Proceedings of the Tenth Asian Congress of Fluid Mechanics
17-21 May 2004, Peradeniya, Sri Lanka

ON THE CONTACT ANGLE IN UNSTEADY FLOWS

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ABSTRACT: We show that in general, the specification of a contact angle condition at the contact line in inviscid fluid motions is incompatible with the classical field equations and boundary conditions generally applicable to them. The limited conditions under which such a specification is permissible are derived; however, these include cases where the static meniscus is not flat. In view of this situation, the status of the many 'solutions' in the literature which prescribe a contact angle in potential flows comes into question. When the fluid is viscous, it is shown that if the contact line is pinned the contact angle has to remain constant. Some implications of this result are discussed.

1. INTRODUCTION

Consider a liquid, under a passive, inert gas, partially filling a smooth walled container. A gravitational field acts on the liquid. The liquid gas interface is subject to surface tension. The interface meets the walls of the container at a line called the contact line. At any point on the contact line, the angle between the normal to the gas-liquid interface and the normal to the solid wall is called the contact angle, α. In the quiescent state, for many pure materials and smooth solid surfaces, the contact angle for many gas/liquid/solid systems is a function of the materials alone. Here, we will assume this to be true and call this contact angle, the static contact angle α_s.

The surface tension at the gas-liquid interface, from now on called the interface, requires that there be a jump in the normal stress across it if it is not flat. In the inviscid case the jump is in the pressure and for a static interface the liquid pressure is just the hydrostatic pressure. Thus the shape of the static interface depends on a Bond number Bo, the ratio of a measure of the gravitational force to a measure of the force due to surface tension. But this shape also depends on the static contact angle α_s, which provides a boundary condition for the differential equation that determines the static interface shape. The static contact angle is therefore a parameter that influences the static meniscus shape and needs to be prescribed in order to calculate the meniscus shape.

When the interface is in motion, the surface tension again requires that a pressure jump, proportional to the interface curvature and to the coefficient of surface tension, exist across it. While the pressure in the liquid will now be determined by the unsteady Bernoulli equation in the inviscid case, in the viscous case the normal stress will be determined from the Navier-Stokes equations. The situation as regards the contact angle, however, is more complicated and confused. If one examines the dynamical equations for the interface it is not at all obvious that one needs to prescribe a condition on the contact angle. Moreover, as is well known, for the case of linearized disturbances between plane, vertical walls where the initial interface is plane, the classical inviscid solution can be obtained without any specification of the contact angle, which turns out to be constant and equal to $\pi/2$ throughout the motion. On the other hand, there are many examples in the literature where analyses and calculations have been made of unsteady potential motions where a condition on the contact angle has been prescribed as a boundary condition at the contact line. Just a short list of these could include Billingham[3], Miles[7] and Shankar[10]. It appears that the motivation to prescribe the contact angle comes from the apparent behaviour of real, viscous interfaces and the need to tailor inviscid models so that they lead to realistic results for real interfaces. The question that we raise here is: are we really free to prescribe the contact angle in inviscid potential flows?

When the liquid is viscous, the situation is made more complicated by the uncertainties associated with a moving contact line, continually made of new material particles. We will restrict ourselves here, in the viscous case, to motions in which it is fixed. Also, since we do not want to deal with situations where the static meniscus itself is not unique, we will through out assume that there is no contact angle hysteresis. The method we use is very similar to the one used by Benjamin & Ursell[2] ; however we will not restrict ourselves to the linearized case or to one where the mean interface is flat and there will be no restriction on the shape of the container. We will in fact show that the contact angle is actually constant for this class of viscous motions.

Some implications of these results will be discussed.

2. ANALYSIS OF INVISCID FLOWS

We consider the inviscid motion of a liquid in an arbitrary, three-dimensional smooth walled container. The motion is generated by the translational motion of the container. The restriction to translational motions is essential to ensure potential flow in the moving frame, a necessary condition for some of the results that will be derived. The fluid is initially in static equilibrium with the gas above it which is at uniform pressure.[1] The motion is assumed to start and continue with a uniform pressure over the interface; we will assume the gas to be passive, i.e. it only exerts a constant pressure on the liquid interface. In §2.1, we write down the equations governing the motion. In §2.2, we consider planar motions and in §2.3 we summarise the main results.

