

## Thermal effects on film development during spin coating

B. S. Dandapat<sup>a)</sup> and B. Santra

*Physics and Applied Mathematics Unit, Indian Statistical Institute, 203, Barrackpore Trunk Road, Calcutta 700 108, India*

A. Kitamura

*Japan Nuclear Cycle Development Institute, 4-33, Tokai, Ibaraki 319-1194, Japan*

(Received 13 July 2004; accepted 12 April 2005; published online 26 May 2005)

This study is an extended version of a previous analysis of thermal effects on the free-surface liquid film flow on a flat, heated rotating disk [A. Kitamura, "Thermal effects of liquid film flow during spin coating," *Phys. Fluids* **13**, 2788 (2001)]. The assumption of a constant disk temperature is extended to a nonuniform disk temperature, and the restriction on small radial distance used in the previous analysis is removed. The evolution equation for the transient film thickness is obtained and solved by the method of characteristics [B. S. Dandapat, P. Daripa, and P. C. Ray, "Asymptotic study of film thinning process on a spinning annular disk," *J. Appl. Phys.* **94**, 4144 (2003)]. The effect of both thermocapillary forces and variable viscosity on the flow is revealed. A physical explanation is provided to justify the results.

[DOI: 10.1063/1.1927525]

### I. INTRODUCTION

Spin coating is a well-known technique by which one can produce a very thin and uniform film of viscous liquid by the action of centrifugal force over a spinning disk. This technique is widely used in the microelectronics industry to manufacture microelectronic devices, optical mirrors, magnetic disks for data storage, etc. The spin-coating mechanism is investigated theoretically in two different ways, viz., (a) spreading of a liquid drop on the surface of a dry rotating disk and (b) development of a thin film on the surface of a wet rotating disk from a liquid blob. In the former type of investigation<sup>1-5</sup> the main interest was to know how the contact line moves during the spreading of the drop on the disk and its stability. In the latter type, starting from the pioneering work of Emslie *et al.*,<sup>6</sup> several models<sup>7-16</sup> have been proposed in subsequent investigations to know the film development on the spinning disk and the literature on the subject is considerably developed. The assumption of a wet wall is justified in the light of the experimental observation on advancing contact lines by Schwartz and Tejada<sup>17</sup> and Giradella and Radigan,<sup>18</sup> in which they have indicated the presence of an unseen precursor layer of fluid ahead of the contact line which is only an angstrom thick. Further it has been argued by physical chemists that the presence of a precursor layer is a very real phenomenon arising as a consequence of evaporation from the drop in a small region local to the contact line followed by diffusion and adsorption (De Gennes<sup>19</sup>).

It is interesting to note that in all the above studies<sup>7-16</sup> it is tacitly assumed that the disk is wet so that the classical no-slip boundary condition can be applied at every point on the disk surface and the film flows under a planar interface

for the entire period of spinning. Further, it has been observed from these studies<sup>6-16</sup> that the rate of film thinning slows down beyond a specific height (depending on the rotational speed) of the film. In general, the final stage of film thickness is proportional to  $t^{-1/2}$  for  $t \rightarrow \infty$ , where  $t$  is the spinning time. So to obtain the desired thinness of the film, one has to operate the spinner for quite a long time. As a result a solid skin may form on the surface layer of the film due to evaporation. Therefore, coating defects may occur if any convective flow that is present does not completely cease before this skin hardens sufficiently. Dandapat and Ray<sup>20</sup> (DR) have tried to accelerate the rate of film thinning so that one may obtain the desired thinness before hardening of the skin. According to them it is always possible to create an external shear stress on the film surface in the form of a surface tension gradient by imposing either a temperature or a concentration gradient along the radial direction of the disk. They have shown that by imposing a specified axisymmetric temperature distribution on the disk it is possible to obtain the film thickness proportional to  $(\alpha t)^{-1}$  for  $t \rightarrow \infty$ , where  $\alpha$  is a new nondimensional thermocapillary parameter. Middleman,<sup>13</sup> and Rehg and Higgins<sup>14</sup> have also noticed that the rate of film thinning increases due to the shear induced by air flow over the film surface. It is to be mentioned here that DR have restricted their analysis to a specified family of temperature distributions that ultimately helps them to search for similarity solutions. In the above studies also, the tacit assumption was that the film remains planar for the entire process.

Recently, Kitamura<sup>21</sup> has removed the barrier of a planar film surface assumption and obtained the transient film thickness through matched asymptotic analysis. Kitamura<sup>22</sup> has also studied the effects of surface tension and viscosity variation with temperature on film planarization and thinning. In this study Kitamura considered a hemispherical-shaped liquid blob placed on the center of the disk, which is heated

<sup>a)</sup> Author to whom correspondence should be addressed. Electronic mail: dandapat@isical.ac.in

uniformly. He derived the evolution equation for the film thickness and solved it by using an expansion for film thickness in powers of  $r^2$ ,  $r$  being the radial distance from the axis of rotation. He observed that the variable viscosity has a more profound effect than that of thermocapillarity on the transient film thickness. A close scrutiny reveals that one may not observe the effective thermocapillarity flow at the free-surface due to uniform heating of the disk. Further the solution obtained by Kitamura is valid only near the axis of rotation. Recently Dandapat *et al.*<sup>23</sup> have removed the restriction of small  $r$  used by Kitamura<sup>21,22</sup> to solve the evolution equation by using the method of characteristics.

