

Report LR-601

# Surface Dilation and Fluid-Dynamical Behaviour of Newtonian Liquids in an Overflowing Cylinder I Pure Liquids

August 1989

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SURFACE DILATION AND FLUID-DYNAMICAL BEHAVIOUR OF  
NEWTONIAN LIQUIDS IN AN OVERFLOWING CYLINDER

I PURE LIQUIDS

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## Summary

The surface dilational viscosity  $\eta_s^d$  relates the surface tension under dynamical conditions  $\sigma_{\text{dyn}}$  to the relative surface expansion rate  $\frac{d \ln A}{dt}$ . The value of  $\eta_s^d$  can be determined by means of the overflowing cylinder technique. The dynamic surface tension is determined by the Wilhelmy plate technique. The key problem is to find the relation between the surface expansion rate and the flow field developed in the cylinder for a particular surface active liquid.

In this paper calculated surface velocity distributions and surface expansion rates are compared to experimental results for a pure liquid. Moreover some experimental results show the large influence of the addition of surfactants on the surface expansion rate.

## 1. Introduction

As we all know, surface active agents play an important role in emulsifying- and foaming properties of liquids. Experience shows that these properties strongly depend on both the nature and the amount of the applied surfactant(s).

The stability of the created foams and emulsions against coalescence is for the greater part determined by the stability of the thin liquid films in between respectively the air bubbles and the emulsion droplets. In turn, the stability of these thin liquid films largely depends on the interfacial properties of the liquid.

However, the liquid-air and liquid-liquid interfaces are continuously exposed to all kinds of disturbances. Especially during preparation, storage and usage of the dispersions, compression and expansion of the interfaces occur. So, examination of especially the dynamic interfacial properties of the liquid may increase our knowledge of the behaviour of dispersions.

This can be illustrated for instance by the behaviour of two dispersions, which contain a different kind of surfactant in different amounts, but have been composed in such a way that the equilibrium interfacial tension of the two dispersions is the same. It is experienced that the dynamic interfacial properties of the two dispersions, and as a result the overall stability of the created dispersions, may differ considerably.

One of the ways in which a mechanical disturbance can be applied to a liquid surface is by means of expansion or compression of that surface. In a so called dynamic measurement the surface is subjected to a sinusoidal change in surface area. In an other type of measurement the surface is continuously expanded or compressed.

Because a thin liquid film can be made unstable especially when it is subjected to an expansion, the study of the behaviour of an expanding surface can result in a better understanding of the foaming behaviour of a liquid.

One of the ways to accomplish a pure dilation (without any shear-components) is with the use of an overflowing cylinder. The physically well defined parameter, which can be determined with the aid of this method, is the surface dilational viscosity  $\eta_s^d$ , which is defined by the following equation:

$$\eta_s^d = \frac{(\sigma_{\text{dyn}} - \sigma_e)}{d \ln A / dt} \quad [\text{Nsm}^{-1}] \quad (1.1)$$

where  $\sigma_{\text{dyn}}$  represents the surface tension during expansion of the surface,  $\sigma_e$  the equilibrium surface tension when the surface is at rest and  $d \ln A / dt$  the relative expansion rate of the surface area A.

It has to be noted that  $\eta_s^d$  is not a constant and that interfaces do not behave like Newtonian systems: the experimental value of the surface dilational viscosity is namely strongly dependent on the value of  $d \ln A / dt$ .

With the overflowing cylinder technique expanding surfaces can be studied. Padday [1] and Piccardi and Ferroni [2,3] have already used the overflowing cylinder technique for dynamic surface tension measurements with various fluids, while Barber [4] and Joos and de Keyser [5] used an overflowing funnel as a method for measuring surface dilational properties. The key problem in these studies is how to relate the surface expansion rate to the flow rate through the funnel or the overflowing cylinder. This problem appears to be a difficult one and none of these investigations resulted in a more detailed description of the physical mechanism which explains the operation of the overflowing cylinder technique.

In practice expanding surfaces can be found during preparation of foams, nucleation of bubbles and during application of thin liquid layers on solid walls like paints, coatings, printing ink, etc. Knowledge of the dynamic interfacial properties of these liquids may lead to improvement of products or production processes.

Because of the observed lack of knowledge the overflowing cylinder technique will be elucidated in this article. First of all a description of the experimental set-up will be given, followed by a theoretical analysis of the flowfield in the overflowing cylinder. Finally results of measurements on pure water will be compared with data resulting from the theoretical treatment.

## 2. Experimental set-up

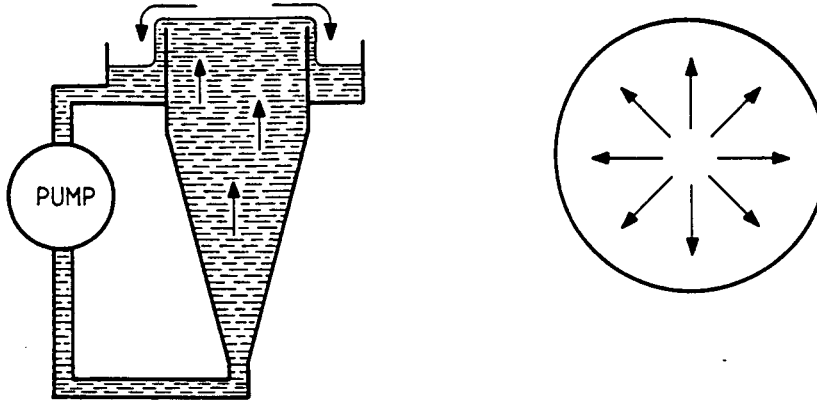


Figure 1: the overflowing cylinder technique. (not to scale)

