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Influence of gravitational forces and fluid flows on the shape of surfaces of a viscous fluid of capillary size

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The Navier-Stokes equations and boundary conditions for viscous fluids of capillary size are formulated in curvilinear coordinates associated with a geometry of the fluid-gas interface. As a result, the fluid dynamics of drops and menisci, which takes into account the influence of gravitational forces and flows on the surface shape, can be described. This gives a convenient basis for numerical studies. Estimations of the effects are presented for the case of an evaporating sessile drop.

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A number of important physical features in studying fluid flows in evaporating liquid drops and menisci of capillary size have been found recently both theoretically and experimentally [1–8]. In particular, it was demonstrated that the vortex convection takes place in evaporating drops and menisci under various conditions [5–8]. The activity in the field is associated now with important applications. Particular examples are the evaporative contact line deposition [1,2,4,9–11], studies of DNA stretching behavior and DNA mapping methods [12–14], developing methods for jet ink printing [15–17], and self-assembly of nanocrystal superlattice monolayer [18–20].

For describing the processes theoretically, one should carry out, in general, a joint study of the fluid dynamics, the thermal conduction, and the vapor diffusion together with respective boundary conditions, in particular at the fluid-gas interface. Standard approximations used in theoretical studies are a spherical cap shape of the drop or menisci and neglect of the hydrodynamical pressure and velocities in the generalized Laplace formula. Though such approximations can be justified under certain conditions, there is a wide range of parameters of the problem when a more accurate theoretical description of liquid surfaces of capillary size is needed.

The shape of a surface is, generally, controlled by the combined effects of surface tension, gravitational forces, hydrodynamic pressure, and velocity distribution near the surface. For solving fluid dynamics problems in the vicinity of curved surfaces of an arbitrary shape, an explicit approach is developed in the present paper, making use of "natural" curvilinear coordinates associated with a surface geometry. Both fluid dynamics equations and the respective boundary conditions are formulated in these coordinates. Equations in such a form are convenient for numerical simulations. We also present analytical estimations for the effects in question for the case of an evaporating sessile drop, which follow from the obtained results.

The Navier-Stokes equations take the form

$$\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v}\cdot\boldsymbol{\nabla})\mathbf{v} + \frac{1}{\rho}\,\mathrm{grad}\,p = \nu\Delta\mathbf{v}. \qquad (1)$$

For simplicity, we assume below that the shape of the surface as well as fluid flows is axially symmetric and $v_\theta=0$, where r,θ,z are cylindrical coordinates. This property is valid for a wide class of problems. Therefore, it is convenient to use cylindrical coordinates in the bulk of an incompressible viscous liquid and to introduce the vorticity $\gamma=\partial v_r/\partial z-\partial v_z/\partial r$ and the stream function ψ, such that $\partial\psi/\partial z=rv_r$ and $\partial\psi/\partial r=-rv_z$ (as distinct from the two-dimensional case [21]). Then rot $\mathbf{v}=\gamma(r,z)\mathbf{i}_\theta$ and the continuity equation div $\mathbf{v}=0$ is naturally satisfied. Equations for the quantities γ and ψ are given by

$$\frac{\partial}{\partial t}\gamma(r,z) + (\mathbf{v}\cdot\boldsymbol{\nabla})\gamma(r,z) = \nu\left(\Delta\gamma(r,z) - \frac{\gamma(r,z)}{r^2}\right), \qquad (2)$$

$$\Delta\psi - \frac{2}{r}\frac{\partial\psi}{\partial r} = r\gamma. \qquad (3)$$

In order to formulate equations close to the surface, it is convenient to choose orthogonal curvilinear coordinates $x^n(x,y,z)$, $x^\tau(x,y,z)$, $x^\theta(x,y,z)$ with local basis vectors normal and tangential to the surface at every point. In order to write down in these curvilinear coordinates the differential forms that enter the hydrodynamic equations, one needs to find explicit expressions for the metric tensor and Christoffel symbols for the chosen class of coordinate systems. Consider both the contravariant coordinates x^n,x^τ,x^θ and the respective physical curvilinear coordinates n,τ,τ_θ. Locally dn is the length along the normal to the surface, $d\tau$ is the surface arclength in the meridian plane, and $d\tau_\theta$ is the surface arclength associated with the rotation angle around the z axis. For an axially symmetric surface, $d\tau_\theta=r(n,\tau)d\theta$. For a differential of radius vector, we have

$$d\mathbf{r} = dx^n\mathbf{e}_n + dx^\tau\mathbf{e}_\tau + dx^\theta\mathbf{e}_\theta = dn\mathbf{i}_n + d\tau\mathbf{i}_\tau + rd\theta\mathbf{i}_\theta, \qquad (4)$$