The present analysis falls in the second type of spin-coating mechanism, which implies the inherent assumption of a wet disk to facilitate the use of the no-slip boundary condition. Further to study the effect of both thermocapillary flow and variable viscosity, a nonuniform heating of the disk surface is assumed. Due to the proximity of the present investigation with the previous paper (Kitamura<sup>22</sup>) we shall state details only where the two papers differ. The paper is organized as follows. The evolution equation for the film thickness is derived in Sec. II using an asymptotic expansion of all dependent variables in terms of the aspect ratio  $\varepsilon(=h_0/L)$ , where  $h_0$  and  $L$ , respectively, denote the characteristic length scales in the vertical (film thickness) and radial directions. The evolution equation is solved in Sec. III by the method of characteristics to remove the constraint of small  $r$ . Results and a discussion are contained in Sec. IV.

## II. EVOLUTION EQUATION

Consider a uniform film of viscous, incompressible, non-volatile liquid on a disk whose radius is large in comparison with the thickness of the film. Initially, the system is at the room temperature  $T_0$ . Simultaneously the system starts rotating with a uniform angular velocity  $\Omega$  about an axis normal to the plane of the disk and an axially symmetric temperature  $T_d(r)$  is imposed on the disk. The origin is fixed at the center of the disk and the  $z$  axis points vertically upward along the axis of rotation. To express the governing equations of motion, continuity, energy, and the corresponding boundary and initial conditions into their nondimensional form, we shall use the characteristic time scale  $t_c(=\nu_0/\Omega^2 h_0^2)$ , the time at which viscous and centrifugal forces balance each other. Specifically, the dimensionless (asterisked) variables are defined as

$$h = h_0 h^*, \quad z = h_0 z^*, \quad r = L r^*, \quad t = t_c t^*, \quad \gamma = h_0 \gamma^*,$$

$$u = (\Omega^2 L h_0^2 / \nu_0) u^*,$$

$$v = (\Omega^3 L h_0^4 / \nu_0^2) v^*, \quad w = (\Omega^2 h_0^3 / \nu_0) w^*,$$

$$p = \rho \Omega^2 L^2 p^*, \quad T = T_0 + (T_{d_0} - T_0) T^*,$$

where  $h_0$  is the maximum height of the initial film thickness  $h(r,0) = \gamma(r)$ ,  $\nu_0$  is the kinematic viscosity of the fluid at room temperature  $T_0$ ,  $\rho$  is the fluid density, and  $T_{d_0}$  is the imposed temperature at the center of the disk.  $u, v, w, p$ , and  $T$ , respectively, denote the three velocity components along

the  $r, \theta, z$  directions, pressure, and temperature. Finally we obtain the dimensionless governing equations and the corresponding boundary and initial conditions as

$$\begin{aligned} \varepsilon \text{Re}(u_r + uu_r + wu_z) = & -p_r + r + 2\varepsilon \text{Re} v + \varepsilon^2 \text{Re}^2 r^{-1} v^2 \\ & + 2\varepsilon^2 [(fu_r)_r + f(u/r)_r] \\ & + [f(u_z + \varepsilon^2 w_r)]_z, \end{aligned} \quad (1)$$

$$\begin{aligned} \varepsilon \text{Re} \left( v_r + uv_r + wv_z + \frac{uv}{r} \right) = & \varepsilon^2 r^{-2} [fr^3(v/r)_r] \\ & + (fv_z)_z - 2u, \end{aligned} \quad (2)$$

$$\begin{aligned} \varepsilon^3 \text{Re}(w_r + uw_r + ww_z) = & -p_z - \varepsilon \text{Fr} + 2\varepsilon^2 (fw_z)_z \\ & + \varepsilon^2 r^{-1} [fr(u_z + \varepsilon^2 w_r)]_r, \end{aligned} \quad (3)$$

$$\frac{1}{r}(ru)_r + w_z = 0, \quad (4)$$

$$\varepsilon \text{Pe}(T_r + uT_r + wT_z) = \varepsilon^2 r^{-1} (rT_r)_r + T_{zz}, \quad (5)$$

at  $z=0$ ,

$$u = v = w = 0, \quad T = \Theta(r), \quad (6)$$

at  $z=h(r,t)$ ,

$$\begin{aligned} -p + 2f\varepsilon^2(1 + \varepsilon^2 h_r^2)^{-1}(\varepsilon^2 u_r h_r^2 + w_z - u_z h_r - \varepsilon^2 w_r h_r) \\ = \varepsilon r^{-1}(rh_{rr} + h_r + \varepsilon^2 h_r^3)(1 + \varepsilon^2 h_r^2)^{-3/2}(\text{We} - \varepsilon^2 \alpha T), \end{aligned} \quad (7)$$