The overflowing cylinder (see figure 1) consists of a metal cylinder (diameter 7.97 cm) through which a liquid is pumped upwards. In order to ensure a laminar flowfield in the cylinder, the fluid first passes through a conical tube, having a small slope (1:10), before it reaches the cylinder. Next the liquid is allowed to flow over the top rim of the vertical cylinder, causing the circular liquid surface to be expanded continuously in a radial way. We are dealing with a steady state situation: this means that the surface dilation pattern does not change in time. However, when of course the flowrate is altered, a new surface expansion rate will be established.

In order to obtain the value of the surface dilational viscosity  $\eta_s^d$ , the dynamic surface tension  $\sigma_{\text{dyn}}$  and the relative expansion rate  $d \ln A / dt$  have to be measured.

The dynamic surface tension can easily be measured by the Wilhelmy plate technique. The most obvious way to do this is in the centre of the surface. The measurement of the expansion rate of the surface, however, is much more complicated. From the symmetry of the cylinder and under the condition that the free surface is almost flat in the neighbourhood of the axis of symmetry it follows that the relative expansion rate depends on the radial velocity of the surface  $v_r(r)$ :

$$\frac{d \ln A}{dt} = \frac{v_r}{r} + \frac{\partial v_r}{\partial r} \quad (2.1)$$

This can be seen in the following way: consider a small surface area  $A$ , see figure 2, which is defined by the radius  $r$ , the radius  $r + \Delta r$  and the angle  $\alpha$  (in radians), as given by:

$$A = \frac{\alpha}{2\pi} \{ \pi(r + \Delta r)^2 - \pi r^2 \} \quad (2.2)$$

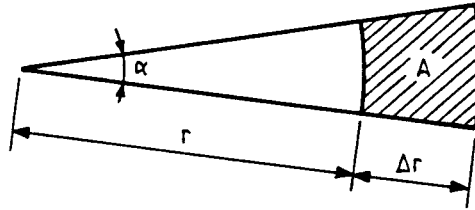


Figure 2: definition of the surface area  $A$ .

Retaining only first order terms the velocity of the surface at  $r + \Delta r$  can be expressed as:

$$v_r(r + \Delta r) = v_r(r) + \frac{\partial v_r}{\partial r} \cdot \Delta r \quad (2.3)$$

By means of equations (2.2) and (2.3) it follows that:

$$\begin{aligned} \frac{d \ln A}{dt} &= \frac{1}{A} \frac{dA}{dt} = \frac{\frac{\alpha}{2} \{ 2(r + \Delta r) \cdot \frac{d}{dt}(r + \Delta r) - 2r \frac{dr}{dt} \}}{\frac{\alpha}{2} \{ (r + \Delta r)^2 - r^2 \}} \\ &= \frac{\{ (r + \Delta r) \cdot (v_r(r) + \frac{\partial v_r}{\partial r} \Delta r) - r \cdot v_r(r) \}}{\Delta r(r + \frac{1}{2} \Delta r)} \\ &= \frac{v_r(r) + \Delta r \cdot \frac{\partial v_r}{\partial r} + \frac{\partial v_r}{\partial r} \cdot r}{r + \frac{1}{2} \Delta r} \end{aligned} \quad (2.4)$$

In the limiting case for  $\Delta r$  going to zero,  $\Delta r$  will be neglectibly small compared to  $r$  and consequently equation (2.4) will turn into equation (2.1). By means of this equation the relative surface expansion rate can be calculated if  $v_r(r)$  is known.

### 3. The flowfield of an overflowing cylinder

#### 3.1. A qualitative description

A semi-infinite cylindrical tube with a circular cross-section is positioned vertically in a gravitational field under atmospheric pressure  $p_0$  at room temperature  $T_0$ . The axis of the cylinder coincides with the direction of the acceleration of gravity (see fig. 3) and the cylinder is cut perpendicular to its axis.

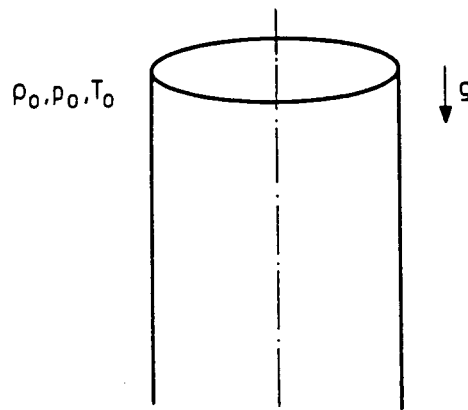


Figure 3: Geometry.

A Newtonian fluid having a density  $\rho_0$  is pumped upwards through the cylinder at a relatively low but constant rate. Hence the fluid rises slightly above the rim of the cylinder, flows over the edge and falls as a thin film downwards along the outside of the cylinder.

For this configuration the form of the fluid-air interface and the velocity field in the fluid that will be reached in the steady state will be analyzed and calculated in their dependence on the boundary conditions at the free surface.

