THEORETICAL AND EXPERIMENTAL RESULTS ABOUT ASYMMETRICAL DROPS AT REST UPON SURFACES HAVING REGULAR ROUGHNESS

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The theory of interfacial tension and related subjects have benefited by a renewed interest during the last thirty years. An important signal of the new trend was the conferment of the Nobel Prize for Physics in 1991 on the French physicist Pierre Gilles de Gennes, renowned theorist of the «soft matter».
A recent and promising trend is nanotechnology, which is exploiting the properties of thin liquid films and drops to construct various miniaturised devices. Another outstanding example is the liquid lens patented by a well-known international electronics firm: the shape of a tiny drop of liquid is modified by an electric field and the curved surface is used to focus a low-power laser.
The computation of the shape of a sessile or pendant drop at rest is a classical problem of Fluid Mechanics. The problem possesses relevant experimental interest to deduce the interfacial tension and the contact angle from the geometrical properties of the drop. The exact knowledge of both quantities is required by many mathematical models of two-phase flow. Especially the subject of oil-water flow has received considerable attention in our Department in Milan, with several papers already published in national and international conferences.
Nowadays a research project is undergoing which is coordinated by Prof. G Cossali at the University of Bergamo and embraces groups in four Universities investigating on thermal and dynamic interactions between sprays and solid surfaces.
Research in our laboratory has confirmed that wettability of the pipe exerts a remarkable influence on the «core annular» flow regimes: in particular «core annular flow», characterized by an oil core surrounded by a water annulus at the wall, gives rise to a minimum pressure drop.This kind of flow is in fact more effective and stable when the pipe is better wetted by water than by oil.
As for symmetrical drops at rest, first of all we have devised and plotted a new kind of graph, not previously found in the literature, in which the shape and size of a symmetrical (sessile or pendant) drop are related to the contact angle β and the Eötvös number $N_{Eö}$, defined as follows: $N_{Eö} = \frac{(\rho_{in} - \rho_{out})\, g\, V^{\frac{2}{3}}}{\sigma}$, where $\rho_{in} - \rho_{out}$ is the density difference between the drop and the surrounding fluid, g is the acceleration of gravity, V is the drop volume, σ is the surface/interfacial tension. Then the drop shape and size are described in dimensionless form by two families of contour lines: e.g. $\frac{H}{D_{\max}}$ and $\frac{D_{\max}}{\sqrt[3]{V}}$, where $D_{\max}$ is the maximum drop diameter and H is their height.

The graph was plotted by a custom made software written by the first author. The graph and the software have been used to check the experimental bench built up to measure the drops.
The bench is equipped with various instruments set up on purpose, including a calibrated syringe driven by a stepper motor and controlled by an electronic card and a high-resolution CCD TV camera to shoot the drops.
After the experimental equipment was properly checked and adjusted on symmetrical drops, whose theory is well known, the most interesting part of the experimental work began.
When the classical theory of interfacial tension is found at variance with experiments, the cause of the discrepancy should be investigated in other respects:
- the chemical homogeneity of the fluids;
- the chemical homogeneity of the solid surface;
- the geometrical regularity of the solid surface.

The difficulty to meet these requirements is that the slightest amount of contaminant could exert a relevant effect if it is localised upon the surface. Moreover the routine tool machining of solid surfaces leaves a roughness which is at least two orders of magnitude the size of a crystal cell in superpolishing and much greater otherwise.

It is known by experience that the larger the drop, the more difficult is to obtain an axisymmetric drop because the probability is enhanced to incur a chemical and/or a geometrical inhomogeneity of the solid surface. That is why the quite gross error of a few degrees in the contact angle is often accepted.

Since the theory of tool machining brings forward over a dozen parameters to describe the roughness of surfaces (none guaranteed to suit the purpose of interfacial tension research), hopes are dim to tackle the problem of drops at rest on rough surfaces using common mechanical surfaces. Because roughness is unavoidable in actual surfaces, only a smart idealisation of roughness may help to achieve the *repeatability* of experimental data.

A similar approach is already documented in the literature. In order to study the effect of variable wettability of the surface, another author has constructed artificial surfaces made of tiny rectilinear microstrips of two alternate materials. Drops laid on these surfaces elongate notably in the strip direction.

Similarly, in order to obtain a regular roughness which is exactly reproducible, we tried to employ a component widely used in Optics acting as a reference specimen: the diffraction grating. It provides two important advantages over common metallic, machine-tooled surfaces:

– roughness is unidirectional, consisting of parallel rectilinear grooves;
– the accuracy is guaranteed by the producer to be a small fraction of the wavelength of visible light.

Another useful surface having similar features in spite of sound modulation is the old microgroove record, whose groove profile is a right-angle symmetrical sawtooth.

From a theoretical point of view, the difficulties of computing the drop surface upon a diffraction grating are the loss of symmetry, because the drop is elongated in the groove direction, and the more complex boundary condition. Moreover the computational mesh size must be a fraction of the undulation wavelength which means that, for a large drop resting on a very closely spaced diffraction grating (having up to hundreds of lines per millimetre), huge meshes with millions of nodes should be used, which cause a heavy computational burden.

Besides the huge computational burden, this approach is not feasible when applied to actual machine-tooled metallic surfaces, whose roughness is neither regular nor exactly known apart from a few statistical parameters.