$$\begin{aligned} f[2\varepsilon^2 h_r(w_z - u_r) + (u_z + \varepsilon^2 w_r)(1 - \varepsilon^2 h_r^2)] \\ = \varepsilon \alpha (T_r + h_r T_z)(1 + \varepsilon^2 h_r^2)^{1/2}, \end{aligned} \quad (8)$$

$$v_z - \varepsilon^2 r h_r (v/r)_r = 0, \quad (9)$$

$$T_z - \varepsilon^2 h_r T_r = -\text{Bi}(1 + \varepsilon^2 h_r^2)^{1/2} T, \quad (10)$$

$$h_t + uh_r = w, \quad (11)$$

and the initial conditions at time  $t=0$ ,

$$u = v = w = 0, \quad T = 0, \quad h(r,0) = \gamma(r), \quad h_t = 0, \quad (12)$$

where the dimensionless parameters that appear are the Reynolds number  $\text{Re} = \Omega^2 L h_0^3 / \nu_0^2$ , the modified Froude number  $\text{Fr} = g / \Omega^2 L$ , the Peclet number  $\text{Pe} = \Omega^2 L h_0^3 / \nu_0 \kappa$ , the Weber number  $\text{We} = \sigma_0 / \rho \Omega^2 L^3$ , and the Biot number  $\text{Bi} (= \beta h_0 / \lambda)$ .  $\beta, \lambda, \kappa$ , and  $\sigma$  denote the heat transfer coefficient, thermal conductivity, thermal diffusivity, and surface tension, respectively.  $\alpha = -(d\sigma/dT)(T_{d_0} - T_0) / (\rho \Omega^2 h_0^2 L)$  is the thermocapillary parameter. Since the aim of this paper is to study the effects of temperature variation along the radial direction of the disk, it is assumed that a small temperature difference  $T_{d_0} - T_0$  is sufficient to change the surface tension  $\sigma$  and viscosity  $\nu$  of the liquid. To derive the above set of equations, linear variations of  $\sigma$  and  $\nu$  with temperature are assumed, of the form  $\sigma = \sigma_0(1 - \alpha T)$  and  $\nu = \nu_0(1 + \varepsilon \delta_1 T)$ , where  $f = \nu / \nu_0$ . It is to be noted here that a few typographical errors in Kitamura<sup>22</sup> are removed in this study; also the notations  $\delta$

and  $\Delta$  of Kitamura<sup>22</sup> are changed to  $\varepsilon\delta_1$  and  $\alpha$ , respectively. In the present analysis  $\delta_1$ ,  $\alpha$ , Re, Fr, and Pe are of order 1 but  $We \sim O(\varepsilon^{-2})$  and  $\varepsilon \ll 1$ .

To obtain an asymptotic solution, all the dependent variables are expanded in powers of  $\varepsilon$  according to the following ansatz:

$$F(r, t, z) \sim F_0(r, t, z) + \varepsilon F_1(r, t, z) + O(\varepsilon^2).$$

On substituting the above form for each dependent variable in the system of Eqs. (1)–(10), and equating terms of like orders, new sets of equations are obtained as usual. Solving these sets from zeroth order to the first order, one gets

$$u = \frac{1}{2}(2h-z)rz + \varepsilon \left[ -\frac{\delta_1 r \Theta(r)}{6} \{-2Az^3 + 3(1+Ah)z^2 - 6hz\} - \alpha \left( \frac{\Theta(r)}{1+Bi h} \right)_r z + Fr \left( \frac{h_r z^2}{2} - h h_r z \right) + Re \left( \frac{r z^6}{360} - \frac{r h z^5}{60} + \frac{r^2 h h_r z^4}{24} + B z^3 - C z \right) + We D_r \left( h z - \frac{z^2}{2} \right) \right], \quad (13)$$

where  $A = Bi/(1+Bi h)$ ,  $B = 2rh^3/9 + rh_r/6$ ,  $C = 3rh^5/5 + r^2 h_r h^4/6 + rh_r h^2/2$ , and  $D = (rh_r)_r/r$  which are same as in Kitamura<sup>22</sup> except of his notation  $\delta$  representing the change of viscosity due to temperature that is replaced by  $\varepsilon\delta_1$  in the present analysis and a negative sign on the first  $O(\varepsilon)$  term inside the angular bracket. It is to be noted here that the solution for  $u$  in Eq. (13) will not satisfy the initial condition (12) due to the large characteristic time scale considered here. To get the initial effect, one needs to consider the short-time analysis as described by Higgins.<sup>12</sup> The surface evolution equation is obtained by integrating the continuity equation (4) and using (11), as

$$\frac{\partial h}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} \int_0^h (ru) dz = 0. \quad (14)$$

