

## Studies on the motion of surface films in two-phase flows

J. SIEKMANN (ESSEN) and W. JOHANN (WERMELSKIRCHEN)

A FLUID interface (for example, liquid-gas) is generally not clean, that is the interface is a region of variable composition owing to its thermodynamic attraction for many of the contaminants present in the adjoining bulk fluid. So much surface-active material (surfactant) may be adsorbed at the interface that the surfactant forms a layer (surface film) with a measurable surface elasticity or surface viscosity of its own. These adsorbed impurities lower the surface tension. The present paper deals with the investigation of the motion of a surface film in a two-phase flow. The theoretical discussion is based on the hypothesis that the film flow is independent of the liquid motion. Simple experimental arrangements (flow in a rotating spherical cavity and between rotating discs) allow a comparison between the theoretical model and the physical reality and show that agreement between theoretical results and experimental data is satisfactory.

Powierzchnia międzyfazowa w płynach (np. ciecz-gaz) nie jest zazwyczaj czysta, to znaczy, że powierzchnia ta jest obszarem o zmiennym składzie z uwagi na jej termodynamiczne oddziaływanie przyciągające wiele zanieczyszczeń znajdujących się w otaczającym płynie. Na powierzchni tej zaobserwować można tyle powierzchniowo czynnego materiału, że zanieczyszczenia te utworzą warstwę (błonkę powierzchniową) o mierzalnej sprężystości lub lepkości. Zaobserwowane zanieczyszczenia zmniejszają napięcie powierzchniowe. W pracy niniejszej rozważa się ruch takiej błonki powierzchniowej w przepływie dwufazowym. Rozważania teoretyczne opierają się na hipotezie, że przepływ błonki jest niezależny od ruchu samego płynu. Proste doświadczenia (przepływ w obracającej się pustce kulistej lub między obracającymi się tarczami) pozwalają porównać model teoretyczny z rzeczywistością fizyczną i pokazują, że zgodność między wynikami teoretycznymi i doświadczalnymi jest zadowalająca.

Межфазовая поверхность в жидкостях (например жидкость-газ) не является обычно чистой, т.е., что эта поверхность является областью с переменным составом из-за ее термодинамического воздействия, притягивающего много загрязнений, находящихся в окружающей жидкости. На этой поверхности может абсорбироваться так много поверхности активного материала, что эти загрязнения образуют слой (поверхностная пленка) с измеримой упругостью или вязкостью. Абсорбированные загрязнения уменьшают поверхностные напряжения. В настоящей работе рассматривается движение такой поверхностной пленки в двухфазном течении. Теоретические рассуждения опираются на гипотезу, что течение пленки не зависит от движения самой жидкости. Простые эксперименты (течение в вращающейся сферической пустоте или между вращающимися дисками) позволяют сравнить теоретическую модель с физической действительностью и показывают, что совпадение между теоретическими и экспериментальными результатами удовлетворительно.

### 1. Introduction

IN GENERAL, fluid interfaces (for example, liquid-gas) are not clean, that is the interface is a region of variable composition owing to its thermodynamic attraction for many of the contaminants present in the adjoining bulk fluid. So much surface-active material may be adsorbed at the (liquid-gas) interface that the surfactant forms a layer (surface film) with a measurable surface elasticity or surface viscosity of its own. These adsorbed impurities lower the surface tension [1, 2, 3, 4]. The present study deals with the theoretical

and experimental investigation of the motion of a surface film in a two-phase flow (water-air). The basic features of the observed phenomena can be described as follows: Liquid-wetted walls are withdrawn from a liquid the surface of which is coated with an incompressible and insoluble surface film of constant monomolecular thickness. The thickness of this film layer is of the order of about 50 Å. A negligible amount of liquid sticks to the wall and carries on its part the surface film. This thin liquid layer, together with the film, is taken along by the wall. Practically one can ignore the existence of the thin liquid layer and assume that the surface film adheres directly to the solid. According to this model, the film is lifted from the liquid if the wall is taken out of the liquid. Conversely, if the film-coated wall is dipped into the liquid, the film is delivered again to the liquid surface. This flow model has been applied to a fluid enclosed in a rotating spherical cavity and between closely-spaced plane discs which are made to rotate in their own planes with steady angular velocity. The fluid (water) occupies about half of the cavity. Furthermore, stearic acid has been added to the fluid. A simple theoretical model, where the influence of the liquid flow on the (slow) film motion has not been taken into account and where a plane, horizontal surface film has been assumed, leads to the biharmonic equation for the stream function of the film flow. This differential equation allows a solution in closed form. Agreement between theoretical and experimental data and observations is satisfactory.

