Spreadings driven by gravity and thermocapillarity

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Abstract

We obtain analytic and approximate solutions for the axisymmetric spreading of a large viscous drop on a smooth horizontal surface driven by gravity and thermocapillarity. The latter forces appear either by uniformly heating or cooling the substrate, or by imposing a quadratically increasing or decreasing temperature profile in the substrate. The flow is described within the lubrication approximation. For uniform cooling and inwards increasing temperature, the spreading is asymptotically described by respective selfsimilar solutions, which are analytically obtained (the drop radius follows power laws with exponents 1/6 and 1/2, respectively). For uniform heating and outwards increasing temperature, the gravity and thermocapillary forces tend to balance, so that respective equilibrium configurations are reached; their shapes and final extensions are obtained. The transition from an initially gravity dominated stage towards the corresponding asymptotic self-similar regime is obtained by using a quasi-self similar approach.

Resumen:

Se obtienen soluciones analíticas aproxi-

madas para el derrame radial de una gota viscosa sobre una superficie horizontal y lisa. flujo es motorizado por las fuerzas gravitatorias y termocapilares. Estas últimas aparecen debido a un calentamiento o enfriamiento uniforme del substrato, o bien cuando se le impone al mismo un perfil de temperatura cuadraticamente creciente o decreciente. El flujo se describe dentro de la aproximación de lubricación. Tanto con enfriamiento uniforme como con temperatura creciente hacia adentro, el derrame está asintoticamente descripto por respectivas soluciones autosimilares, las cuales se obtienen analíticamente. El radio de la gota sigue leyes de potencias con exponentes 1/6 y 1/2, respectivamente. Por otro lado, para calentamiento uniforme o temperatura decreciente hacia afuera, las fuerzas gravitatorias y termocapilares tienden a balancearse, de modo que se alcanzan respectivas configuraciones de equilibrio. Aquí se obtienen sus formas y extensiones finales. La transición de la etapa inicial dominada por la gravedad hacia el correspondiente regimen autosimilar asintótico se obtiene empleando un formalismo cuasi-autosimilar.

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I. Introduction

The problem of the spreading of a viscous liquid drop has been widely investigated theoretically and experimentally in the last years. 1-9 Such interest is mainly related to the occurrence of this problem in several industrial processes, such as coating, soldering and casting. Usually, driving forces such as gravity and Laplace pressure have been considered; however, much less attention has been given to those cases in which differences of temperature between the fluid and the environment give place to thermocapillary effects. As it is well known, thermocapillary forces appear because hot regions of the free surface have smaller values of the surface tension than cold regions. For wetting fluids which spread indefinitely, the thermocapillary forces asymptotically overcome other driving forces such as gravity and Laplace pressure. They may either accelerate the spreading or limit the maximum drop extension depending upon the temperature distribution at the free surface.

An study of an axially symmetric drop spreading on a smooth horizontal surface which is uniformly heated or cooled was done by Ehrhardt & Davis. 10,11 They considered Laplace pressure, gravity and thermocapillary as driving forces and the drop evolution was obtained by assuming static equilibrium, the spreading rate being determined via a contact angle - front velocity relation. Their analysis and experiments were intended for moderate values of both the Bond number Bo (ratio between gravitational and capillary forces) and the Marangoni number Ma (ratio between thermocapillary and capillary forces). An extension of their work was lately done by Smith¹² who studied the spreading of a two-dimensional droplet when a linear temperature distribution is imposed at the substrate. On the other hand, a study of the same problem for large values of Bo and Ma was done by Kalinin & Starov¹³ by using a quasi-steady approach.

This work concerns with the axisymmetric drop spreading driven by gravity and thermocapillarity (Bo, $Ma \gg 1$) when the substrate is uniformly cooled or heated and also when it has a parabolic radially symmetric temperature distribution. This last case may represent a local maximum or minimum of a rather general temperature profile. When the substrate is uniformly heated or the temperature increases outwards, the drop asymptotically reaches an equilibrium shape, which is analytically obtained. Instead, when it is uniformly cooled or the temperature decreases outwards, a transition between a gravity-dominated regime to a thermocapillarydominated regime is obtained. It is well known that the gravitational regime is described by a self-similar solution in which the drop radius follows the power law $r_f \propto t^{1/8}$; likewise, in the thermocapillary regime we find $r_f \propto t^{1/6}$ for the uniform cooling case and $r_f \propto t^{1/2}$ for the parabolic case (in both cases all the results are analytical).