where $\mathbf{e}_\ell=\mathbf{i}dx/dx^\ell+\mathbf{j}dy/dx^\ell+\mathbf{k}dz/dx^\ell$ are contravariant base vectors. Unlike the contravariant base vectors $\mathbf{e}_n,\mathbf{e}_\tau,\mathbf{e}_\theta$, the Cartesian base vectors $\mathbf{i},\mathbf{j},\mathbf{k}$ and physical curvilinear base vectors $\mathbf{i}_n,\mathbf{i}_\tau,\mathbf{i}_\theta$ are orthonormalized: $\mathbf{i}_n=\mathbf{e}_n/\sqrt{g_{nn}}$, $\mathbf{i}_\tau$

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$=\mathbf{e}_\tau/\sqrt{g_{\tau\tau}}$, $\mathbf{i}_\theta=\mathbf{e}_\theta/\sqrt{g_{\theta\theta}}$. The validity of the following relations is necessary for constructing the contravariant basis:

$$\frac{\partial \mathbf{e}_i}{\partial x^j}=\frac{\partial \mathbf{e}_j}{\partial x^i}. \tag{5}$$

The unit vectors of the physical coordinate system do not satisfy such a requirement, in contrast to contravariant basis vectors, due to the difference in their normalizations. We note that the requirement (5) will be satisfied if the local angle φ between the normal vector to the surface and the symmetry axis depends only on x^τ, does not depend on x^n and x^θ, and

$$\frac{\partial \tau}{\partial x^\tau}=n\frac{d\varphi}{dx^\tau}+f_0(x^\tau), \quad \text{i.e.,} \quad \frac{\partial^2 \tau}{\partial x^\tau \partial n}=\frac{d\varphi}{dx^\tau}. \tag{6}$$

Here the function $f_0(x^\tau)$ is defined by the geometry of the problem and, in particular, by the choice of the origin for the coordinate n. For a spherical surface $x^\tau\equiv\varphi$, $x^n\equiv n=R\equiv\sqrt{x^2+y^2+z^2}$, $x^\tau\equiv\varphi=\tau/n$, $x^\theta\equiv\theta$, and $r=n\sin\varphi$.

Relations (6) permit us to determine the contravariant basis near the surface: $\mathbf{e}_n=\mathbf{i}_n$, $\mathbf{e}_\tau=\mathbf{i}_\tau\partial\tau/\partial x^\tau$, $\mathbf{e}_\theta=r\mathbf{i}_\theta$. One obtains for the basis the following components of the metric tensor: $g_{nn}=1$, $g_{\tau\tau}=(\partial\tau/\partial x^\tau)^2$, $g_{\theta\theta}=r^2$, $g_{\tau n}=g_{\tau\theta}=g_{n\tau}=0$, $g=\det g_{ik}=r^2(\partial\tau/\partial x^\tau)^2$, and the corresponding Christoffel symbols

$$\Gamma^\tau_{n\tau}=\Gamma^\tau_{\tau n}=\frac{\partial\varphi}{\partial x^\tau}\frac{1}{\partial\tau/\partial x^\tau}, \quad \Gamma^\theta_{n\theta}=\Gamma^\theta_{\theta n}=\frac{\sin\varphi}{r},$$

$$\Gamma^n_{\tau\tau}=-\frac{\partial\varphi}{\partial x^\tau}\frac{\partial\tau}{\partial x^\tau}, \quad \Gamma^\tau_{\tau\tau}=\frac{\partial^2\tau}{\partial x^{\tau 2}}\frac{1}{\partial\tau/\partial x^\tau},$$

$$\Gamma^\theta_{\tau\theta}=\Gamma^\theta_{\theta\tau}=\frac{\cos\varphi}{r}\frac{\partial\tau}{\partial x^\tau}, \quad \Gamma^n_{\theta\theta}=-r\sin\varphi, \quad \Gamma^\tau_{\theta\theta}=-\frac{r\cos\varphi}{\partial\tau/\partial x^\tau}. \tag{7}$$

The expressions for the metric tensor and Christoffel symbols allow us to obtain explicit formulas for all differential forms according to general rules of differential geometry [22]. In particular, for an arbitrary vector $\mathbf{F}$ one finds

$$\text{rot }\mathbf{F}=\frac{1}{r}\left(\frac{\partial(rF_\theta)}{\partial\tau}-\frac{\partial F_\tau}{\partial\theta}\right)\mathbf{i}_n+\frac{1}{r}\left(\frac{\partial F_n}{\partial\theta}-\frac{\partial(rF_\theta)}{\partial n}\right)\mathbf{i}_\tau + \left(\frac{\partial F_\tau}{\partial n}-\frac{\partial F_n}{\partial\tau}+\frac{d\varphi}{d\tau}F_\tau\right)\mathbf{i}_\theta. \tag{8}$$