New ideas are needed to tackle the problem effectively. A new mathematical model, not previously found in the literature, has been developed which replaces the rough surface with an equivalent smooth plane. The “trick” is to replace the ideal static contact angle γ previously described with an equivalent contact angle γ_W which obeys the Wenzel equation (R. N. Wenzel, 1936):

$$\cos(\beta_W) = s\,\cos(\beta)$$

where s is the ratio of the actual to the planar area in the direction considered, always greater than 1. The dimensionless factor is easily computed when the roughness profile is perfectly known, as in the case of diffraction gratings. Since s depends upon the direction, β_W is anisotropic too.

The partial differential equation governing the drop interface was deduced using both the Laplace equation and the Calculus of variations, leading to the same equation in cylindrical coordinates:

$$\frac{R^3\dfrac{\partial^2 R}{\partial z^2} - R^2\left[1+\left(\dfrac{\partial R}{\partial z}\right)^2\right] + R\left\{\dfrac{\partial^2 R}{\partial z^2}\left(\dfrac{\partial R}{\partial \varphi}\right)^2 - 2\,\dfrac{\partial R}{\partial z}\,\dfrac{\partial R}{\partial \varphi}\,\dfrac{\partial^2 R}{\partial z\,\partial \varphi} + \left[1+\left(\dfrac{\partial R}{\partial z}\right)^2\right]\dfrac{\partial^2 R}{\partial \varphi^2}\right\}}{R\left\{R^2\left[1+\left(\dfrac{\partial R}{\partial z}\right)^2\right]^{\frac{3}{2}} + \left(\dfrac{\partial R}{\partial \varphi}\right)^2\right\}} + \frac{\Delta p(0) + (\rho_{in} - \rho_{out})\,g\,z}{\sigma} = 0$$

where R is the radial co-ordinate, φ is the azimuthal angle, z is the vertical coordinate, R is treated as a function of z and φ ; $\Delta p(0)$ is the excess pressure inside the drop at its apex. The z axis is oriented downwards, so that $\dfrac{\partial^2 R}{\partial z^2} < 0$ at the apex.

The above mentioned PDE contains the symmetrical drop as a special case. When R is independent of φ : $\dfrac{\partial R}{\partial \varphi} = 0$, then the PDE reduces to the ordinary differential equation of the symmetrical drop at rest on a smooth horizontal plane.

No analytical solution of the symmetrical drop equation, which is a 2nd order non-linear ODE, is known; an analytical solution is far less likely to exist in the asymmetrical case, which adds complexity. However the numerical solution is not obvious at all because not only the solution must obey the PDE, but must satisfy the boundary condition and the volume condition as well.
Although this problem pertains to the Statics of fluids, a similar difficulty is encountered in Fluid dynamics where a set of at least two groups of nonlinear PDEs must be solved simultaneously.
A numerical method has been developed which bears a resemblance to the SIMPLE method by Patankar and Spalding, which inspired much of the later work.

Two estimates $R = R(z, \varphi)$ and $R' = R'(z, \varphi)$ are to be employed. At the beginning of the iteration $R' = R'(z, \varphi)$ is constructed as an estimate having a given shape (for instance ellipsoidal), an arbitrary volume and obeying the boundary condition (governed by the Wenzel equation). Afterwards the estimate is uniformly magnified in all directions so as to embrace the given drop volume. It can be easily seen that, thanks to the nature of the Wenzel equation, the magnification leaves the boundary condition unaltered.
Then a central difference discretization of the PDE is performed which casts the PDE in a nominally linear form because, apart from the highest order derivatives $\frac{\partial^2 R}{\partial z^2}$ and $\frac{\partial^2 R}{\partial \varphi^2}$ in the first fraction and z in the second fraction, all other occurrences of $R = R(z, \varphi)$ are replaced by the known estimate $R' = R'(z, \varphi)$. Discretization is performed in such a way that respects the «all positive coefficients» rule in order to avoid unphysical oscillations in the solution.
Thus the PDE turns into a large system of linear equations which can be solved with one of the usual methods (for instance the «conjugate gradient» method).

The $R = R(z, \varphi)$ estimate, which obeys the PDE but not the boundary condition and the volume condition, is corrected first locally to satisfy the boundary condition and then globally, with a uniform magnification, to correct the volume condition too. This way $R = R(z, \varphi)$ is turned into a new $R' = R'(z, \varphi)$, which is ready to start another iteration.

The numerical method goes on until converges within a chosen threshold.
Experiments have been carried out on a diffraction grating etched with 20 lines per millimetre and an obtuse-angles triangular profile. As a consequence of the anisotropic roughness, drops of water laid on the grating elongate in the groove direction, taking an approximately elliptic shape. Photographs have been taken showing the drop contour on the grating and two profiles along the groove direction and the perpendicular direction.
The comparison between numerical and experimental data shows a satisfactory agreement: in the worst case the predicted size differs from the observed size less than 8%, but in most cases the agreement is much better.
As for the other option, a 33 r.p.m. microgroove record has been used. The groove spacing is 20 micrometers and the groove angle at the apex is 90 degrees. The sharper edge implies an interesting consequence. The drop is no longer free to spread on the surface because the liquid boundary is somewhat stopped by the crests; the parallel crests act as a sort of «trench» which prevent the liquid to «advance». Hence drops take a peculiar «biscuit-shaped» border.
The study of liquid drops laid on solid surfaces having quite regular roughness is attractive from the point of view of heat transfer, because rough surfaces may be interpreted as micro-finned surfaces which are known to enhance heat transfer. This kind of research is expected to be the future development of the theme.