#### 3.2. Formulation of the problem

In order to describe the flowfield in the overflowing cylinder quantitatively a cylindrical polar coordinate system is introduced. The axis of the coordinate

system coincides with the axis of the cylinder and the origin of the coordinate system is chosen at a distance H below the rim of the cylinder (see figure 4).

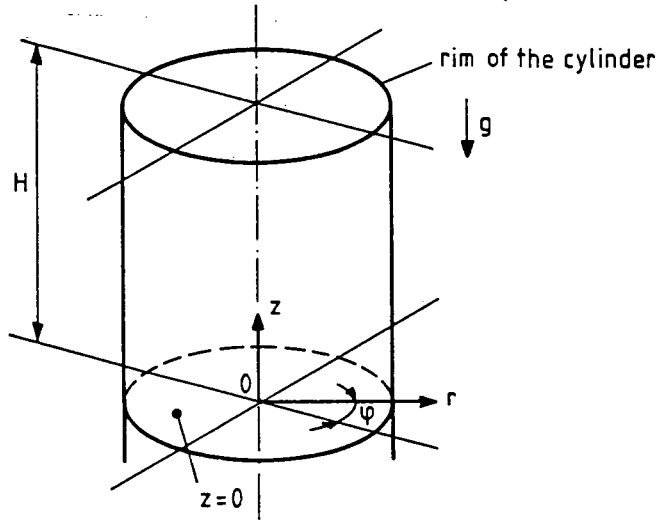


Figure 4.

The incompressible viscous flow in the cylinder is governed by the conservation equations: the conservation of mass and the three equations for the conservation of linear momentum: The Navier-Stokes equations. Since the geometry is axially symmetrical and the flow is stationary the Navier-Stokes equations reduce to:

$$\frac{\partial}{\partial r} (rv_r) + \frac{\partial}{\partial z} (rw) = 0 \quad (a)$$

$$v_r \frac{\partial v_r}{\partial r} + w \frac{\partial v_r}{\partial z} = -\frac{1}{\rho} \frac{\partial p}{\partial r} + \nu \left( \nabla^2 v_r - \frac{v_r}{r^2} \right) \quad (b) \quad (3.1)$$

$$v_r \frac{\partial w}{\partial r} + w \frac{\partial w}{\partial z} = -\frac{1}{\rho} \frac{\partial p}{\partial z} + \nu \nabla^2 w - g \quad (c)$$

In these equations the symbol  $\nabla^2$  indicates the Laplace operator

$$\nabla^2 = \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} + \frac{\partial^2}{\partial z^2} \quad (3.2)$$

$v_r$  and  $w$  are the velocity components in the  $r$  and  $z$  direction respectively,  $\rho$  is the fluid density,  $\nu$  is the kinematic viscosity,  $p$  is the pressure and  $g$  is the acceleration due to gravity.

The equations (3.1) describe the flow in a crossplane through the axis of the cylinder. Because of the symmetry of the flowfield only half of the velocity field has to be considered. (c.f. figure 5).

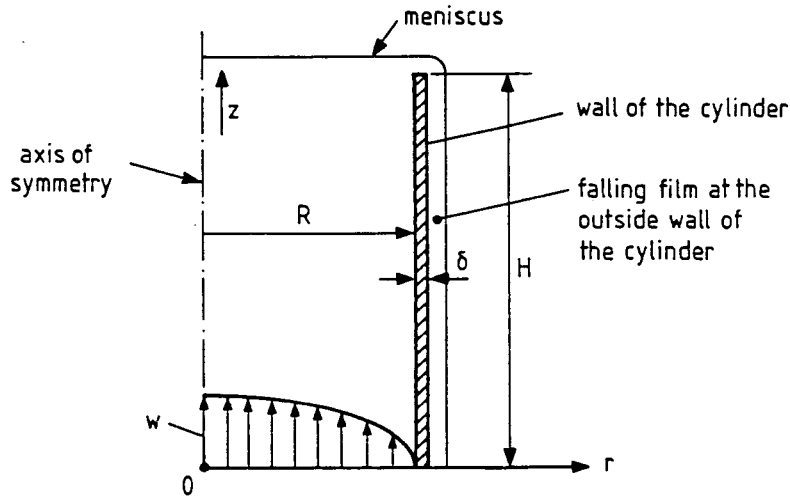


Figure 5.

If a proper set of boundary conditions is specified for the equations (3.1) a numerical solution for the velocity field and the pressure distribution can be obtained on a finite domain.

In the solution procedure a choice is made for an explicit time dependent strategy. In this strategy the flow is switched on at time  $t=0$  by imposing a velocity profile at the 'entrance' of the cylinder ( $z=0$ ) and after some time convergence to steady state flow is obtained. This implies that in the left hand side of the equations for the conservation of linear momentum (3.1) the terms  $\frac{\partial v_r}{\partial t}$  and  $\frac{\partial w}{\partial t}$  which were deleted in (3.1) are added again yielding the time dependent Navier-Stokes equations

$$\frac{\partial}{\partial r} (rv_r) + \frac{\partial}{\partial z} (rw) = 0 \quad (a)$$

$$\frac{\partial v_r}{\partial t} + v_r \frac{\partial v_r}{\partial r} + w \frac{\partial v_r}{\partial z} = - \frac{1}{\rho} \frac{\partial p}{\partial r} + \nu \left( \nabla^2 v_r - \frac{v_r}{r} \right) \quad (b) \quad (3.3)$$

$$\frac{\partial w}{\partial t} + v_r \frac{\partial w}{\partial r} + w \frac{\partial w}{\partial z} = - \frac{1}{\rho} \frac{\partial p}{\partial z} + \nu \nabla^2 w - g \quad (c)$$

One of the advantages of this strategy is that difficulties with boundary conditions on the velocity profile of the falling film on the outside of the cylinder are avoided. Introduction of a continuity condition at the point where the film leaves the calculation domain is sufficient.