## 2. Film motion in closed rotating tanks

In what follows we discuss the motion of a surface film caused by the rotation of a closed tank, half of which is filled by a liquid (water), the rest by a gas (air). Due to contamination and the presence of surface active agents (stearic acid), a surface film appears at the gas-liquid interface. Two tank geometries are considered: the spherical one and the cylindrical one, the latter having a circular cross section and the bases very close together. The axes of symmetry are chosen as the axes of rotation. They are perpendicular to the direction of gravity. In both cases the steady rotation of the tank (angular speed  $\Omega$ ) causes a movement of the film on the gas-liquid interface and on the tank wall in contact with the gas.

In order to determine the motion of the film, the following idealized assumptions are made:

- (i) the influence of the liquid flow on the film motion is negligible,
- (ii) the surface of the film covering the liquid is plane and positioned horizontally,
- (iii) the surface film is insoluble, incompressible, and of constant thickness and surface viscosity,
- (iv) the steady tank rotation causes the movement of the surface film on the gas-liquid interface and on the tank wall in contact with the gas,
- (v) the motion of the surface film is slow (creeping flow).

In connection with first assumption it should be pointed out that this postulate is indeed a very strong restriction. Actually the motion of the film and the bulk fluid should be treated simultaneously. This leads to a rather complex flow problem since both flow

fields are coupled. The underlying liquid exerts shear stresses on the film. This effect, however, has not been taken into account in the present analysis. Neglecting this effect results, of course, in a noticeable difference between theoretical and experimental data. Nevertheless, the elementary theory describes the observed phenomena qualitatively correct; quantitative agreement between theoretical predictions and test results is satisfactory.

Assuming the film to be a Newtonian liquid, and because of the assumptions (ii), (iii) and (iv), we note that the film motion is governed by the Stokes equations of plane flow in circular or rectangular cross sections, respectively. Since the gas-liquid interface is exactly at the equatorial or axial cross section, respectively, the film does not deform or break at the wall. Introducing a film stream function  $\psi(r, \vartheta)$  or  $\psi(x, y)$ , respectively, the solution of the problem under consideration is reduced to the solution of the biharmonic equation

$$(2.1) \quad \nabla^4 \psi \equiv \nabla^2 \nabla^2 \psi = 0,$$

where  $\nabla^2$  denotes the Laplacian in polar or Cartesian coordinates, respectively. Solutions of the basic equation (2.1), subject to appropriate boundary conditions, are discussed in the following section.

### 2.1. Film flow in a rotating spherical cavity

Let us consider the film flow in a rotating spherical cavity of radius  $R$  as shown schematically in Fig. 1. Thus, if  $(r, \vartheta, z)$  are cylindrical polar coordinates with origin at the center  $O$  of the cavity, we have for the radial velocity component  $u_0$  and the azimuthal velocity component  $v_0$  of the film flow in the equatorial plane of the sphere

$$(2.2) \quad u_0 = -\frac{1}{r} \psi_{\vartheta}, \quad v_0 = \psi_r,$$

where a subscript denotes partial differentiation. The boundary conditions to be imposed on Eq. (2.1) are

$$(2.3) \quad u_0(R, \vartheta) = R\Omega \sin \vartheta, \quad v_0(R, \vartheta) = 0.$$

Next we introduce dimensionless quantities by making lengths and velocities dimensionless by  $R$  and  $R\Omega$ , respectively. For the sake of simplicity no special notation will be employed. The solution of the biharmonic equation (2.1) can be written as [5]

$$(2.4) \quad \psi = \psi_1 + r^2 \psi_2,$$

where  $\psi_1$  and  $\psi_2$  are harmonic functions. Since the determination of the solution is straightforward, details will be omitted. A few standard manipulations yield

$$(2.5) \quad \psi = \frac{3}{2} \left( r - \frac{r^3}{3} \right) \cos \vartheta$$

for the stream function, and

$$(2.6) \quad u_0 = \frac{3}{2} \left( 1 - \frac{r^2}{3} \right) \sin \vartheta, \quad v_0 = \frac{3}{2} (1 - r^2) \cos \vartheta$$

for the velocity components.

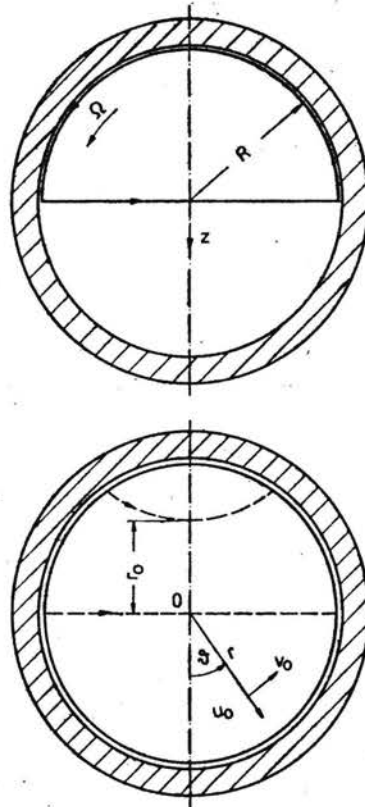


FIG. 1. Film flow in a rotating spherical cavity, geometry and notation.