The transition from the initial gravity-dominated stage to the corresponding asymptotic regime is described by a quasi-self-similar approach, ¹⁵ which assumes an instantaneous self-similarity of the flow. The thickness profile during the evolution is analytically obtained, but the time dependence of the drop radius is numerically solved.

II. Lubrication approximation and temperature field

Consider a liquid drop on a smooth, horizontal rigid plane (substrate) located at z=0 in which there is an axisymmetric temperature distribution $T(r,0)=T_s(r)$ (see Fig. 1). The drop is composed of a non-volatile, wetting and very viscous Newtonian liquid surrounded by an ambient gas at zero temperature (i.e., T is measured respect to the ambient gas temperature). The lubrication approximation reduces the Stokes equa-

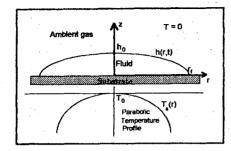


Figure 1: Sketch of a large viscous drop placed on a substrate with a parabolic temperature profile.

tion to:

$$0 = -\frac{\partial p}{\partial r} + \mu \frac{\partial^2 v_r}{\partial z^2},\tag{1}$$

$$0 = -\frac{\partial p}{\partial z} - \rho g, \tag{2}$$

where v_r is the radial component of the velocity, p the pressure, ρ the density and g the gravity (here, we have assumed that μ weakly depends on T).

From Eq. (2) the hydrostatic pressure is

$$p(r,z) = p_0 - \gamma c + \rho g(h-z) \tag{3}$$

where γ is the liquid-ambient gas surface tension, h = h(r,t) the thickness profile and c the curvature given by,

$$c = \frac{\partial^2 h}{\partial r^2} + \frac{1}{r} \frac{\partial h}{\partial r}.$$
 (4)

By replacing Eq. (3) into Eq. (1), and setting the boundary conditions of no-slip at the bottom $v_r(z=0)=0$ and of continuity of the tangential stress at the free surface

$$\mu \frac{\partial v_r}{\partial z} = \frac{\partial \gamma}{\partial r} \text{ at } z = h , \qquad (5)$$

we get a parabolic velocity profile $v_r(z)$. By vertically averaging $(v = h^{-1} \int_0^h v_r(z) dz)$ and neglecting terms with hc << 1, we get¹⁰:

$$v = \frac{\gamma}{3\mu} h^2 \frac{\partial c}{\partial r} - \frac{\rho g}{3\mu} h^2 \frac{\partial h}{\partial r} - \frac{\gamma'}{2\mu} h \left. \frac{\partial T}{\partial r} \right|_{r=h} \tag{6}$$

where $\gamma' = -d\gamma/dT = const. > 0$.

The evolution equation for the drop shape is given by the continuity equation

$$\frac{\partial h}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} (rvh) = 0, \tag{7}$$

which must be solved together with Eq. (6), under the symmetry conditions at the center and the constraint:

$$\int_0^{r_f(t)} 2\pi r h(r,t) dr = V = const.$$
 (8)

where V is the drop volume and $r_f(t)$ the front position.

For sufficiently thin and extended drops, the heat transport equation reduces to $\partial^2 T/\partial z^2 = 0$. Therefore, the temperature distribution T(z) is linear and must satisfy the following boundary conditions at the substrate and the free surface¹⁰:

$$T = T_s(r)$$
 at $z = 0$, $-k\frac{\partial T}{\partial z} = k_g \frac{T}{\delta}$ at $z = h$.