In addition to these initial conditions the following boundary conditions are imposed.

On the inside and outside of the solid wall obviously a no slip condition applies so

$$v_r = w = 0 \text{ for } \begin{matrix} r = R \\ r = R + \delta \end{matrix} \quad \} \quad z \leq H \quad (3.4)$$

where  $\delta$  is the thickness of the wall.

In the axis of symmetry a free slip condition is imposed, hence both the velocity normal to the axis of symmetry  $v_r$  and the normal derivative of the velocity tangential to the axis of symmetry  $\frac{\partial w}{\partial r}$  are zero.

$$\begin{aligned} (v_r) &= 0 \\ &\} \text{ for } r = 0 \\ \left( \frac{\partial w}{\partial r} \right) &= 0 \end{aligned} \quad (3.5)$$

What remains are the boundary conditions to be applied at the free surface. Because of the axial symmetry the unknown position of the free surface may be described by

$$z = f(r) \quad (3.6)$$

where the function  $f(r)$  has to be found as a result of the calculations. On the outside of the surface the pressure is equal to the atmospheric pressure  $p_0$ .

The ultimate goal is to calculate the velocity profile in z and r direction for a Newtonian liquid taking into account the effect of a surface tension gradient operating at the free surface of the liquid. In order to build up the procedure of the calculations for this complex configuration first three simpler systems will be considered.

a) in which no surface tension is taken into account

b) in which a constant surface tension is taken into account (so no surface tension gradient)

and c) in which a constant surface tension gradient is taken into account.

It must be noted here that even the condition c) is a simplification of a practical situation because it is to be expected that the surface tension gradient will depend on r.

ad a) The simplest form is the free slip condition and a pressure of the liquid equal to the atmospheric pressure implying

$$\left. \begin{array}{l} p=p_o \\ \frac{\partial v_t}{\partial n} = 0 \end{array} \right\} \text{ for } z=f(r) \quad (3.7)$$

where n is the direction normal to the free surface and  $v_t$  is the velocity of the surface tangential to the surface. This means that the effect of surface tension is not taken into account.

ad b) If a constant surface tension is incorporated a more complex form of boundary condition is imposed. In that case the combination of free slip and capillary pressure is applied.

Hence

$$p = p_0 - \sigma \left( \frac{1}{R_1} + \frac{1}{R_2} \right) \quad \text{for } z=f(r) \quad (7)$$

$$\frac{\partial v_t}{\partial n} = 0$$

where  $\sigma$  is the actual value of the surface tension and  $R_1$  respectively  $R_2$  are the principal radii of curvature. For an axially symmetrical configuration these radii shown in figure 6 are given by

$$\frac{1}{R_1} = \frac{d}{dr} \frac{f'}{\sqrt{1+(f')^2}} \quad \text{for } z=f(r) \quad (8)$$

$$\frac{1}{R_2} = \frac{1}{r} \frac{f'}{\sqrt{1+(f')^2}}$$

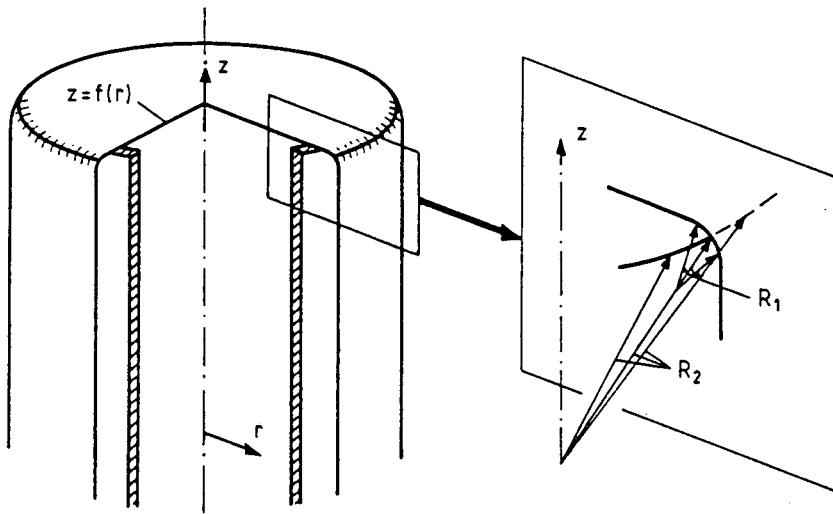


Figure 6.

ad c) The most complex form of boundary condition occurs if surface tension gradients are taken into account. In that case a tangential shear stress is present in the free surface. The tangential shear stress at the surface is balanced by the viscous shear stress in the fluid, yielding the following set of boundary conditions (see figure 7)

$$\frac{\partial v_t}{\partial n} = \frac{1}{\mu} \frac{\partial \sigma}{\partial s}$$

(9)

$$p = p_0 - \sigma \left( \frac{1}{R_1} + \frac{1}{R_2} \right)$$

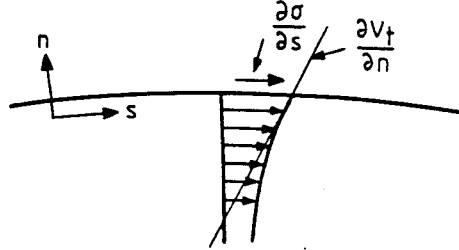


Figure 7.