Streamlines having from the origin the distance  $r_0$  (for  $\vartheta = 0$  and  $\vartheta = \pi$ , respectively) are given by

$$(2.7) \quad \vartheta(r) = \arccos \frac{3r_0 - r^3}{3r - r^3}.$$

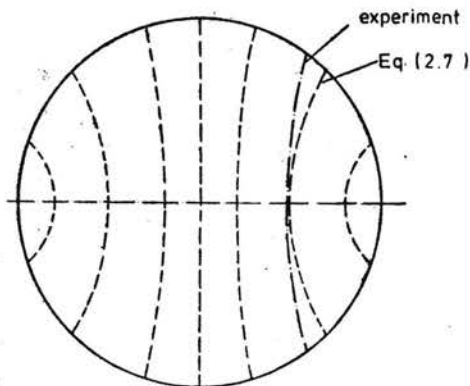


FIG. 2. Streamlines of film flow in a half-filled rotating spherical cavity.

Computations based on Eq. (2.7) are presented in Fig. 2. One observes that the experimentally-determined streamline exhibits a weaker curvature than the theoretically-calculated curve. This deviation which is more pronounced for points close to the equator is to some extent due to the simplifying assumptions (in particular (i)). A rigorous analysis should begin with the simultaneous solution of the (coupled) equations of motion for film and fluid flow.

For the mean velocity  $u_{0m}^*$  of film particles on the streamline passing through the origin of the coordinate system, we obtain

$$(2.8) \quad u_{0m}^* = \int_0^1 u_0 \left( r, \frac{\pi}{2} \right) dr = \frac{4}{3},$$

or, in dimensional form,

$$(2.9) \quad \bar{u}_{0m} = \frac{4}{3} R\Omega \approx 1.33 R\Omega.$$

Time measurements for particles moving on the mean film streamline yield for the mean film velocity approximately  $\bar{u}_{0m} \approx 1.15 R\Omega$ . Thus the experimental value is somewhat smaller than the theoretically predicted result. However, it should be pointed out that in view of the assumptions made agreement is very satisfactory. Figure 3 shows the test

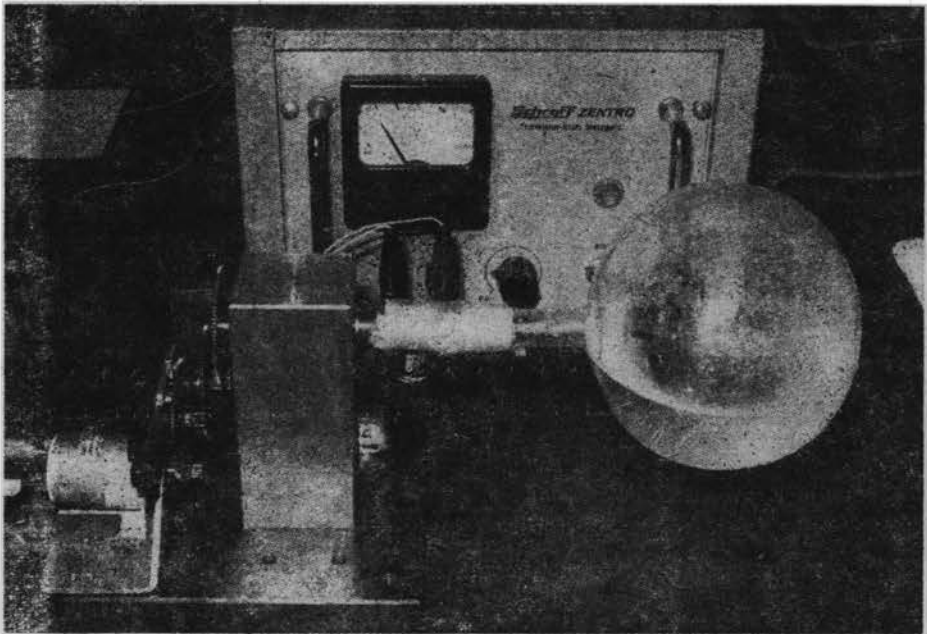


FIG. 3. Test apparatus.

apparatus designed by the junior author [6]. The connecting-piece of a glass-piston (radius 60 mm) is mounted on the driving shaft (diameter 12 mm) of a small gearing as seen schematically in Fig. 4. Film streamlines (Fig. 2) were determined experimentally by means of aluminium particles on the upper surface of the contaminant.