Here, k and k_g are the thermal conductivities of the liquid and the ambient gas, respectively, and δ is the thickness of the thermal boundary layer established within the gas. Then, the temperature profile within the drop is

$$T(r,z) = T_s(r) \left(1 - \frac{z}{\ell + h(r)} \right) \tag{10}$$

where we have defined the length $\ell = k\delta/k_g$; at the free surface we have,

$$T(z=h) = \frac{T_s(r)}{1+h/\ell} \tag{11}$$

and

$$\left. \frac{\partial T}{\partial r} \right|_{z=h} = -\frac{T_s(r)}{\ell \left(1 + h/\ell \right)^2} \frac{\partial h}{\partial r} + \frac{1}{(1 + h/\ell)} \frac{\partial T_s}{\partial r}$$
(12)

Eq. (11) shows that the temperature at the free surface may vary from 0 for $\ell=\delta=0$ (adiabatic limit, with no thermocapillary effect) to $T_s(r)$ for $\ell=\delta=\infty$ (perfectly conducting limit). Usually, silicon oils are employed in the experiments; in Ref. 11 helium was used as the ambient gas $(k_g\simeq k)$ with an estimated value of $\ell\simeq 0.1\,cm$. If air were the ambient gas, by assuming that δ is of the same order, it is expected that $\ell\simeq 1\,cm$ since $k_g\simeq 0.1\,k$. Thus, in what follows we shall assume that the ratio h/ℓ (usually called Biot number) is much smaller than unity.

Here, we shall consider that the temperature distribution at the substrate is given by

$$T_s(r) = T_0 \left(1 + \frac{r^2}{L^2} \right),$$
 (13)

where L is a length and T_0 is a constant temperature. For $L \to \infty$, the uniformly heated $(T_0 > 0)$ or cooled $(T_0 < 0)$ case is obtained. For finite L, Eq. (13) is an axially symmetric parabolic temperature distribution which may represent a local maximum $(T_0 > 0)$ or minimum $(T_0 < 0)$ of a rather general temperature profile. For instance, the temperature field of a circular substrate whose border is kept at constant temperature is described by Eq. (13) near the center. Thus, Eq. (12) takes the form

$$\left. \frac{\partial T}{\partial r} \right|_{r=h} = -\frac{T_0}{\ell} \left(1 + \frac{r^2}{L^2} \right) \frac{\partial h}{\partial r} + 2T_0 \frac{r}{L^2} \tag{14}$$

In the following, we shall be concerned with spreadings involving drop volumes V of some cubic centimeters; therefore, only gravitational and thermocapillary forces will be considered since the driving force due to the gradient of the Laplace pressure is negligible in most of the fluid volume. The study of this regime strongly simplifies the problem because the contact line does not

require any special treatment. The spreadings driven by gravity and surface tension in which the drop volumes are of some cubic millimeters have been longly studied in the literature.^{8,9,15} In the present case, the second and third terms of Eq. (6) dominate over the first one, so that the validity conditions of our approach are that both gravity and thermocapillary forces largely overcome capillary forces. By considering that $\partial h/\partial r \approx h_0/r_f$ and $\partial c/\partial r \approx h_0/r_f^3$, where $h_0 \approx V/r_f^2$ is the thickness at the centre of the drop, Eq. (6) leads to the conditions:

$$r_f^2 \gg \frac{\gamma}{\rho g} , \frac{r_f^4}{V\ell} + \frac{r_f^8}{V^2 L^2} \gg \frac{\gamma}{\gamma' |T_0|}.$$
 (15)

Eq. (15) shows that for unlimited spreadings $(r_f \to \infty)$, the Laplace pressure can always be asymptotically neglected. Therefore, Eq. (6) reduces to

$$v = -\frac{\rho g}{3\mu} h^2 \frac{\partial h}{\partial r} - \frac{\gamma'}{2\mu} h \left. \frac{\partial T}{\partial r} \right|_{r=h} \tag{16}$$

with the temperature gradient given by Eq. (14).

III. Asymptotic solutions

In the early stages of the spreading, the first term in Eq. (16) dominates whatever is the temperature distribution. Then; the flow is described by the self-similar viscous-gravity solution known as Barenblatt-Pattle's solution¹⁴:

$$h = h_0(t) \left(1 - r^2/r_f^2\right)^{1/3}, \quad (17)$$

$$r_f(t) = 0.894... \left(\frac{\rho g V^3}{3\mu} t\right)^{1/8}.$$
 (18)

where $h_0(t) = 4V/3\pi r_f^2$. As the spreading develops, the thermocapillary effects become increasingly important.