Therefore,

$$\gamma=(\text{rot }\mathbf{v})_\theta=\frac{\partial v_\tau}{\partial n}-\frac{\partial v_n}{\partial\tau}+v_\tau\frac{d\varphi}{d\tau}, \tag{9}$$

$$\Delta\mathbf{v}=-\text{rot}(\gamma\mathbf{i}_\theta)=-\mathbf{i}_n\left(\frac{\partial\gamma}{\partial\tau}+\frac{\cos\varphi}{r}\gamma\right)+\mathbf{i}_\tau\left(\frac{\partial\gamma}{\partial n}+\frac{\sin\varphi}{r}\gamma\right), \tag{10}$$

$$(\mathbf{v}\cdot\nabla)\mathbf{v}=\left[v_n\frac{\partial v_n}{\partial n}+v_\tau\left(\frac{\partial v_n}{\partial\tau}-\frac{d\varphi}{d\tau}v_\tau\right)\right]\mathbf{i}_n + \left[v_n\frac{\partial v_\tau}{\partial n}+v_\tau\left(\frac{\partial v_\tau}{\partial\tau}+v_n\frac{d\varphi}{d\tau}\right)\right]\mathbf{i}_\tau. \tag{11}$$

Thus, the components of Eq. (1) may be rewritten as

$$\frac{\partial p}{\partial\tau}=-\rho\left[\frac{\partial v_\tau}{\partial t}+v_\tau\frac{\partial v_\tau}{\partial\tau}+v_n\left(\frac{\partial v_n}{\partial\tau}+\gamma\right)\right]+\eta\left(\frac{\partial\gamma}{\partial n}+\frac{\sin\varphi}{r}\gamma\right), \tag{12}$$

$$\frac{\partial p}{\partial n}=-\rho\left[\frac{\partial v_n}{\partial t}+v_n\frac{\partial v_n}{\partial n}+v_\tau\left(\frac{\partial v_\tau}{\partial n}-\gamma\right)\right]-\eta\left(\frac{\partial\gamma}{\partial\tau}+\frac{\cos\varphi}{r}\gamma\right). \tag{13}$$

In the more general case when $v_\theta\neq 0$, the terms $\rho v_\theta^2\cos\varphi/r$ and $\rho v_\theta^2\sin\varphi/r$ should be added to the right-hand member of the Eqs. (12) and (13), correspondingly.

The components of the viscous stress tensor $\sigma'_{ik}=\eta(\partial v_i/\partial x_k+\partial v_k/\partial x_i)$, which describe momentum transfer through the boundary, take the form

$$\sigma'_{nn}=2\eta\frac{\partial v_n}{\partial n}, \quad \sigma'_{n\tau}=\eta\left(\frac{\partial v_n}{\partial\tau}+\frac{\partial v_\tau}{\partial n}-v_\tau\frac{d\varphi}{d\tau}\right). \tag{14}$$

The boundary condition at the surface is [21]

$$\left[P-p_v-\sigma\left(\frac{1}{R_1}+\frac{1}{R_2}\right)\right]n_i=\eta\left(\frac{\partial v_i}{\partial x_k}+\frac{\partial v_k}{\partial x_i}\right)n_k-\frac{\partial\sigma}{\partial x_i}. \tag{15}$$

The normal vector is directed here in the outward direction, toward the atmosphere. Here p_v is the pressure of the gas and atmosphere, P is the hydrodynamic pressure on the surface, $R_{1,2}$ are the main local radii of curvature on the surface, and σ is the surface tension. Projections of (15) onto the local tangential and normal directions to the surface are

$$\frac{d\sigma}{d\tau}=\eta\left(\frac{\partial v_n}{\partial\tau}+\frac{\partial v_\tau}{\partial n}-v_\tau\frac{d\varphi}{d\tau}\right), \tag{16}$$

$$P-p_v=\sigma\left(\frac{1}{R_1}+\frac{1}{R_2}\right)+2\eta\frac{\partial v_n}{\partial n}. \tag{17}$$

Equation (16) is the boundary condition for the velocities on the surface. In particular, one gets the boundary condition for the vorticity at the surface:

$$\gamma=\frac{1}{\eta}\frac{d\sigma}{d\tau}+2\left[v_\tau\frac{d\varphi}{d\tau}-\frac{\partial v_n}{\partial\tau}\right]. \tag{18}$$

Boundary conditions for the stream function at the surface may be obtained by integrating the expression $\partial\psi/\partial\tau=-rv_n(\tau)$, where $v_n(\tau)$ is the normal component of the velocity to the boundary. The boundary conditions for the stream function are particularly simple if the motion of the surface is much slower than the typical fluid velocities of the problem, when one can put $v_n\approx 0$.

