.9	33.1	30.2	27.8	25.9
0.07	102.0	53.8	38.6	30.8	26.1	22.9	20.5	18.6	17.2	16.0
0.10	61.7	32.5	23.3	18.6	15.7	13.8	12.3	11.2	10.3	9.6
0.20	23.1	12.2	8.7	7.0	5.9	5.1	4.6	4.2	3.8	3.6
0.50	7.0	3.6	2.6	2.1	1.8	1.5	1.4	1.3	1.2	1.1
1.00	3.1	1.6	1.2	0.9	0.8	0.7	0.6	0.6	0.5	0.5
1.50	2.1	1.1	0.8	0.6	0.5	0.5	0.4	0.4	0.3	0.3

TABLE III. THE COLLECTION EFFICIENCY OF SUBMICRON CLOUD DROPLETS BY A WATER DROP FOR THE CASE OF $x_u = \pi/2.$ ($E \times 10^5$)

r_1 (mm)	r_2 (μ) 0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0
0.01	1638.0	868.3	624.0	500.4	424.5	372.5	334.2	304.9	281.5	262.3
0.02	606.4	320.6	230.0	184.1	156.0	136.7	122.6	111.7	103.0	95.9
0.03	339.6	176.3	128.5	102.8	87.1	76.3	68.3	62.2	57.4	53.4
0.05	164.0	86.5	61.9	49.6	41.9	36.7	32.8	29.9	27.5	25.6
0.07	101.8	53.6	38.4	30.7	25.9	22.7	20.3	18.5	17.0	15.8
0.10	61.6	32.4	23.1	18.5	15.7	13.7	12.3	11.1	10.3	9.5
0.20	23.1	12.2	8.7	6.9	5.9	5.1	4.6	4.1	3.8	3.6
0.50	7.0	3.7	2.6	2.1	1.8	1.5	1.4	1.2	1.2	1.1
1.00	3.1	1.6	1.2	0.9	0.8	0.7	0.6	0.6	0.5	0.5
1.50	2.1	1.1	0.8	0.6	0.5	0.5	0.4	0.4	0.3	0.3

TABLE IV. THE DISTANCE BETWEEN THE ABSORBENT SURFACE AND THE DROP SURFACE. ($I \times 10^4$ mm)

$r_2(\mu)$	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0
AgCl Aerosols	0.162	0.194	0.215	0.231	0.245	0.257	0.267	0.276	0.284	0.291
Submicron Cloud Droplets	0.136	0.163	0.180	0.194	0.205	0.214	0.223	0.231	0.238	0.244

The radius of absorbent surface can be expressed by:

$$d_o = (r_1 + r_2) + I.$$

— — — CONCENTRATION BOUNDARY LAYER PROFILE

- - - - - - STREAMLINE

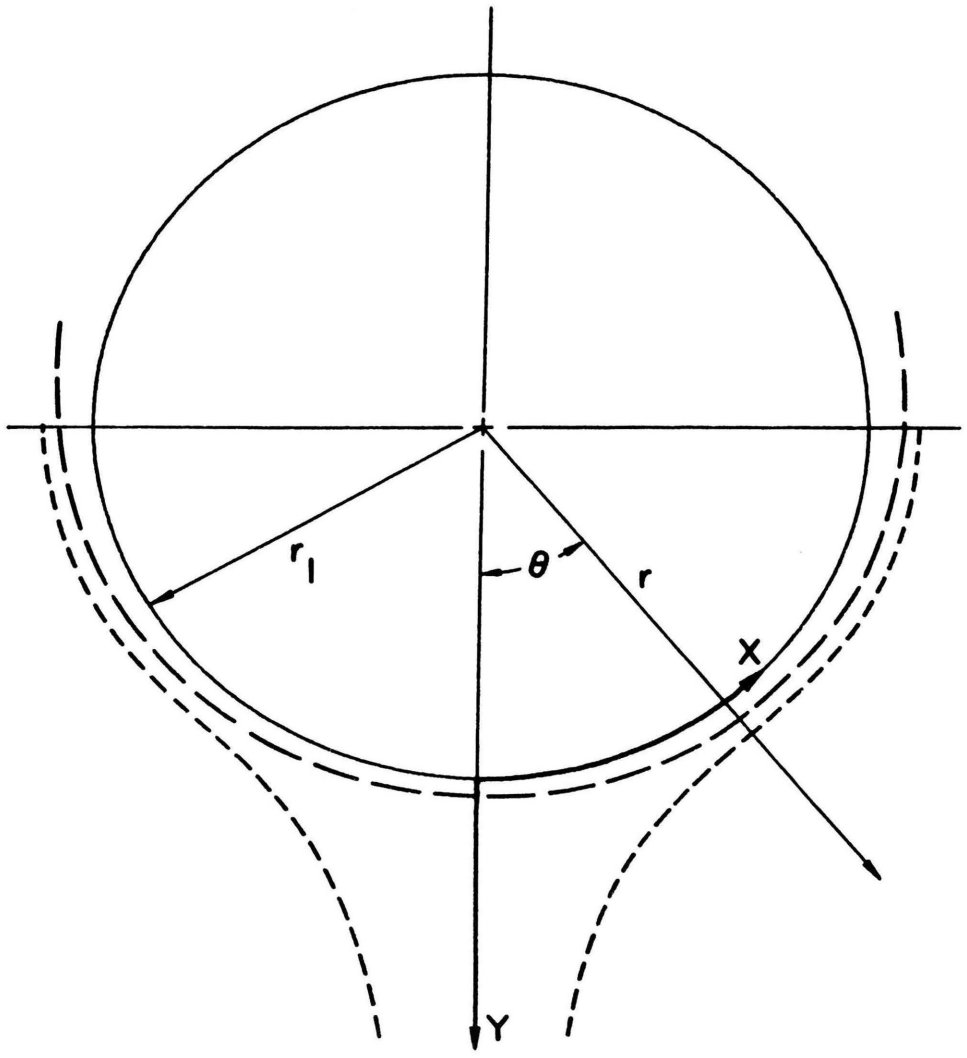

Fig. 1

VITA

Ming-shian Wu, son of Mr. and Mrs. Ben-nan Wu, was born in Hsinchu, Taiwan, on Feb. 19, 1940. He received his primary and secondary education in Hsinchu. After graduating from Hsinchu High School in 1959, he was employed by the National United Industrial Research Center, Hsinchu, for a year. He attended the National Taiwan University, Taipei, Taiwan, from 1960 to 1964, and graduated with a degree of Bachelor of Science in Chemical Engineering. He was drafted into the military service, subsequently, in Chinese Air Force for a year and served as a Second Lieutenant. He taught Chemistry in Hsinchu High School in 1965-1966, and then worked for the National Taiwan University as a fulltime teaching and research assistant in Analytical Chemistry, Department of Chemistry.

He came to the United States of America and enrolled at the University of Missouri-Rolla, for graduate work toward a Ph. D. degree in Physical Chemistry, in 1967.

On June 21, 1969, he married the former Miss Justina Chieh-Hui Hsieh of Taipei, Taiwan, a Ph. D. candiate of the University of Missouri-Rolla, in Department of Chemistry.