Finally, Eqs. (13) and (14) together yield

$$h_t = -\frac{1}{3r}(r^2 h^3)_r + \frac{\varepsilon}{12r} \left\{ -\delta_1 \left( \frac{r^2 \Theta(r)(4h^3 + 3Bi h^4)}{1+Bi h} \right) + 6\alpha r h^2 \left( \frac{\Theta(r)}{1+Bi h} \right)_r + Re \left( \frac{136}{105} r^2 h^7 - \frac{8}{5} r^3 h^6 h_r \right) + 4Fr r h^3 h_r - 4We r h^3 \left( \frac{1}{r} (r h_r)_r \right) \right\}. \quad (15)$$

It is to be noted here that for  $\Theta(r)=1$ , one can obtain Kitamura's<sup>22</sup> equation (11) from Eq. (15) above, except that of the negative sign in front of the first of the  $O(\varepsilon)$  terms on the right-hand side (rhs) of (15) and  $\Delta$  is replaced by  $\alpha$ . For  $\Theta(r)=0$ , Eq. (15) coincides with Eq. (35) of Reisfeld *et al.*<sup>24</sup> with  $E=0$ . Further it is to be noted that although the circumferential velocity component  $v$  exists, it is not necessary for deriving Eq. (14) and the flow pattern is independent of the circumferential coordinate because of axial symmetry assumption.

### III. SOLUTION PROCESS

From now onwards, the method of solution procedure differs from that of Kitamura<sup>22</sup> and follows closely Dandapat *et al.*<sup>23</sup> Expanding  $h(r, t)$  in powers of  $\varepsilon$  as

$$h(r, t) \sim h_0(r, t) + \varepsilon h_1(r, t) + O(\varepsilon^2) \quad (16)$$

and using Eq. (16) in Eq. (15) and equating the coefficients of order up to  $\varepsilon$ , one gets

$$h_{0t} + r h_0^2 h_{0r} = -\frac{2}{3} h_0^3, \quad (17)$$

and

$$h_{1t} + r h_0^2 h_{1r} = -2(h_0^2 + r h_0^2 h_{0r} h_1) + \frac{1}{12r} \left[ -\delta_1 \left\{ \frac{r^2 \Theta(r)(4h_0^3 + 3Bi h_0^4)}{1+Bi h_0} \right\} + 6\alpha r h_0^2 \left( \frac{\Theta(r)}{1+Bi h_0} \right)_r + Re \left( \frac{136}{105} r^2 h_0^7 - \frac{8}{5} r^3 h_0^6 h_{0r} \right) + 4Fr r h_0^3 h_{0r} - 4We r h_0^3 \left( \frac{1}{r} (r h_{0r})_r \right) \right]. \quad (18)$$

From Eq. (17), one can obtain

$$\frac{d}{dt} h_0[r(t), t] = -\frac{2}{3} h_0^3[r(t), t] \quad (19)$$

along the characteristic curve  $r(t)$  satisfying

$$\frac{d}{dt} r(t) = r(t) h_0^2[r(t), t]. \quad (20)$$

Upon integration, Eqs. (19) and (20) give

$$h_0[r(t), t] = C_0 \chi^{-1/2}, \quad (21a)$$

along with

$$r(t) = C_1 \chi^{3/4}, \quad (21b)$$

where  $\chi = 1 + (4/3)C_0^2 t$ . It also follows from Eq. (21) that, along each characteristic curve,

$$r(t) h_0^{3/2}[r(t), t] = C_1 C_0^{3/2} = \text{const.} \quad (22)$$

Integrating Eq. (18) along the same characteristic curve, Eq. (20) reduces to

$$h_1[r(t), t] = -\frac{C_0}{12} \delta_1 \chi^{-1/2} \Theta(r) (3 + \zeta^{-1}) + \frac{\alpha}{2} C_1^{-1} \chi^{-3/4} \left( \Theta_r(r) \zeta^{-1} + \frac{2}{3} Bi C_0 C_1^{-1} \chi^{-5/4} \Theta(r) \zeta^{-2} \right) + \frac{62}{315} Re C_0^5 \chi^{-5/2} - \frac{2}{9} Fr C_0^2 C_1^2 \chi^{-5/2} + \frac{32}{81} We C_0^2 C_1^{-4} \chi^{-4} + C_2 \chi^{-1/2}, \quad (23)$$

where  $\zeta = (1 + Bi C_0 \chi^{-1/2})$  and  $C_0$ ,  $C_1$ , and  $C_2$  are constants of