A. Substrate uniformly heated or cooled

In this case Eqs. (16) and (14) lead to $(L \rightarrow \infty)$:

$$v = \left(-\frac{\rho g}{3\mu}h + \frac{\gamma' T_0}{2\mu\ell}\right)h\frac{\partial h}{\partial r}.$$
 (19)

If the substrate is colder than the ambient gas $(T_0 < 0)$, the thermocapillary force is directed outwards like the gravity force, so that both promote the spreading. Thus, while no other forces appear, the drop spreads indefinitely. As r_f increases, the second term in Eq. (19) becomes dominant, so that is given by

$$v = \frac{\gamma' T_0}{2\mu\ell} h \frac{\partial h}{\partial r} \tag{20}$$

for

$$r_f \gg \left(\frac{\rho g \ell V}{\gamma |T_0|}\right)^{1/2}.$$
 (21)

We shall look for an asymptotic self-similar solution of Eq. (20) in the form,

$$h(r,t) = h_0(t)H(\eta) , \ v(r,t) = v_f(t)U(\eta)$$
 (22)

where $v_f = dr_f/dt$ is the front velocity, and $H(\eta)$, $U(\eta)$ are nondimensional functions of $\eta = r/r_f$. The function $U(\eta)$ may be obtained from the continuity equation; in fact, by replacing these expressions into Eq. (7), we get

$$\omega H - \eta H' + \eta^{-1} (\eta U H)' = 0$$
 (23)

where the prime denotes derivative with respect to η , and

$$\omega = \frac{r_f}{v_f h_0} \frac{dh_0}{dt} = const. \tag{24}$$

In these variables, the constant volume condition, Eq. (8), becomes:

$$V = Ih_0 r_f^2 = const. (25)$$

where we have defined the shape factor

$$I = \int_0^1 2\pi \eta H(\eta) \ d\eta. \tag{26}$$

Since I and V are constants, Eq. (25) gives $\omega = -2$; then, Eq. (23) admits the analytical solution $U = \eta$, that is the velocity profile is linear. It should be noted that the self-similar solution of Eq. (17) may be obtained with this formalism when gravity is the only driving force.

By replacing Eq. (22) into Eq. (20) we have,

$$\frac{H}{\eta}H' = \frac{2\mu\ell r_f v_f}{\gamma' T_0 h_0^2} = const. \tag{27}$$

With the condition H(1) = 0, we obtain the self-similar solution:

$$H = (1 - \eta^2)^{1/2} , \qquad (28)$$

$$r_f(t) = \left[-\frac{27}{4\pi^2} \frac{\gamma' T_0 V^2}{\mu \ell} t \right]^{1/6},$$
 (29)

$$I = 2\pi/3. \tag{30}$$

If the substrate is warmer than the ambient gas $(T_0 > 0)$, the thermocapillary force is directed inwards unlike the gravity force, so that they compete till balancing in an asymptotic configuration with the front at rest. The equilibrium drop shape given by the condition v = 0 leads to a flat profile of constant thickness.

B. Substrate with a parabolic temperature profile

Now, the direction of the thermocapillary forces depends on whether the drop is placed on a minimum $(T_0 > 0)$ or a maximum $(T_0 < 0)$ of $T_s(r)$, so that the drop may reach a final equilibrium shape or spread indefinitely. In this case,

CEILAP CITEFA - CONICET ZUFRIATEGUI Y VARELA 1603 VILLA MARTELLI REPUBLICA ARGENTINA the first term in Eq. (14) may be neglected for $L \ll r_f \sqrt{\ell/h_0}$, a condition that is satisfied since we have already assumed $h_0/\ell \ll 1$. The governing equation is

$$v = -\frac{\rho g}{3\mu}h^2 \frac{\partial h}{\partial r} - \frac{\gamma T_0}{\mu L^2} hr.$$
 (31)

For $T_0 < 0$ (outwards decreasing temperature) and

$$r_f \gg \left(\frac{\rho g V^2 L^2}{\gamma T_0}\right)^{1/6},$$
 (32)

substitution of Eq. (22) into Eq. (47) (without the gravity term) leads to

$$H = \frac{\mu L^2 v_f}{\gamma' T_0 h_0 r_f} = 1, \tag{33}$$

so that the asymptotic self-similar solution is given by a constant thickness profile $(I = \pi)$ whose front advances as

$$r_f(t) = \left[-\frac{2\gamma' T_0 V}{\pi \mu L^2} t \right]^{1/2}.$$
 (34)

This means that the wetted area grows linearly with time, what represents a very high spreading rate when compared, for instance, with the gravity driven spreading.