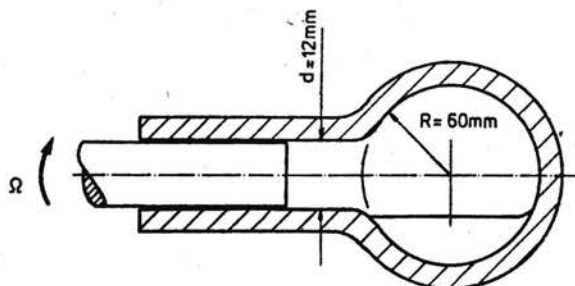


FIG. 4. Sketch of apparatus.

## 2.2. Flow phenomena between closely-spaced rotating discs

The test set-up described in detail by JOHANN [6] is shown in Fig. 5. The surface film, enclosing the upper half-space between the rotating discs, is delivered to the liquid in the long and narrow canal between the discs by the walls in the region of the advancing

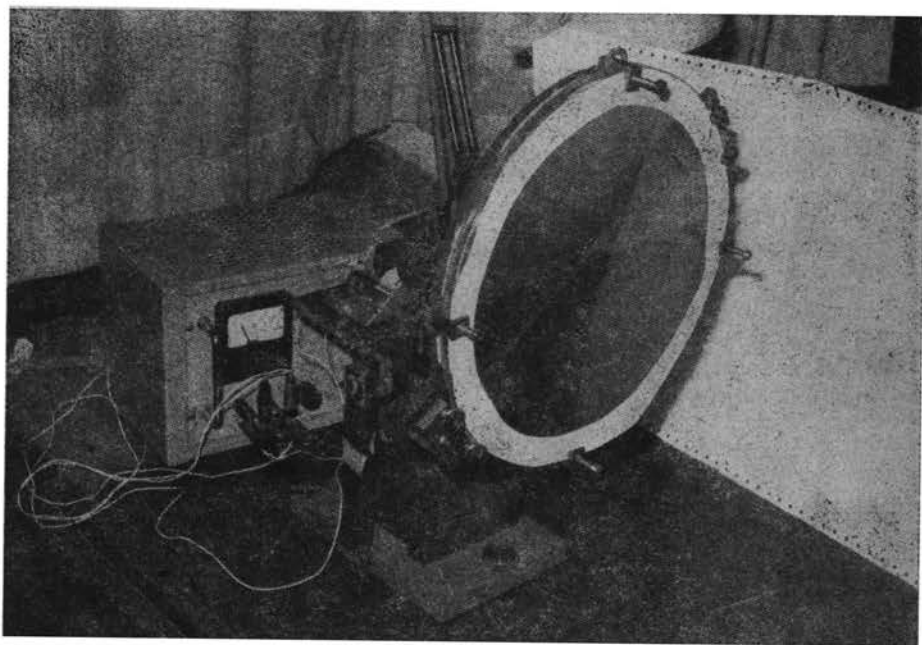


FIG. 5. Test apparatus.

meniscus and is taken up again by the walls in the region of the receding meniscus. Since the ratio  $R/h$  ( $R$ —radius of the discs,  $h$ —half-width of the gap between the discs) is rather large, there occur, in virtue of the continuity of the flow of the surface-active agent (film), considerable film speeds in the central range of the canal.

The theoretical treatment of the film movement within the canal is based on the flow pattern displayed in Fig. 6. In addition to the assumptions (i)–(v), we suppose that

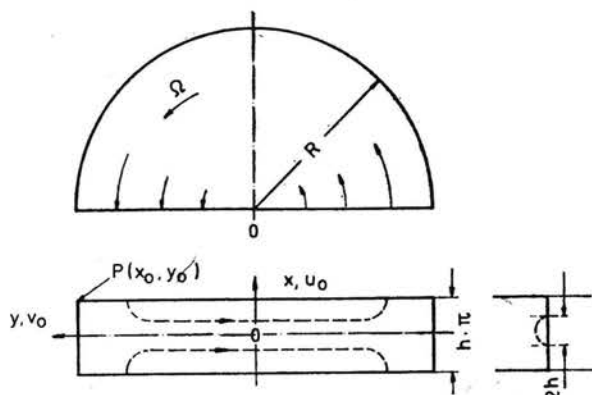


FIG. 6. Model of idealized film flow, geometry and notation — — — streamlines of film flow (not true to scale).