2.1 Governing equations

We write the equations in a reference frame attached to the container. The container wall is given by $f(x, y, z) = 0$. Rectangular cartesian coordinates are employed with gravity generally in the negative z-direction. Our analysis is restricted to the case when the interface is representable by a single valued smooth function, e.g. by $z = \zeta(x, y, t)$. The interface motion can be of finite amplitude however. The equations governing the liquid motion are the continuity and Euler equations -

$$\nabla \cdot \mathbf{u} = 0, \tag{1}$$

$$\mathbf{u}_t + (\mathbf{u} \cdot \nabla)\mathbf{u} = -\nabla p + \mathbf{F} \tag{2}$$

where $\mathbf{u}$ is the liquid velocity, p is the pressure and $\mathbf{F}$ is the net body force, both real and fictitious. $\mathbf{F}$ can be an arbitrary function of time. These have to be solved subject to the boundary conditions on (a) the container wall and (b) the interface. The condition on the wall is the no-penetration condition $\mathbf{u} \cdot \nabla \mathbf{f} = 0$. The conditions on the interface $z = \zeta(x, y, t)$ are

$$\zeta_t + u\zeta_x + v\zeta_y = w, \tag{3a}$$

$$p - p_a = -\frac{1}{Bo}\kappa \tag{3b}$$

[1] The condition of static equilibrium can be relaxed and, in fact, has to be in the case of time periodic wave motions.

which respectively are the kinematic condition on the interface and the normal stress condition on it. Bo is a suitably defined Bond number, p_a is the constant ambient pressure over the interface and κ is the local interface curvature. The interface $z = \zeta(x, y, t)$ is assumed to intersect the container wall $f(x, y, z) = 0$ in a smooth curve called the contact line. It is further assumed that ζ is analytic in all variables. The possibly time dependent angle α made by the contact line with the wall is given by

$$\frac{f_z - f_x\zeta_x - f_y\zeta_y}{|\nabla f|[1 + \zeta_x^2 + \zeta_y^2]^{\frac{1}{2}}} = \cos\alpha(t) \tag{4}$$

where (4) is to be evaluated at any point $(x_c(t), y_c(t), \zeta(x_c(t), y_c(t), t))$ on the contact line. We will show that no *a priori* prescription of $\alpha(t)$ is possible though in a few cases, it turns out to be a constant equal to its initial value α_s.

2.2 Planar inviscid motions with flat side walls

The container is straight walled, but not necessarily rectangular, and the motion is 2-D in the $x - z$ plane. The contact line in this case consists of just two points (A and B, say). The x and z axes are chosen such that, locally, the body surface is a line of constant x, say $x = 0$. There will be an x component of gravity in this case; the gravity direction has no bearing on the analysis however. The no-penetration condition implies $u(0, z, t) = 0$ from which it follows that $u_{z(n)}(0, z, t) = 0$ where $u_{z(n)}$ is the n^{th} derivative of u with respect to z.

Now, differentiate (3a) with respect to x to obtain an equation valid on the interface and hence on the contact line -

$$\begin{aligned}\zeta_{xt}(x,t) + u(x, \zeta(x,t), t)\zeta_{xx}(x,t) + [u_z(x, \zeta(x,t), t)\zeta_x(x,t) \\ +u_x(x, \zeta(x,t), t)]\zeta_x(x,t) = w_x(x, \zeta(x,t), t) + w_z(x, \zeta(x,t), t)\zeta_x(x,t).\end{aligned} \tag{5a}$$

Using the fact that an inviscid flow starting from rest has to be irrotational both in an inertial frame and a frame translating with respect to the inertial, we get $u_z - w_x = 0$, which with the continuity equation $u_x + w_z = 0$ allows (5a) to be written in the form

$$\zeta_{xt} = u_z - (2u_x + \zeta_x u_z)\zeta_x - u\zeta_{xx}. \tag{5b}$$

where the arguments in (5a) have been dropped to reduce the clutter. When (5b) is applied at the contact line we obtain

$$\zeta_{xt}(0,t) = -2u_x(0, \zeta(0,t), t)\zeta_x(0,t). \tag{6}$$

Note that, if the container were to be rotating as well, $u_z - w_x \neq 0$ and the above analysis would not apply. (6) shows that ζ_{xt} at the contact line cannot be specified arbitrarily; it certainly is not zero in general. This means that the contact angle changes with time in a manner that cannot be prescribed beforehand. However, for $\alpha = \pi/2$, not only is $\zeta_{xt}(0,0) = 0$ but also $\zeta_{xt^{(k)}}(0,0) = 0$ for all $k = 2, 3, \cdots$. We will show this by mathematical induction. Let the induction proposition be

$$P(k) : \zeta_{xt^{(m)}}(0,0) = 0 \;\forall\, m = 0, ..., k.$$

$P(0)$ is true because $\alpha_s = \pi/2$. Assume $P(k)$ is true; we will show that $P(k+1)$ is true. Rewriting (5b) as $\zeta_{xt} = u_z - a(u,\zeta)\zeta_x - u\zeta_{xx}$, the k^{th} time derivative of this equation can be written

$$\zeta_{xt^{(k+1)}} = u_{zt^{(k)}} - [a\zeta_x]_{t^{(k)}} - (u\zeta_{xx})_{t^{(k)}}.$$