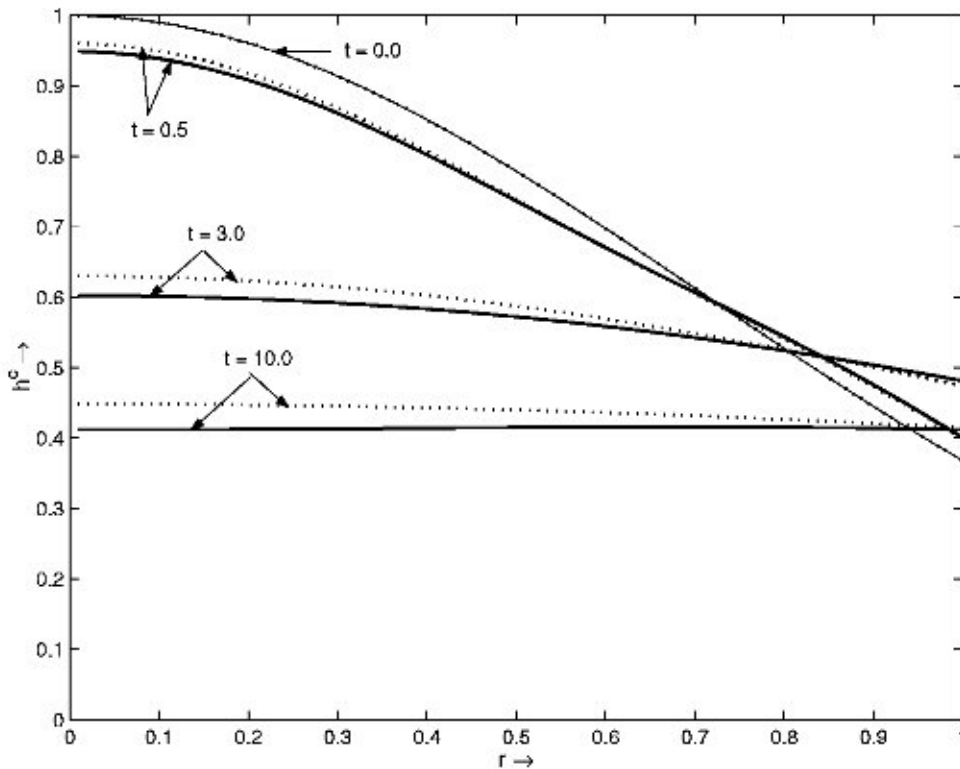


FIG. 1. Composite height  $h^c$  vs  $r$  at  $t = 0.0, 0.5, 3.0, 10.0$  for  $Bi=0.0$  and  $\gamma(r)=\exp(-r^2)$ . Solid lines for temperature distribution— $\Theta(r)=\exp(-r^2)$  and dotted lines for constant temperature distribution— $\Theta(r)=1$ .

integration, which are to be evaluated through matching with the corresponding small time solution. The short-time scale analysis can be done by simply stretching the temporal coordinates as  $\tau=t/\varepsilon$  and keeping other variables the same as before except  $h$  is replaced by  $H$  to mark short-time scale variable. Following as outlined by Higgins,<sup>12</sup> one can obtain

$$h_S = \gamma(r) - \varepsilon \left[ \left( \frac{2}{3} H_0^3 + Re H_0^2 H_{0r} \right) \tau + Re \left\{ (4H_0^5 + 10rH_0^4 H_{0r}) \sum \frac{\exp(-\lambda_p^2 \tau / Re H_0^2) - 1}{\lambda_p^6} \right\} + 4H_0^2 H_{0r} \sum \frac{\tau \exp(-\lambda_p^2 \tau / Re H_0^2)}{\lambda_p^4} \right]. \quad (24)$$

Here  $h_S$  denotes the surface profile obtained from short-time analysis. The matching condition that is derived from the requirement that the flow is continuous from the start of spinning of the disk to all succeeding time, suggests

$$\lim_{\tau \rightarrow \infty} h_S(\tau) = \lim_{t \rightarrow 0} h(t),$$

which implies  $C_0 = \gamma(\xi)$ ,  $C_1 = r(t=0) = \xi$  (say) and

$$C_2 = \frac{\delta_1}{12} \gamma \Theta(\xi) (3 + \xi_0^{-1}) - \frac{\alpha}{2} \xi^{-1} \left( \Theta \xi_0^{-1} + \frac{2}{3} \Theta(\xi) Bi \gamma \xi^{-1} \xi_0^{-2} \right) - \frac{62}{315} Re \gamma^5 + \frac{2}{9} Fr \gamma^2 \xi^{-2} - \frac{32}{81} We \gamma^2 \xi^{-4} + 0.066666 Re(4\gamma^5 + 10\xi \gamma^4 \gamma_\xi), \quad (25)$$

where  $\xi_0 = 1 + Bi C_0$ .

Using (25) in Eqs. (21) and (23), the composite film thickness is obtained as

$$h^c = h(r, t) + h_S(r, \tau) + \frac{2}{3} \gamma^3 \tau - [\gamma + 0.06666 \varepsilon Re(4\gamma^5 + 10\xi \gamma^4 \gamma_\xi)]. \quad (26)$$