In (9)  $\sigma$  again is the local surface tension,  $\frac{\partial \sigma}{\partial s}$  is the tangential derivative of the surface tension and  $\mu$  is the dynamic viscosity coefficient of the liquid.

### 3.3. Numerical solution technique and results.

In the solutions presented in this paper only free surface boundary conditions of type a) and b) have been taken into account. Calculations with boundary conditions of type c) are in preparation and results will be presented in a separate paper.

The unsteady Navier-Stokes equations are solved by an explicit Eulerian finite difference scheme on an orthogonal non-uniform Cartesian grid (see figure 8).

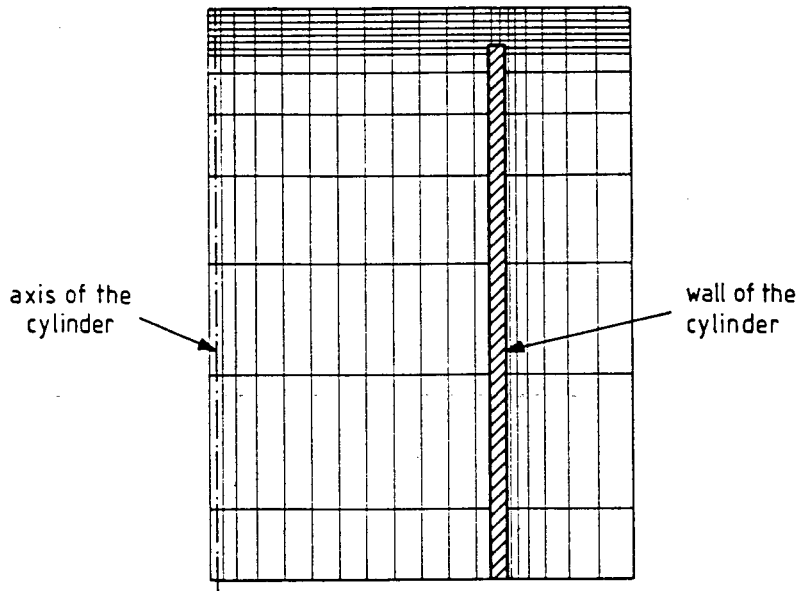


Figure 8.

The Newtonian fluid considered is pure water. At the start the cylinder is filled to the rim with fluid and at time  $t=0$  the flow is switched on. The displacement of the free surface is tracked and the velocity field is calculated. After some time the flow converges to a steady state.

This type of calculation has been performed for several mass fluxes with and without surface tension. A switch option to switch on or off surface tension during calculation has been build into the solution routine.

The results presented are the rising height of the interface above the rim of the cylinder calculated at the axis of symmetry and plotted as a function of the mass flow rate through the cylinder with surface tension switched on and with surface tension switched off (figure 9). It is noticed that the rising height is strongly dependent on the value of the surface tension. For moderate flow rates of a pure liquid good agreement exists between the calculated results and the experiments. For low flow rates experimental as well as calculated results show some instabilities (indicated by the dashed line in figure 9). These instabilities are probably caused by the falling film on the outside of the cylinder.

Further for a typical mass flow rate the velocity distribution in the fluid is presented with surface tension switched on (figure 10). From figure 10 it is noticed that in the neighbourhood of the rim of the cylinder the maximum radial

velocity is occurring below the free surface. This implies that the free surface is accelerated via viscous forces by the bulk fluid. With respect to the velocity distribution in the surface comparisons are made with experiments (see next section).

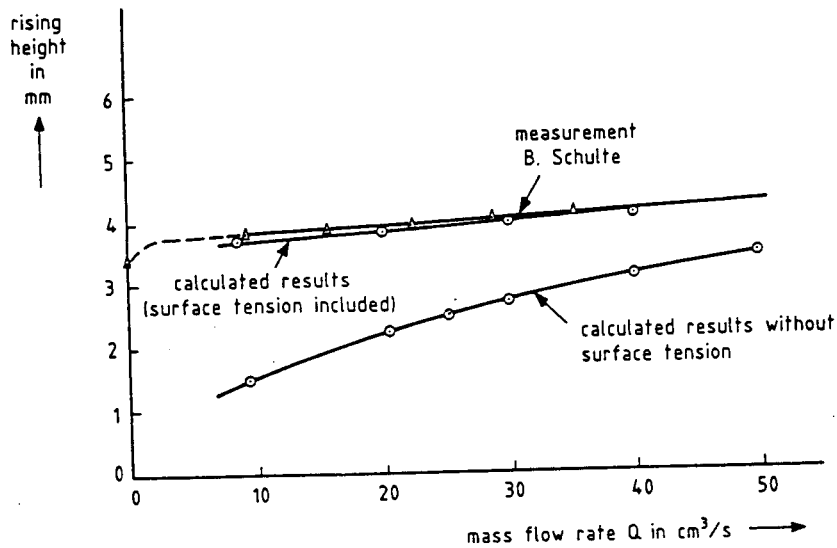


Figure 9.