With respect to the above equation, we observe the following -

1. $\partial\{u_z\}/\partial t^k = 0$ at x = 0 $\forall$ k = $0, 1, 2, \cdots$

2. $\partial\{a\zeta_x\}/\partial t^k = \sum_{m=0}^{k} b_m a_{t^{(k-m)}} \zeta_{xt^{(m)}}$ where b_m is the binomial coefficient $C(k,m)$. By the truth of $P(k)$, this sum is zero at $x = 0$.

3. $\partial\{u\zeta_{xx}\}/\partial t^k = \sum_{m=0}^{k} C(k,m) u_{t^{(k-m)}} \zeta_{xxt^{(m)}}$. This sum is zero $\forall\, k = 0, 1, \cdots$ as u and all its time derivatives vanish on the contact line.

Thus we have $\zeta_{xt^{(k+1)}}(0,0) = 0$ which means $P(k+1)$ is true. Thus, by mathematical induction, $P(n)$ is true $\forall n = 0, 1, \cdots$ and so $\zeta_{xt}(0,t) = 0$. This in turn implies that $\zeta_x(0,t) = 0$ for all time and the contact angle remains at $\pi/2$ for all time. Some observations are noteworthy -

1. Irrotationality of the motion is necessary but not sufficient.

2. This is a nonlinear result i.e. it holds irrespective of the perturbation amplitude and the shape of the static meniscus as long as the initial contact angle α_s is $\pi/2$. In particular, the static meniscus need not be flat.

3. The result holds as long as the region of the body surface over which the contact line moves is flat. The shape of the body elsewhere is immaterial.

2.3 A summary of the results in the inviscid case

A similar analysis can be carried out for 2-D inviscid flows where the side walls are curved; it is found that the contact angle is not preserved even for linearized motions with $\alpha_s = \pi/2$. In 3-D it is found that apart from small disturbances to a flat interface ($\alpha_s = \pi/2$) in a right cylinder of arbitrary cross section, the contact angle is not preserved. Thus the important point is that in inviscid fluid motions starting from rest in a container, the contact angle cannot be prescribed in advance and neither is there need for such a prescription. However, if one insists on prescribing the contact angle, this would necessarily result in a 'weak-type' solution - one that is in violation of the actual behaviour of the contact angle. However, in the special case of $\alpha_s = \pi/2$, there exist situations where the contact angle remains constant throughout the motion. These situations include cases of curved static menisci.

3. ANALYSIS OF VISCOUS FLOWS

We write the equations in a reference frame attached to the moving container. The container wall is given by $f(x,y,z) = 0$. Rectangular cartesian coordinates are employed with gravity in the negative z-direction. Our analysis is restricted to the case when the interface is representable by a single valued smooth function i.e. by $z = \eta(x,y,t)$. The interface motion can be of finite amplitude however. The equations governing the liquid motion are the continuity and the Navier-Stokes equations -

$$\nabla \cdot \mathbf{u} = 0, \tag{7a}$$

$$\mathbf{u}_t + (\mathbf{u} \cdot \nabla)\mathbf{u} = -\nabla p + \mathbf{F} + \frac{1}{Re}\nabla^2 \mathbf{u} \tag{7b}$$

where $\mathbf{u}$ is the liquid velocity, $\mathbf{F}$ is the net body force, both real and fictitious and Re a suitably defined Reynolds number. $\mathbf{F}$ can be an arbitrary function of time. These have to be solved subject to the boundary conditions on (a) the container wall and (b) the interface. The condition on the wall is the no-slip condition $u = v = w = 0$ where u, v and w are the x, y and z components of velocity. The conditions on the interface $z = \eta(x, y, t)$ are[6]

$$\eta_t + u\eta_x + v\eta_y = w, \tag{8a}$$

$$\eta_x(v_z + w_y) - \eta_y(u_z + w_x) + 2\eta_x\eta_y(u_x - v_y) - (\eta_x^2 - \eta_y^2)(u_y + v_x) = 0, \tag{8b}$$

$$2\eta_x^2(u_x - w_z) + 2\eta_y^2(v_y - w_z) + 2\eta_x\eta_y(u_y + v_x) + (\eta_x^2 + \eta_y^2 - 1)\{\eta_x(u_z + w_x) + \eta_y(v_z + w_y)\} = 0, \tag{8c}$$

$$p - \frac{2}{Re}\frac{\eta_x^2 u_x + \eta_y^2 v_y - \eta_x(u_z + w_x) - \eta_y(v_z + w_y) + \eta_x\eta_y(u_y + v_x) + w_z}{1 + \eta_x^2 + \eta_y^2} = p_a - \frac{1}{Bo}\kappa \tag{8d}$$

which respectively are the interface integrity, the two tangential stress and the normal stress conditions on the interface.