#### IV. RESULT AND DISCUSSION

The variation of the film thickness  $h$  with  $r$  given by Eq. (26), for different temperature distributions and times, is depicted in Fig. 1. It is clear from this figure that the film thins faster for temperature decreasing radially outwards than for a constant distribution. In this study, only two thermal factors, the thermocapillary parameter  $\alpha$  and the viscosity variation with temperature  $\delta_1$  act on the system. It is natural to ask the question which of these parameters affects the flow most? A close look into the solution for  $h$  reveals that both of these parameters appear in the first-order term  $h_1$ . Collecting all the terms containing  $\delta_1$  and  $\alpha$  separately and denoting them by  $h_{1\delta_1}$  and  $h_{1\alpha}$ , respectively, one can obtain

$$h_{1\delta_1} = \frac{\gamma \delta_1}{12 \chi^{1/2}} \{ 3[\Theta(\xi) - \Theta(r)] + [\Theta(\xi) \xi_0^{-1} - \Theta(r) \xi^{-1}] \}, \quad (27)$$

and

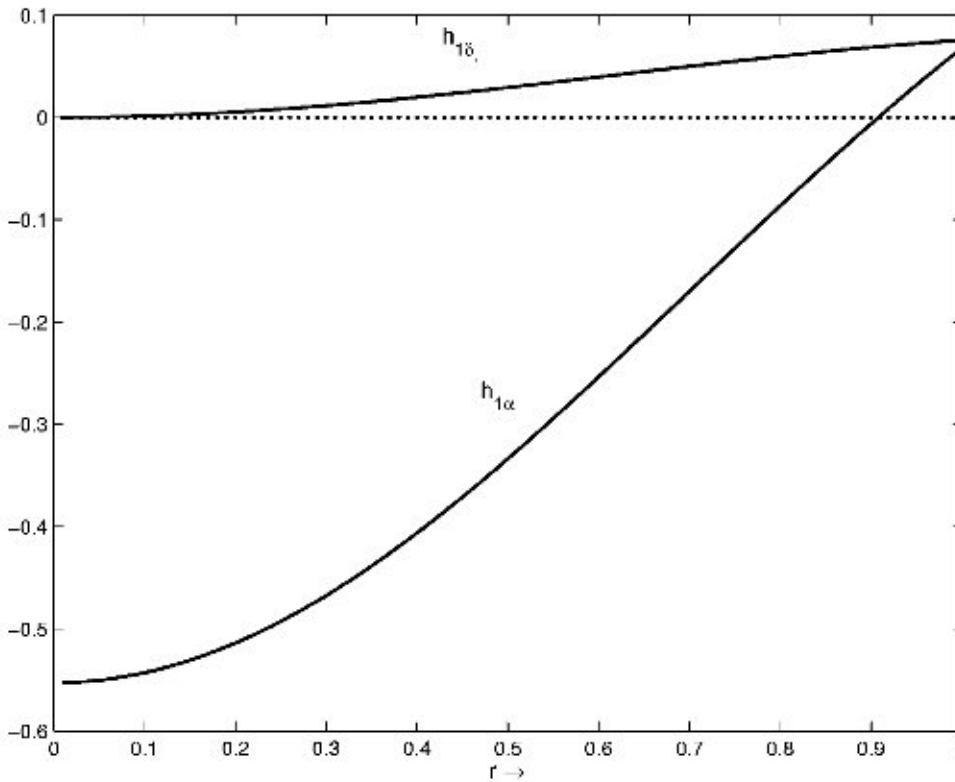


FIG. 2.  $h_{1\delta_1}$  and  $h_{1\alpha}$  vs  $r$  at  $t=3.0$  for  $\text{Bi}=0.0$  and  $\gamma(r)=\exp(-r^2)$ . Solid lines for temperature distribution— $\Theta(r)=\exp(-r^2)$  and dotted line represents both  $h_{1\delta_1}$  and  $h_{1\alpha}$  for constant temperature distribution— $\Theta(r)=1$ .

$$h_{1\alpha} = \frac{\alpha}{2\xi\chi^{1/2}} \left[ \frac{\Theta_r(r)\chi^{1/4}}{\chi^{1/2} + \text{Bi}\gamma} - \frac{\Theta_\xi}{1 + \text{Bi}\xi} + \frac{2}{3}\text{Bi}\gamma\xi^{-1} \left\{ \frac{\Theta(r)}{\chi^{1/2}(\chi^{1/2} + \text{Bi}\gamma)^2} - \frac{\Theta(\xi)}{(1 + \text{Bi}\gamma)^2} \right\} \right], \quad (28)$$

where  $\Theta(\xi)$  is the initial temperature distribution. The influence of  $\delta_1$  and  $\alpha$  on film thinning is delineated in Fig. 2, and it is clear that both  $h_{1\delta_1}$  and  $h_{1\alpha}$  increase with  $r$  for  $\text{Bi}=0$  at time  $t=3$ . From this figure one can infer that  $h_{1\alpha}$  has a stronger influence on film thinning than  $h_{1\delta_1}$ . This behavior of  $h_{1\alpha}$  and  $h_{1\delta_1}$  may be explained as follows: The thermocapillary parameter  $\alpha$  is a measure of the variation of surface tension with temperature. Since the disk is allowed to cool radially outward, the surface tension is low at the center of the disk and hence the thermocapillary force acts as a tangential stress on the surface of the film along the favorable flow direction. This leads  $\alpha$  to enhance film thinning. However, the role of  $\alpha$  will be adverse if the disk is heated radially outward. On the other hand,  $\delta_1$  is the measure of the increase of the viscous resistance as the system is allowed to cool radially outward. Due to this reason,  $h_{1\delta_1}$  is increasing with  $r$  in Fig. 2. However, if the temperature increases radially outward, then the viscous resistance decreases and as a result  $\delta_1$  helps thinning the film. Now looking at the expressions for  $h_{1\delta_1}$  and  $h_{1\alpha}$  in Eqs. (27) and (28), one can see that for a constant temperature distribution and for  $\text{Bi}=0$ , both  $h_{1\delta_1}$  and  $h_{1\alpha}$  vanish, as reflected by the dotted line in Fig. 2. Further one can also see that as time increases  $h_{1\delta_1}$  decreases through the increase of  $\chi$  but remains positive for cooling radially outward.