(a) the cross section of the interface has the shape of a semicircle over the entire length of the canal; by means of stretching there results a plane film surface of breadth  $2x_0 = h\pi$ . A Cartesian coordinate system  $(0; x, y)$  is located in this plane;

(b) the surface film sticks to the walls of the discs  $(\pm x_0, y)$ . At the boundaries  $(x, \pm y_0)$ , the film flow has to fulfill the inflow, or outflow condition, respectively,

$$(2.10) \quad \left| \int_{-x_0}^{x_0} v_0(x, \pm y_0) dx \right| = n \cdot 2x_0 y_0 \Omega,$$

where  $n = 1$  corresponds to a closed hollow cylinder without reverse flow in the corners, while  $n = 0$  corresponds to the case of an open canal between rotating discs dipping into a liquid which is covered by a surface-active agent.

First of all it can be shown that the integral condition (b) is satisfied by a parabolic velocity distribution:

$$(2.11) \quad |v_0(x, \pm y_0)| = \frac{3}{2} n y_0 \Omega \left( 1 - \frac{x^2}{x_0^2} \right).$$

Next, we have to solve the biharmonic equation (2.1) for the film stream function under appropriate boundary conditions. The velocity components are given by

$$(2.12) \quad u_0 = -\psi_y, \quad v_0 = \psi_x.$$

Making all lengths dimensionless by  $y_0 = R$ , all velocities by  $y_0 \Omega$ , we obtain for the boundary conditions, taking into account (b) and Eq. (2.11), the following expressions (again, no special notation for dimensionless quantities will be employed)

$$(2.13) \quad \begin{aligned} u_0(x_0, y) &= u_0(-x_0, -y) = y, \\ u_0(-x_0, y) &= u_0(x_0, -y_0) = -y, \\ v_0(x_0, y) &= v_0(-x_0, y) = 0, \\ |v_0(x, \pm 1)| &= \frac{3}{2} n \left( 1 - \frac{x^2}{x_0^2} \right). \end{aligned}$$

The solution of the governing equation (2.1) is posed in the form [7]

$$(2.14) \quad \psi = Axy^2 + Bx + ax \cos \lambda x \cosh \lambda y + bx \cos \lambda y \cosh \lambda x,$$

where  $A$ ,  $B$ ,  $a$  and  $b$  are integration constants (to be determined from the boundary conditions), and  $\lambda$  is a parameter. The solution of this boundary value problem requires elementary but laborious operations only. In order to obtain simple analytical expressions, we develop the trigonometric and hyperbolic functions in series form and neglect terms of higher than the second order. Dropping terms of higher order is suggested by the fact that the free parameter  $\lambda$  can be chosen as a small quantity. With

$$(2.15) \quad \nu = \lambda x_0$$

we obtain for the unknowns  $A$ ,  $B$ ,  $a$  and  $b$

$$(2.16) \quad A = -\frac{\lambda}{4\nu^3} (\lambda^3 + 3\nu^2 + 2n\lambda\nu), \quad a = \frac{1 - \frac{\lambda^2}{2} - n\lambda\nu}{2\lambda\nu^3},$$

$$(2.17) \quad B = \frac{1 + \frac{3}{4}\lambda^2\nu^2 + \frac{3}{2}n\lambda\nu^3}{\lambda\nu^3}, \quad b = -\frac{1 + \frac{\lambda^2}{2} + n\lambda\nu}{2\lambda\nu^3}.$$

The streamlines are calculated from the simplified stream function

$$(2.18) \quad \psi = Axy^2 + Bx + ax \left(1 - \frac{\lambda^2}{2} x^2\right) \left(1 + \frac{\lambda^2}{2} y^2\right) + bx \left(1 + \frac{\lambda^2}{2} x^2\right) \left(1 - \frac{\lambda^2}{2} y^2\right),$$

where the cosine and hyperbolic cosine functions of Eq. (2.14) are replaced by their corresponding series expansions (up to the quadratic terms). Denoting points on the boundary of the rectangular film domain by  $\tilde{x}$  and  $\tilde{y}$ , introducing the constants  $A$ ,  $B$ ,  $a$ ,  $b$  as given by Eq. (2.17), recalling Eq. (2.16), and setting

$$(2.19) \quad X = \frac{\tilde{x}}{x_0}, \quad Y = \tilde{y}, \quad m = \frac{x_0}{y_0}, \quad \xi = \frac{x}{x_0},$$

it is only a matter of algebra to work out the equation of the streamlines

$$(2.20) \quad y = \frac{1}{\sqrt{X^3 - 3X}} [(3 + 6nm)(X - \xi) - (1 + 2nm)(X^3 - \xi^3) + Y^2(X^3 - 3\xi)]^{1/2}.$$

Plots of streamlines, according to Eq. (2.20), are shown in Fig. 7. We note in passing that these streamlines are qualitatively in good agreement with observed trajectories of particles. To compare theoretical and experimental data we calculate the mean film velocity  $\bar{v}_{0m}$  in the range  $-r_M \leq y \leq r_M$ , i.e. within a test section of length  $2r_M$ .