Analysis similar to that in §2 shows that if the contact line is pinned the contact angle α is constant and remains at its initial value α_s.

4. DISCUSSION

Recall that in the classical works the question of the contact angle never arose because confined flows with boundaries and capillarity were not normally studied. Early studies on the latter were confined to linearized, two-dimensional flows between vertical walls; here the classical, exact solutions correspond to a contact angle of $\pi/2$, which is maintained throughout the motion. In the 1960s the space programmes required solutions to more general problems involving curved static interfaces and static contact angles other than $\pi/2$. The difficulty posed by these problems forced approaches that were either semi-analytical or numerical and some like Moore & Perko[8] did not attempt to impose a dynamical contact angle condition; α_s affected the initial conditions alone through the static meniscus. The imperative to impose a contact angle condition at the contact line, not permitted in general by the classical inviscid formulation, appears to have come from experimental observations of real, viscous contact lines. It is well known[4] that real, dynamic viscous contact lines display complex behaviour and are not at all well understood with many parameters playing a role. It is in attempting to model this complicated behaviour in an inviscid frame work that the need for contact angle conditions began to be felt and then applied. The earlier models of a constant contact angle and the one used by Reynolds and Satterlee[9] (essentially to model contact angle hysteresis) are in a sense non-dynamic. Hocking's[5] model is a dynamic one, attempting in an inviscid framework to account for contact line hysteresis or viscous wetting effects at the contact line. In any case, the purpose is to account for viscous and other real effects in a inviscid, potential model of the flow.

Thus the need to more realistically model the dynamic contact line appears to require the freedom to *impose* a condition at the inviscid contact line. But as was shown in §2 the classical field equations and boundary conditions do not in general provide this freedom. Then the natural question is, what is the status of the very large and important body of work in which a contact angle condition is imposed, in violation of the classical formulation? Let us call solutions which are obtained without such a condition 'classical'. Then this body of work referred to does not deal with classical solutions. This means that these 'solutions' will be found to violate at least some of the boundary conditions at the contact line. The best of these will exactly satisfy the field equations and boundary conditions everywhere else. For want of a better terminology, we will call these solutions 'weak-type solutions', well aware of how imprecise this is.[2] . Even if the 'solutions' found in the literature were exact it is unlikely that they would all fall into the same class of 'weak-type solutions' because the formulations are so different. To make matters worse all the 'solutions' in the literature are approximate; there are no exact solutions even for the simplest of problems. However, the situation is not as bad as it appears. Checks are made between different approaches and with experimental results for the same problem; when there is reasonable agreement between these, which can be improved with further work, one can have some confidence that, at least from a phenomenological point of view if not a mathematical one, the 'solutions' are satisfactory.

On the other hand when the fluid is viscous, we have shown that if the contact line is pinned the contact angle has to remain constant. There are a number of important implications of this result. For example, if a viscous liquid is slowly draining from a container, the contact angle has to remain constant in the draining film at the wall, at least as long as the continuum model holds. Similarly if a plate is withdrawn from a viscous liquid the contact angle has to remain constant. Other interesting consequences will be considered in the talk.

5. CONCLUSION

We have shown that the classical field equations governing the motion of a confined inviscid liquid under a passive gas do not, in general, permit the independent specification of a contact angle condition at the contact line. In fact the only cases where such a condition may be permissible are when the static contact angle is $\pi/2$, the container is straight walled and (i) the motion is two-dimensional or (ii) the motion is a small three-dimensional disturbance from a flat initial interface. The restrictions are indeed surprising as is the difference between two- and three-dimensional motions.

These results have a somewhat serious bearing on the vast literature that exists in which 'solutions' have been found to inviscid motions in which various contact angle conditions have been imposed. It is our contention that these cannot be classical solutions to the classical field equations since classical solutions do not permit the imposition of a contact angle condition. It is suggested that these 'solutions' belong to an improperly defined class of 'weak-type solutions', in the sense that they attempt to solve the field equations in an approximate sense, with some of the equations being solved exactly. The need for such 'solutions' is driven by the compulsion to try to model in an inviscid frame work, the complicated behaviour of moving viscous contact lines.

For a viscous liquid, the contact angle has to remain constant if the contact line is pinned. This result has a number of important consequences, for example in liquid draining and in the drag out problem.

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