Figure 3 shows the variation of the composite film height with respect to time at  $r=0.8$ , when the disk is either cooled or heated outwards for different  $\text{Bi}$ . It is clear from the figure that cooling outward helps thinning for all  $\text{Bi}$  except for  $\text{Bi}=0$ . At  $\text{Bi}=1.0$ , the film thins faster for cooling. According to Fig. 3, it seems that for  $\text{Bi}=0$  the film thins faster for heating. For  $\text{Bi}=0$ ,  $h_1$  reduces to

$$h_1 = \frac{\alpha}{2}\xi^{-1}\chi^{-3/4}(\Theta_r - \Theta_\xi\chi^{1/4}) + \text{terms containing (Re, We, and Fr)}. \quad (29)$$

The first term on the rhs of Eq. (29) gives the change of film height due to the difference of temperature distribution. Since  $|\Theta_r| \ll |\Theta_\xi|$  either for heating or cooling outwards, the term in the bracket becomes negative or positive depending on the temperature distribution. This explains why heating outward gives faster thinning for  $\text{Bi}=0$ .

Looking first at Fig. 3, one may wonder what is the reason for  $h^c$  increasing initially, before decreasing? One may resolve this question by scrutinizing Fig. 1 and will find that at  $r=0.8$ ,  $h^c$  has increased at time  $t=0.5$  from its original height at  $t=0.0$ . Due to the assumption of the no-slip boundary condition and a constant volume of liquid, a sharp wave front gradually develops at the initial stage and moves along the radial direction, resulting in the increase of  $h^c$  for some  $r$ . This result is also observed in the classical work of Emslie *et al.*<sup>6</sup> in their Fig. 3.

Finally, it is to be pointed out here that the present paper is the first to investigate the thermal effects on spin coating with disk temperature variation and predicts the transient

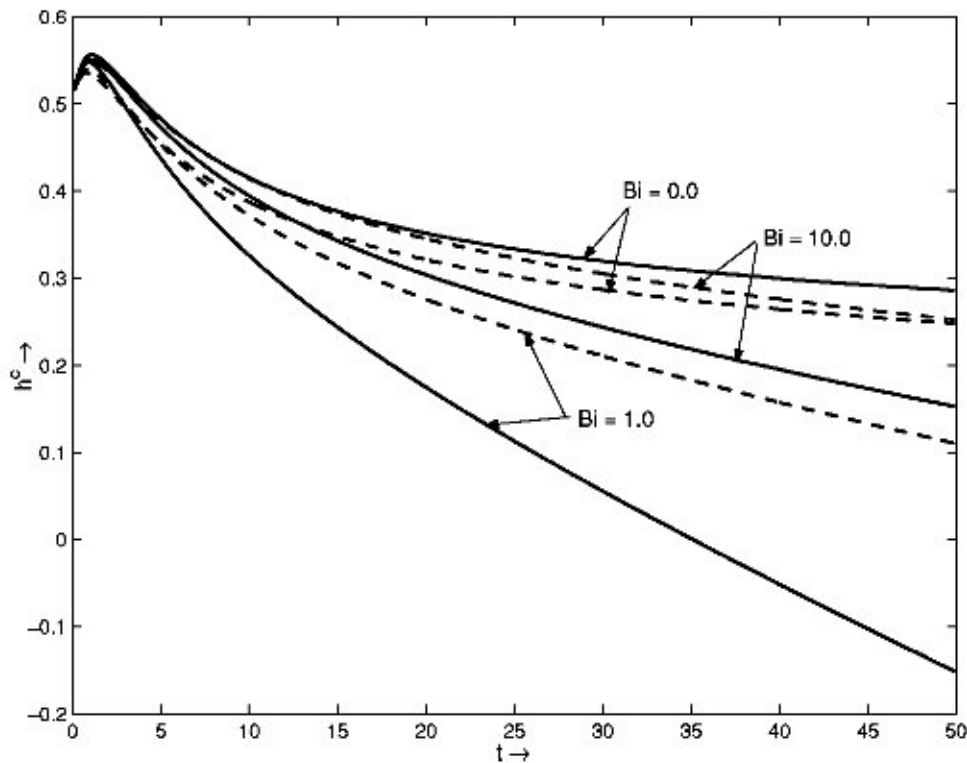


FIG. 3. Composite height  $h^*$  vs  $t$  at  $r = 0.8$  for  $Bi=0.0, 1.0, 10.0$ , and  $\gamma(r) = \exp(-r^2)$ . Solid lines for temperature distribution— $\Theta(r)=\exp(-r^2)$  and dashed lines for temperature distribution— $\Theta(r)=\exp[-(r-1)^2]$ .

shape of the interfacial film boundary. Unfortunately, the authors do not know any experimental result which can be compared with the present observations.