Defining  $\bar{v}_{0m}$  by

$$(2.21) \quad \bar{v}_{0m} = \frac{1}{2r_M} \int_{-r_M}^{r_M} v_0(0, y) dy$$

and making use of Eqs. (2.12), (2.14) and (2.17), we obtain

$$(2.22) \quad \bar{v}_{0m} = y_0 \Omega \left[ \frac{1}{4} \frac{x_0}{y_0} \left( 3 - \frac{r_M^2}{y_0^2} \right) + \frac{3}{2} n \right].$$



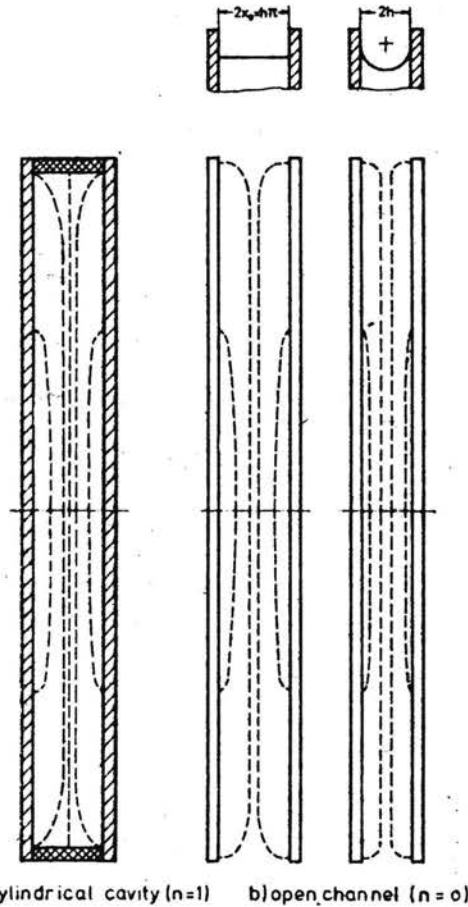


FIG. 7. Streamlines of film flow in the long and narrow canal between rotating discs.

It is obvious to neglect the inflow or outflow, respectively, of the film at the narrow wall of the hollow cylinder ( $2h \ll R$ ). Furthermore, we introduce the actual dimensions of the canal. This leads to the following approximate formula for the mean film velocity:

$$(2.23) \quad \bar{v}_{0m} \approx R\Omega \left[ \frac{R}{2h\pi} \left( 3 - \frac{r_M^2}{R^2} \right) \right].$$

For frosted glass discs the expression for  $\bar{v}_{0m}$  should be multiplied by a roughness factor  $\kappa > 1$ .

Figure 8 shows a plot of the mean film velocity ( $\bar{v}_{0m}$ ) as a function of the angular speed ( $\Omega$ ). The experimentally-determined curve  $\bar{v}_{0m}(\Omega)_{LM}$  denotes the mean film velocity over the entire test range  $L_M = 2r_M$ , which extends symmetrically with respect to the center of the disc. Furthermore,  $\bar{v}_{0A} = \bar{v}_{0A}(\Omega)_{LMA}$  is the mean velocity over half of the test range ( $L_{MA}$ ) in the region of the advancing meniscus, while  $\bar{v}_{0R} = \bar{v}_{0R}(\Omega)_{LMR}$  is the corresponding velocity in the region of the receding meniscus. Since tiny particles scattered on the upper part of the film surface travel with the film flow, the mean film velocity