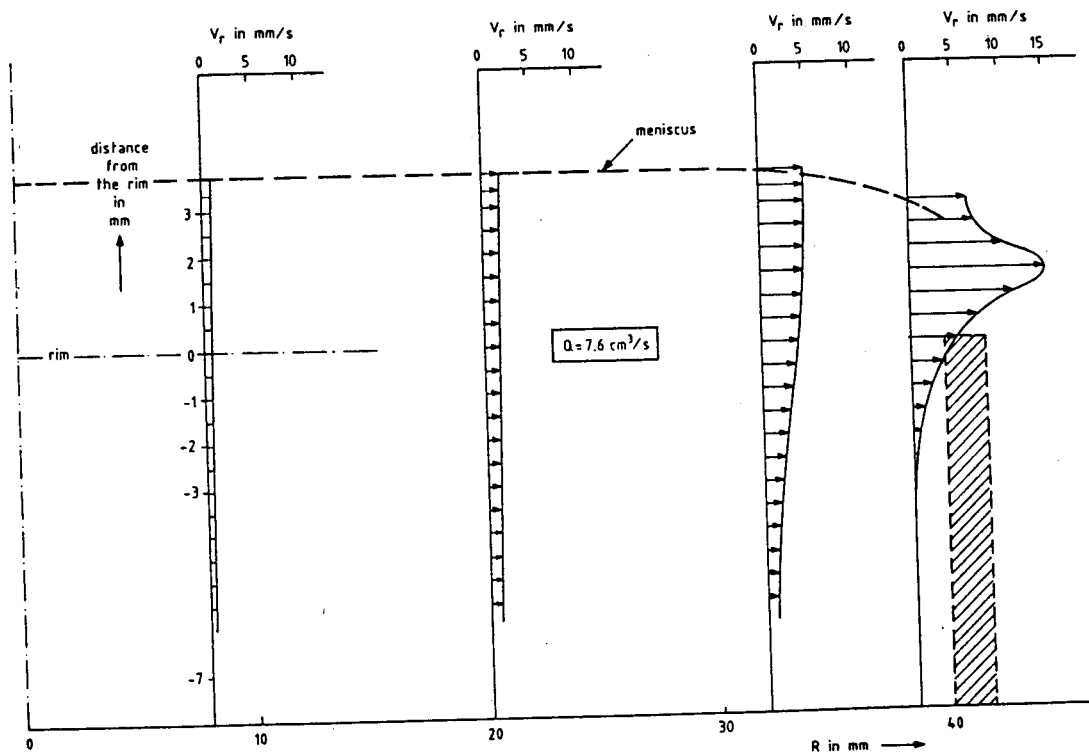


Figure 10.

#### 4. Experiments

In order to obtain the value of the surface dilational viscosity  $\eta_s^d$  the quantities  $\sigma_{\text{dyn}}$  and  $\frac{d\ln A}{dt}$  have to be known (compare equation 1.1). The dynamic surface tension can easily be measured by means of the Wilhelmy plate technique and the value of  $\frac{d\ln A}{dt}$  can be determined by measuring the radial velocity at the surface  $v_r(r)$  as a function of the distance  $r$  from the centre of the surface according to equation (2.1).

Before a measurement is carried out, the overflowing cylinder has to be adjusted in such a way that the cylinder is in a perfect vertical position. This ensures a purely radial symmetric velocity profile having a zero radial velocity of the surface at the centre of the circular meniscus. The dynamic surface tension  $\sigma_{\text{dyn}}$  and the radial velocity of the surface  $v_r(r)$  are a function of the flowrate. The flow through the cylinder can easily be adjusted and determined by means of a needle valve and a flowmeter. It has to be taken care of that the flowrate is adjusted in such a way that the outside wall of the cylinder is completely wetted by the falling film.

At present we are dealing with water and in that case the dynamic surface tension equals the equilibrium surface tension of pure water, independently of the flowrate. The radial velocity of the surface  $v_r(r)$ , is of course a function of the flowrate. The magnitude of  $v_r(r)$  is established by determining the velocity of small particles floating on the expanding surface. Here polyethylene particles with a diameter of 100  $\mu\text{m}$  and a density of 900  $\text{kg.m}^{-3}$  are used. Experimentally it is found that these particles do not influence the equilibrium surface tension of water. At the start of an experiment the polyethylene particles are sprinkled on the surface of the overflowing liquid as uniformly as possible. Shortly after that a photo of the expanding surface is taken, using two flashes with a fixed time interval. Both the time interval and the exposure time are chosen in such a way that the photo shows two sharp images of each particle. Figure 11 is an example of a photo obtained in this way.