For $T_0 > 0$ (outwards increasing temperature), the equilibrium shape given by v=0 leads

$$h = h_0 \left(1 - r^2/r_{f,eq}^2\right)^{1/2}$$
, (35)

$$r_{f,eq} = \left(\frac{3\rho g L^2}{4\pi^2 \gamma' T_0}\right)^{1/6} V^{1/3},$$
 (36)

$$I = 2\pi/3. \tag{37}$$

IV. Gravity thermocapillary transition

In principle, no single self-similar solution for fixed values of β , M, D and N given by r_f ,

though it is not self-similar for long time, it instead approximates an instantaneous self similar behaviour. This approximation has been successfully used 15 previously to deal with spreadings without thermocapillarity. Here, we shall suppose that Eq. (22) approximately holds by considering that H is actually a slowly varying function of time, so that t only determines its coefficients. Substitution of Eq. (22) into Eq. (7), we get Eq. (23) where ω is now given by

$$\omega = -2 + \frac{r_f}{v_f} \left(\frac{\partial H}{\partial t} - \frac{H}{I} \frac{dI}{dt} \right); \qquad (38)$$

therefore, the quasi self-similar evolution will be valid when the last term is much less than 2. We expect this is a good approximation provided the departures from the self-similar behaviour are small. Under this condition (which is fulfilled a posteriori), we get $U = \eta$ as before, and

$$\beta \eta = -H^2 H' + M \left(1 + D^2 \eta^2 \right) H H' - N \eta H.$$
 (39)

Here, we have introduced the dimensionless num-

and

$$\beta = \frac{3\mu}{\rho g} \frac{r_f}{h_0^3} v_f. \tag{41}$$

Here, β is the ratio between the viscous and gravity forces; note that M, N are two different ratii between the thermocapillary and gravity forces. Eq. (39) must be solved under the boundary conditions

$$H = 1, H' = 0 \text{ at } \eta = 0$$
 (42)
 $H = 0 \text{ at } \eta = 1$

can describe the complete flow evolution. How- h_0 and v_f at time t. The key idea of the approxever, we shall look for a solution which, even imation is to suppose that during a small time interval from t to $t + \Delta t$, the solution is given by the function $H(\eta)$ which results from Eq. (39). Once obtained the value of β that allows $H(\eta)$ to satisfy Eq. (42), the front velocity is given by

$$v_f = \frac{\rho g V^3}{3\mu} \frac{\beta(t)}{I^3 r_f^7}.$$
 (43)

Then, the updated values of x_f and h_0 give place to another solution of Eq. (39) and so on.

A. Substrate uniformly cooled (M < 0)

A direct integration of Eq. (39) with $L \to \infty$ gives,

$$\frac{\beta\eta^2}{2} = -\frac{H^3 - 1}{3} + M\frac{H^2 - 1}{2} \tag{44}$$

whence

$$\beta = \frac{2}{3} - M. \tag{45}$$

Here, it is convenient to calculate the shape factor as $I = \pi \int_0^1 \eta^2 dH$; thus, we get

$$I = \frac{\pi}{2} \left(\frac{3 - 4M}{2 - 3M} \right). \tag{46}$$

Eqs. (45) and (46) allow to perform the time integration of Eq. (43) to obtain the spreading dynamics. In Fig. 2 we show $r_f(t)$ as obtained from the complete time integration for a set of typical values. It can be seen the transition from the gravity dominated regime, Eq. (17), to the thermocapillary regime, Eq. (28). The limit $\beta \to -M$ for $|M| \gg 1$ (see Eq. (45)) indicates a balance between the viscous and thermocapillary forces, so that gravity plays no role in this asymptotic (thermocapillary) regime.