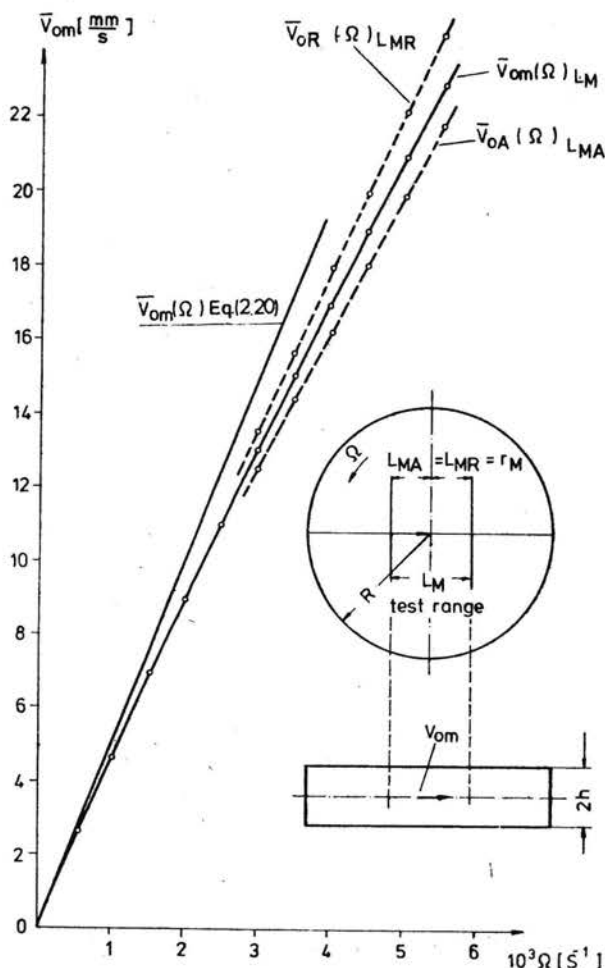


FIG. 8. Plot of  $\bar{v}_{om}$  vs.  $\Omega$ ;  $R = 140$  mm,  $2h = 3.6$  mm,  $L_M = 100$  mm, disc material: frosted glass.

$\bar{v}_{om}(\Omega)_{LM} \equiv \bar{v}_{om,exp} = L_M/T_M$  is found easily by timing ( $T_M$ ) of the distance ( $L_M$ ) covered by the aluminum chips placed on the surfactant. The discrepancy between experimental results and the theoretically (Eq. 2.23) predicted linear behavior of  $\bar{v}_{om}(\Omega)$  is very probably due to the compressibility of the surface film and a reverse flow in the corners of the hollow cylinder. After stopping of the discs one observes a flow of the surface from the advancing to the receding meniscus until the film pressure gradient vanishes. Finally, the complicated flow phenomena in the region of the advancing and receding meniscus give rise to a visible deviation of the real interface from the equilibrium interface. The real interface is inclined towards the horizontal (Fig. 9). Test results of the interface displacement  $\Delta H$  vs.  $\Omega$ , and vs. the normal film velocity  $U_s$  are displayed in Figure 10. Experiments were carried out by tilting the (vertical) axis of the discs towards the horizontal ( $\alpha$ —angle of inclination). We note that  $\Delta H$  is approximately proportional to the effective gravitational acceleration  $g^* = g \sin \alpha$ . The

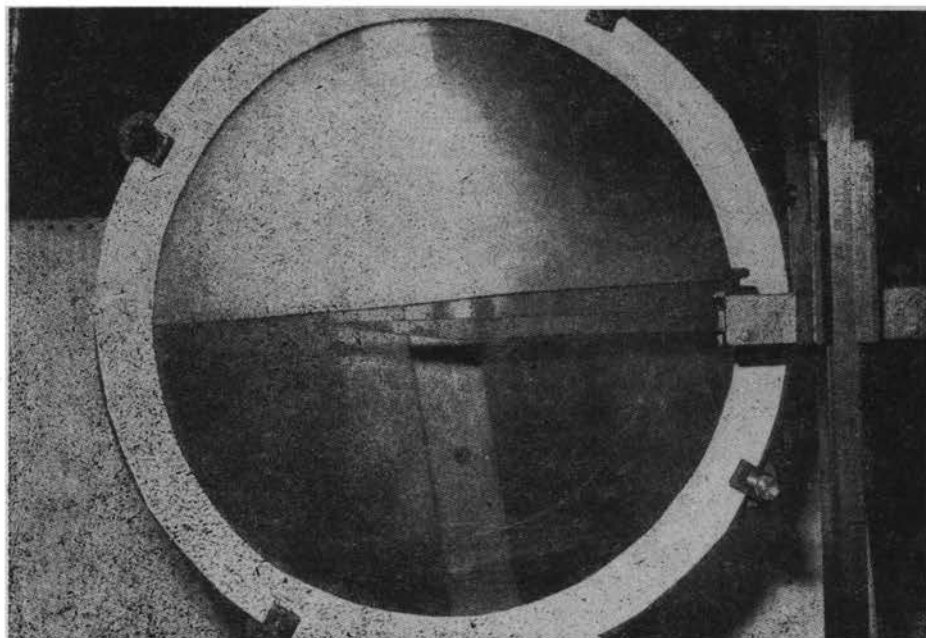


FIG. 9. View of surface film level,  $g^* = g$ ,  $\Omega = 0.5 \text{ s}^{-1}$ .