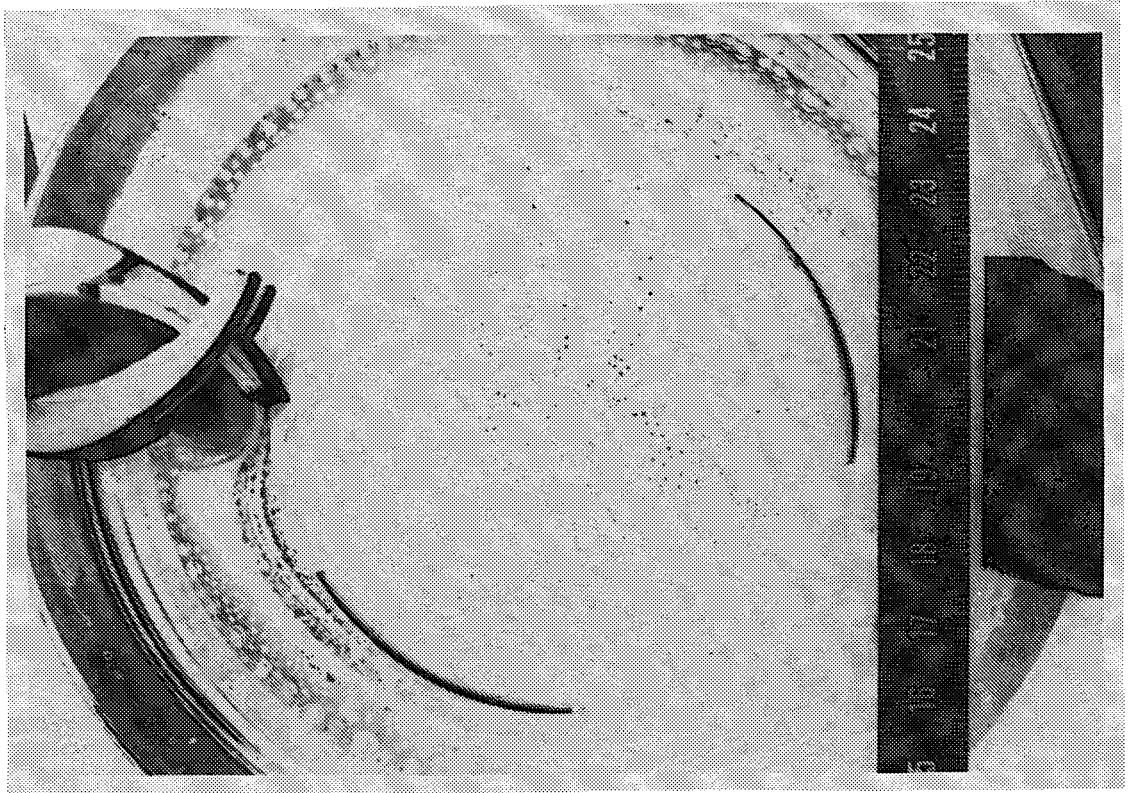


Figure 11: example of a photo of polyethylene particles floating on the expanding surface, taken by using two successive flashes.

The radial velocity of the surface  $v_r(r)$  is obtained from the distance between the position of a particle in the two images  $r_2 - r_1$  and the time lapse between the two flashes  $\Delta t$  according to:

$$v_r(r) = \frac{r_2 - r_1}{\Delta t} \quad (4.1)$$

This velocity belongs to a radial distance  $r$  for which as an approximation the mean distance is taken

$$r = \frac{r_2 + r_1}{2} \quad (4.2)$$

The resulting curves, like for example figure 12, present  $v_r$ , as a function of  $r$  at a certain flowrate. For reasons of comparison the numerically calculated distribution is also plotted in this figure.

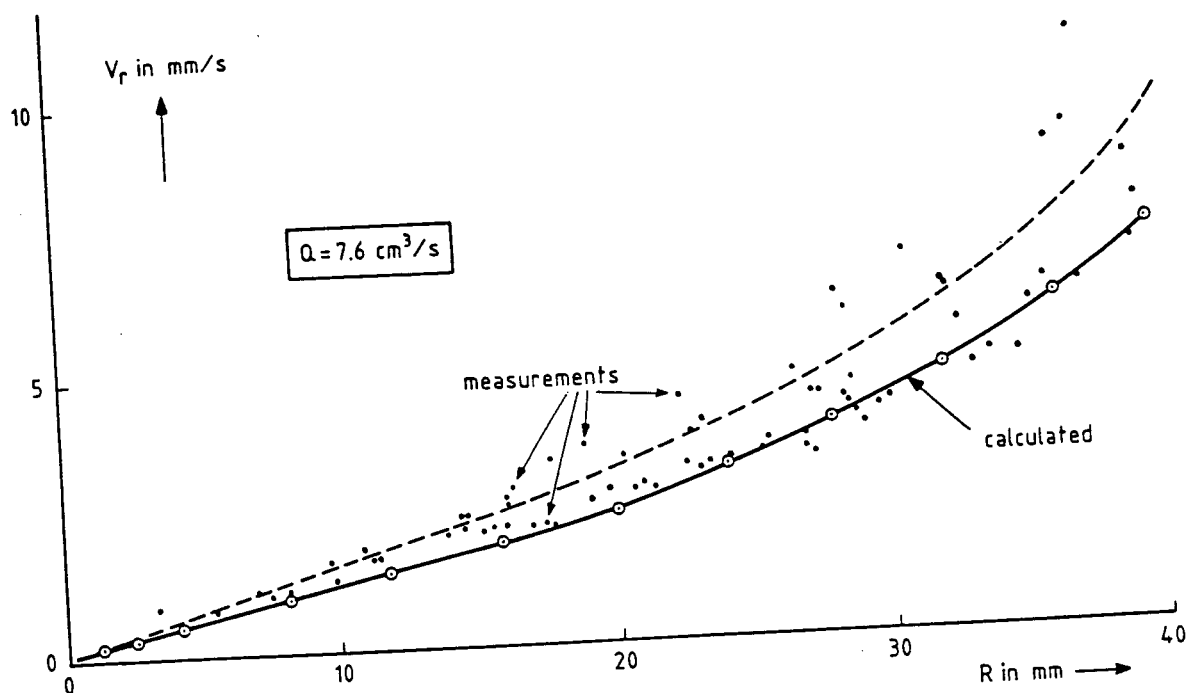


Figure 12: the velocity of the water surface as a function of the radius at a flowrate of  $7.6 \text{ cm}^3 \cdot \text{s}^{-1}$