B. Substrate with a parabolic temperature profile

In this case, the equation of motion is (see Eq.(39))

$$\beta \eta = -H^2 H' - N \eta H. \tag{47}$$

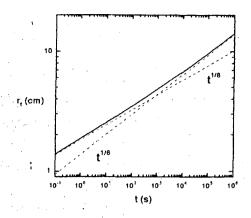


Figure 2: Front position r_f as a function of time for a drop placed on a uniform cooled substrate with $T_0 = -20^{\circ}C$ and $\ell = 0.1 \, \mathrm{cm}$. The drop parameters are: $V = 1 \, \mathrm{cm}^3$, $\mu = 1 \, \mathrm{g/cm} \, \mathrm{s}$, $\rho = 0.9 \, \mathrm{g/cm}^3$ and $\gamma = 0.05 \, \mathrm{dyn/cm}^{\circ}C$. The dashed lines correspond to the asymptotic solutions given by Eqs. (17) and (28)

A direct integration leads to

$$N\eta^{2} = \left(1 - H^{2}\right) - 2\beta^{*} \left(1 - H\right) - 2\beta^{*2} \ln\left(\frac{H + \beta^{*}}{1 + \beta^{*}}\right) \tag{48}$$

where

$$\beta^* = \frac{\beta}{N} = \frac{\mu}{\gamma' T_0} \frac{L^2}{h_0 r_f} v_f \tag{49}$$

is the relation between viscous and thermocapillary forces. By putting $H(1)=0,\ \beta^*$ is given by the root of the equation:

$$\frac{1-N}{2} - \beta^* - \beta^{*2} \ln \left(\frac{\beta^*}{1+\beta^*} \right) = 0.$$
 (50)

The shape factor I may also be calculated analytically as:

$$I = \frac{\pi}{N} \left[\frac{2}{3} - \beta^* + 2\beta^{*2} - 2\beta^{*3} \ln \left(\frac{1 + \beta^*}{\beta^*} \right) \right]. \tag{51}$$

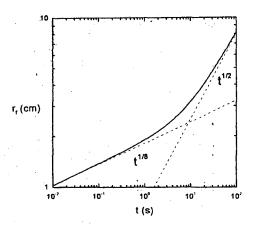


Figure 3: Front position r_f and as a function of time for a drop placed on a uniform cooled substrate with $T_0 = -20^{\circ}C$ and $\ell = 0.1 \, \text{cm}$. The drop parameters are: $V = 1 \, \text{cm}^3$, $\mu = 1 \, g/\text{cm} \, s$, $\rho = 0.9 \, g/\text{cm}^3$ and $\gamma' = 0.05 \, dyn/\text{cm}^{\circ}C$. The dashed lines correspond to the asymptotic solutions given by Eqs. (17) and (28).

The complete calculation of β^* as a function of N allows to perform the time integration. The results for $r_f(t)$ (see Fig. 3) show the transition from the gravity regime to the thermocapillary regime. For $|N| \gg 1$, Eq. (50) leads to

$$\beta^* \approx -1 - \exp(N/2 - 3/2);$$
 (52)

this shows that the asymptotic self-similar thermocapillary solution is exponentially reached. Besides, Eq. (51) gives $I \to \pi$ as $|N| \to \infty$, i.e. the thickness profile becomes planar in this regime. The asymptotic balance between thermocapillary forces and viscous forces is evidenced by $\beta^* \to -1$ as $|N| \to \infty$; in fact, $\beta^* = -1$ leads to the asymptotic power law $r_f(t)$ given by Eq. (34).

V. Final remarks

We have described the spreading dynamics of a large viscous liquid drop placed on a heated or cooled substrate under the action of gravity and thermocapillary forces. Uniform and parabolic radially increasing or decreasing temperature profiles on the substrate have been considered. It has been shown that if the temperature distribution gives place to thermocapillary forces pointing outwards, the spreading asymptotically reaches a thermocapillary dominated self-similar regime. Otherwise, the drop tends to a configuration at rest with a steady flow inside; thus, thermocapillarity balances gravity by means of the viscous forces within the drop.

Solutions for the asymptotic regimes have been analytically obtained. A quasi-self similar approach has been developed to describe the transition from the initial viscous-gravity regime to the asymptotic self-similar regimes in which the front advances following power laws of time. We believe that the asymptotic thermocapillary regime achieved with the parabolic temperature profile may be of interest in the coating industry because its uniform thickness profile and its high spreading rate (see Eq. (34)) are desirable features for this process.

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