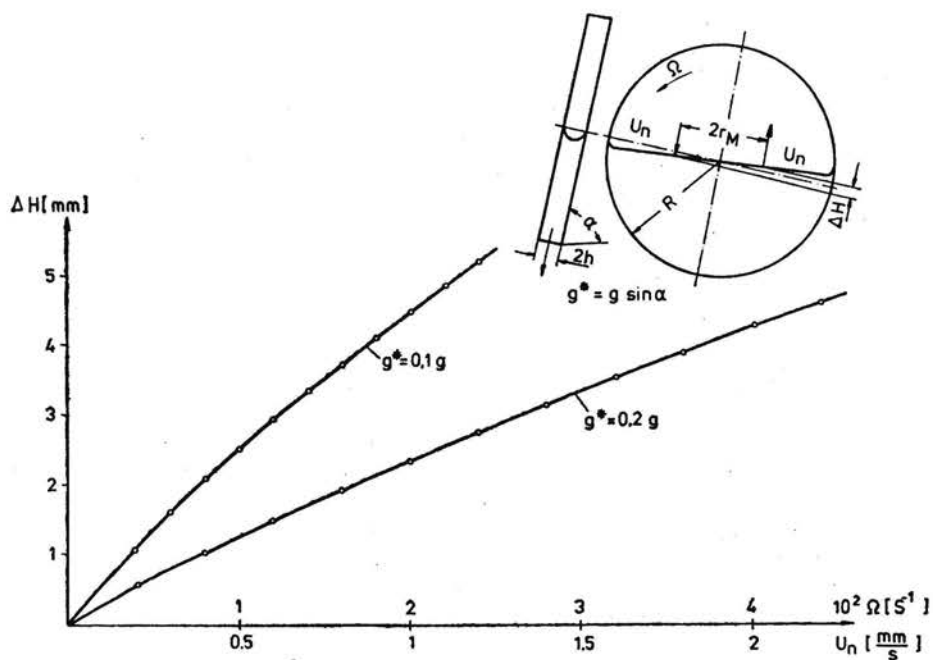


FIG. 10. Plot of  $\Delta H$  vs.  $\Omega$  and vs.  $U_n$ ;  $R = 140 \text{ mm}$ ,  $2h = 3 \text{ mm}$ ,  $2r_M = 100 \text{ mm}$ , disc material: frosted glass.

degressive trend of  $\Delta H(\Omega)$  may be explained by the effect of the thickness of the water-film on the frosted glass of the discs. In case of small wall speeds and thus small film thicknesses it is conjectured that the material properties of the frosted glass plates play a decisive role. However, a detailed comprehension of these properties is beyond the control of the experimenter. The remarkable influence of the water film thickness on the displacement of the interface can be demonstrated easily. If the angular speed is decreased suddenly, one observes that in the first instant the deviation of the interface from the equilibrium position is smaller than at the steady final state with the same angular speed but smaller film thickness. Experiments were made also with pure water. It was noticed that in this case  $\Delta H$  is considerably smaller than in the case of a liquid coated by a surface active agent. For an uncontaminated liquid the flow pattern in the neighbourhood of the interface is basically different from the corresponding pattern of a fluid which is contaminated by impurities.

### 3. Conclusions

In the preceding section the motion of a surface film in a two-phase flow has been dealt with under the very restrictive hypothesis that in a first approximation the film motion is independent of the liquid motion, the remaining gas being at rest. In reality both motions are coupled since shear stresses are exerted from the supporting liquid on the surface film, where the film is supposed to be a very thin, prestressed, elastic (or viscoelastic, respectively), insoluble and incompressible material layer of constant thickness and large surface (shear) viscosity, carried by a viscous homogeneous and incompressible Newtonian liquid. In spite of the restrictive assumptions, it has been shown that the simple theoretical model yields reasonable results. The very careful experimental investigations carried out by the junior author have provided test data which may serve to develop a more rigorous hydrodynamic theory. The problem under consideration is a problem with a free boundary. The latter is a boundary which is not prescribed, but which must be found as part of the solution of the problem. The surface of a moving interface in contact with vacuum, air or another fluid is, however, such a boundary.

### Reference

1. V. G. LEVICH, *Physicochemical hydrodynamics*, Prentice Hall, Englewood Cliffs, N. J., 1962.
2. K. L. WOLF, *Physik und Chemie der Grenzflächen*, Bd. 2, Springer 1959.
3. J. T. DAVIES, E. K. RIDEAL, *Interfacial phenomena*, Academic Press, New York und London, 1961.
4. G. L. GAINES, *Insoluble monolayers at liquid-gas interfaces*, Wiley and Sons, New York, 1969.
5. PH. FRANK, R. V. MISES, *Die Differential — und Integralgleichungen der Mechanik und Physik*, Band 1, 2. Auflage, Dover Publications, Inc. New York, Fr. Vieweg u. Sohn, Braunschweig 1961.
6. W. JOHANN, *Über den Einfluss von Oberflächenfilmen auf Strömungsvorgänge*, Dissertation, Technische Hochschule Darmstadt, 1977.
7. C. B. BREZENO und R. GRAMMEL, *Technische Dynamik*, Band 1, 2. Auflage, Springer 1953.

UNIVERSITÄT ESSEN, GESAMTHOCHSCHULE, FRG.

Received October 25, 1979.