Equation (17) represents the boundary condition that allows one to obtain the shape of the surface. The pressure

$P(\tau)$ satisfies the Navier-Stokes equations (12) and (13) with corresponding projections of the gravitational force added to the right-hand part of the equations. Therefore, the quantity $p(\tau)=P(\tau)+\rho gz$ satisfies Eqs. (12) and (13) without the additional terms. To further simplify the equations and boundary conditions, one can introduce the quantities $p_3(\tau)-p_3(0)=p(\tau)-p(0)-2\eta\partial v_n/\partial n|_0^\tau$, $p_4(\tau)-p_4(0)=p(\tau)-p(0)+\rho(v_\tau^2+v_n^2)/2|_0^\tau$, and $k=R_1^{-1}+R_2^{-1}=d\varphi/d\tau+\sin\varphi/r$, $k_0=(R_1^{-1}+R_2^{-1})|_{\tau=0}$, $z_0=z|_{\tau=0}$. Then we get

$$p_3(\tau)-p_3(0)=\sigma(k-k_0)+\rho g(z-z_0), \tag{19}$$

$$\frac{d\varphi}{d\tau}=k_0+\frac{p_3(\tau)-p_3(0)-\rho g(z-z_0)}{\sigma}-\frac{\sin\varphi}{r}. \tag{20}$$

In the particular case when one can disregard the term with the pressure, Eq. (20) reduces to the Young-Laplace equation in the form obtained in [23]. The tangential component (12) of the Navier-Stokes equation may be represented as

$$\frac{dp_4}{d\tau}=-\rho\left(\frac{\partial v_\tau}{\partial t}+v_n\gamma\right)+\eta\left[\frac{\partial\gamma}{\partial n}+\frac{\sin\varphi}{r}\gamma\right]. \tag{21}$$

Equation (20) turns out to be quite convenient for determining the shape of the surface. The shape of an axially symmetric surface is unambiguously described by the function $\varphi(\tau)$. Because all expressions contain either the difference $p_4(\tau)-p_4(0)$ or the derivative $dp_4(\tau)/d\tau$, the initial value of $p_4(0)$ is still an arbitrary constant. It is convenient to take

$$p_4(0)=2\eta\frac{\partial v_n}{\partial n}\bigg|_{\tau=0}+\frac{\rho(v_\tau^2+v_n^2)}{2}\bigg|_{\tau=0}. \tag{22}$$

Then

$$p_3(\tau)-p_3(0)=p_4(\tau)-2\eta\frac{\partial v_n}{\partial n}-\frac{\rho(v_\tau^2+v_n^2)}{2}. \tag{23}$$

Introducing the vector $\mathbf{y}=(r(\tau),\varphi(\tau),z(\tau),p_4(\tau))^T$ allows one to represent Eqs. (20), (21), and (23), $dr(\tau)/d\tau=\cos\varphi$, $dz(\tau)/d\tau=-\sin\varphi$, in the form

$$\frac{d\mathbf{y}}{d\tau}=f(\tau,\mathbf{y}). \tag{24}$$

At the initial point one has $\mathbf{y}(0)=(r(0),\varphi(0),z(0),p_4(0))^T$. Here $p_4(0)$ is defined in (22). The Cauchy problem for the system of differential equations (24) with initial conditions derived above can be solved by standard numerical methods to obtain the surface profile.

It is of interest to find out a relative role of terms in Eq. (19) under specific physical conditions. Below we carry out the respective estimations for an evaporating sessile drop lying on a substrate in the regime of a pinned contact line. Evaporation results in an inhomogeneous spatial temperature distribution in the drop and along the drop surface. The corresponding Marangoni forces result in vortex flows of the liquid in the drop.