## ACKNOWLEDGMENT

The authors express their sincere thanks to the referees for their constructive suggestions and comments to improve the text of this paper.

- <sup>1</sup>S. M. Troian, E. Herbolzheimer, S. A. Safran, and J. F. Joanny, "Fingering instabilities of driven spreading films," *Europhys. Lett.* **10**, 25 (1989).
- <sup>2</sup>F. Melo, J. F. Joanny, and S. Fauve, "Fingering instability of spinning drops," *Phys. Rev. Lett.* **63**, 1958 (1989).
- <sup>3</sup>J. A. Moriarty, L. W. Schwarz, and E. O. Tuck, "Unsteady spreading of thin liquid films with small surface tension," *Phys. Fluids A* **3**, 733 (1991).
- <sup>4</sup>I. S. McKinley, S. K. Wilson, and B. R. Duffy, "Spin coating and air-jet blowing of thin viscous drops," *Phys. Fluids* **11**, 30 (1999).
- <sup>5</sup>L. W. Schwartz and R. V. Roy, "Theoretical and numerical results for spin coating of viscous liquids," *Phys. Fluids* **16**, 569 (2004).
- <sup>6</sup>A. G. Emslie, F. J. Bonner, and L. G. Peck, "Flow of a viscous liquid on a rotating disk," *J. Appl. Phys.* **29**, 858 (1958).
- <sup>7</sup>A. Acrivos, M. J. Shah, and E. E. Petersen, "On the flow of a non-Newtonian liquid on a rotating disk," *J. Appl. Phys.* **31**, 963 (1960).
- <sup>8</sup>D. Meyerhofer, "Characteristics of resist films produced by spinning," *J. Appl. Phys.* **49**, 3993 (1960).
- <sup>9</sup>B. T. Chen, "Investigation of the solvent-evaporation effect on spin coating of thin films," *Polym. Eng. Sci.* **23**, 399 (1983).
- <sup>10</sup>W. W. Flack, D. S. Soong, A. T. Bell, and D. W. Hess, "A mathematical

- model for spin coating of polymer resists," *J. Appl. Phys.* **56**, 1199 (1984).
- <sup>11</sup>P. C. Sukanek, "Spin coating," *J. Imaging Technol.* **11**, 184 (1985).
- <sup>12</sup>B. G. Higgins, "Film flow on a rotating disk," *Phys. Fluids* **29**, 3522 (1986).
- <sup>13</sup>S. Middleman, "The effect of induced air flow on the spin coating of viscous liquids," *J. Appl. Phys.* **62**, 2530 (1987).
- <sup>14</sup>T. J. Rehg and B. G. Higgins, "The effect of inertia and interfacial shear on film flow on a rotating disk," *Phys. Fluids* **31**, 1360 (1988).
- <sup>15</sup>C. J. Lawrence, "Spin coating with slow evaporation," *Phys. Fluids A* **2**, 453 (1990).
- <sup>16</sup>B. S. Dandapat and P. C. Ray, "Film cooling on a rotating disk," *Int. J. Non-Linear Mech.* **25**, 369 (1990).
- <sup>17</sup>A. M. Schwartz and S. B. Tejada, "Studies of dynamic angles on solids," *J. Colloid Interface Sci.* **38**, 359 (1972).
- <sup>18</sup>H. Giradella and W. Radigan, "Electrical resistivity changes in spreading liquid films," *J. Colloid Interface Sci.* **51**, 522 (1975).
- <sup>19</sup>P. G. De Gennes, "Wetting: Statics and dynamics," *Rev. Mod. Phys.* **57**, 827 (1985).
- <sup>20</sup>B. S. Dandapat and P. C. Ray, "The effect of thermocapillarity on the flow of a thin liquid film on a rotating disc," *J. Phys. D* **27**, 2041 (1994).
- <sup>21</sup>A. Kitamura, "Asymptotic solution for film flow on a rotating disk," *Phys. Fluids* **12**, 2141 (2000).
- <sup>22</sup>A. Kitamura, "Thermal effects of liquid film flow during spin coating," *Phys. Fluids* **13**, 2788 (2001).
- <sup>23</sup>B. S. Dandapat, P. Daripa, and P. C. Ray, "Asymptotic study of film thinning process on a spinning annular disk," *J. Appl. Phys.* **94**, 4144 (2003).
- <sup>24</sup>B. Reisfeld, S. G. Bankoff, and S. H. Davis, "The dynamics and stability of thin liquid films during spin coating I. Films with constant rates of evaporation or absorption," *J. Appl. Phys.* **70**, 5258 (1991).