Ultimately we are interested in the dynamic surface behaviour of a surfactant solution. Therefore it is interesting to notice that already the addition of a small amount of a surface active agent raises the velocity of the aqueous surface considerably (see figure 13). This phenomenon will be discussed in a forthcoming paper.

By means of equation (2.1) it follows from the figures 11 and 13 that the value of the relative surface expansion rate  $\frac{d \ln A}{dt}$  in an area near the centre of the circular meniscus equals  $0.29 \text{ s}^{-1}$  for water and equals  $3.37 \text{ s}^{-1}$  for Teepol. But since the dynamic surface tension of pure water equals the equilibrium surface tension, the surface dilational viscosity  $\eta_s^d$  is zero in the case of pure water.

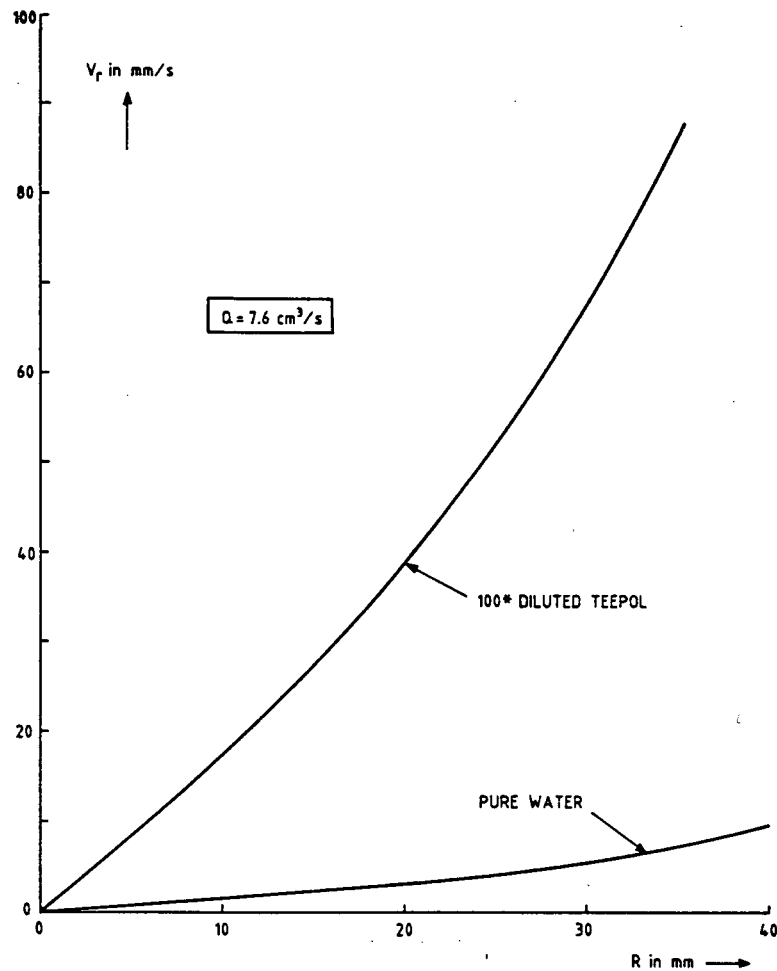


Figure 13: the velocity of the surface as a function of the radius at a flowrate of  $7.6 \text{ cm}^3 \cdot \text{s}^{-1}$  for

- water and
- 1 volume per cent dilution of a commercially available concentrated Teepol solution in water.

The height of the meniscus above the rim of the overflowing cylinder,  $h_{\text{dyn}}$  has been measured in the centre of the circular meniscus by means of a micrometer. First the position of the rim of the cylinder is established by positioning a rod of known thickness over the top rim of the cylinder, and next the position of the liquid surface is measured by screwing down the micrometer until its cone touches the liquid surface. The value of  $h_{\text{dyn}}$  in the centre of the circular meniscus is obtained from the difference between those two measured distances. The value of  $h_{\text{dyn}}$  of pure water as a function of the flowrate has been shown in figure 9. There too the measured values were compared with the results of a numerical calculation and good agreement was found.

## 5. Conclusions

It has been shown in this paper that for the parameters of a pure fluid flowing through the overflowing cylinder a better than qualitative agreement exists between calculations and measurements.

It has been noticed from the calculated results that for a pure fluid in particular in the neighbourhood of the rim of the cylinder the bulk fluid is driving the surface layer. If a small amount of surfactant is added surface tension gradients are introduced resulting in a large change in magnitude of the surface velocity. This suggests that in that case the maximum radial velocity indeed occurs at the free surface and the free surface is driving the bulk.

## 6. References

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