The motion of the surface is considered to be much slower than typical fluid velocities. This property is valid for a wide class of evaporating drops. Then one can take approximately $v_n\approx 0$. The fluid motion is considered as a quasistationary vortex flow. In the following expressions n_0 is the characteristic distance between the surface of the drop and the vortex center, r_0 is the contact line radius, $\sigma'=-\partial\sigma/\partial T$, ΔT is the temperature difference between the substrate and the apex of the drop, and θ_c is the contact angle. Therefore, $d\varphi/d\tau\approx\sin\theta_c/r_0$ and $d\sigma/(\eta d\tau)\approx-\sigma'\Delta T\sin\theta_c/(\eta r_0\theta_c)$. Using the condition (16) and taking n_0 as a characteristic distance for a change of v_τ along the normal to the surface, one obtains $v_\tau\approx(\partial v_\tau/\partial n)n_0=n_0 d\sigma/(\eta d\tau)+n_0 v_\tau d\varphi/d\tau$—i.e., $v_\tau(1-n_0\sin\theta_c/r_0)\approx-\sigma' n_0\Delta T\sin\theta_c/(\eta r_0\theta_c)$—hence,

$$|v_\tau|\lesssim\frac{\sigma'\Delta T n_0}{\eta r_0}. \tag{25}$$

Therefore $|v_\tau d\varphi/d\tau|\approx|v_\tau|\sin\theta_c/r_0\approx n_0\sigma'\Delta T\sin^2\theta_c/(\eta r_0^2\theta_c)\ll\sigma'\Delta T/(\eta r_0)$, i.e., the term $v_\tau d\varphi/d\tau$ in (16) and (18) is much smaller than $d\sigma/(\eta d\tau)$. It follows from (18) that

$$|\gamma|\approx\frac{\sigma'\Delta T}{(\eta r_0)}. \tag{26}$$

It follows from $|\partial^2 v_\tau/\partial n^2|\approx|v_\tau|/n_0^2$ and

$$\frac{\partial\gamma}{\partial n}=\frac{\partial^2 v_\tau}{\partial n^2}+\frac{d\varphi}{d\tau}\frac{\partial\sigma}{\eta\partial\tau} \tag{27}$$

and (25) that

$$\left|\frac{\partial\gamma}{\partial n}\right|\approx\frac{\sigma'\Delta T}{\eta r_0 n_0}\left(\frac{\sin\theta_c}{\theta_c}+\frac{n_0\sin\theta_c}{2r_0}\right)\approx\frac{\sigma'\Delta T}{\eta r_0 n_0}. \tag{28}$$

We substitute (26) and (28) to (21) and integrate the obtained expression over τ. This gives the estimation of relative effects of pressures and velocities as compared with gravitational forces in Eq. (19):

$$p_4(\tau)-p_4(0)\approx\frac{\sigma'\Delta T\theta_c}{n_0\sin\theta_c},\qquad 2\eta\frac{\partial v_n}{\partial n}\bigg|_0^\tau\ll p_4(\tau)-p_4(0), \tag{29}$$

$$\frac{|p_4(\tau)-p_4(0)|}{\rho gh}\approx\frac{\sigma'\Delta T}{\rho g n_0 h}\frac{\theta_c}{\sin\theta_c}, \tag{30}$$

$$\frac{|\rho v_\tau^2/2|}{\rho gh}\lesssim\frac{1}{2gh}\left(\frac{n_0}{\eta}\frac{\sigma'\Delta T}{r_0}\right)^2.$$

For estimating the term $\rho v_\tau^2/2$, we used (25).

The ratio of gravitational force to the term with surface tension in (19) is characterized by the dimensionless number $B_0=\rho g h r_0/(2\sigma\sin\theta_c)$, which is analogous to the Bond number. Therefore, (30) may be represented as

$$\frac{|p_4(\tau)-p_4(0)|}{|\sigma(k-k_0)|}\approx\frac{\sigma'\Delta T\theta_c}{2\sigma\sin^2\theta_c}\frac{r_0}{n_0}, \tag{31}$$

$$\frac{|\rho v_\tau^2/2|}{|\sigma(k-k_0)|}\approx\frac{\rho}{4r_0\sigma\sin\theta_c}\left(\frac{n_0\sigma'\Delta T}{\eta}\right)^2.$$

Based on a geometry of the fluid surface, we have derived Eqs. (12) and (13) and the boundary condition (19), which allow one to obtain numerically a surface profile dynamics and to take into account the influence of fluid dynamics and gravitational forces on the shape of the fluid-gas interface.

The equations and boundary conditions derived in this paper were used in [24] to find numerically the profile of the evaporating sessile drop surface. According to Eq. (30), the effects of pressure become more important with an increase of the temperature drop in the liquid and with the temperature derivative of the surface tension. The analytical estimations (30) applied to the conditions of [24] show that the relative contribution of pressures and velocities as compared with gravitational forces in the Laplace formula (19) is not too large. Numerical results confirm this qualitative conclusion and give approximately one-tenth for the value of this quantity under the conditions of [24].

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