



A free boundary problem for evaporating layers

B.W. van de Fliert

University of Twente, 7500 AE Enschede, The Netherlands

Abstract

Subject is the free boundary problem of a liquid layer that is dried by evaporation. Using a Stefan type problem, we model the diffusion driven drying of a layer of liquid paint consisting of resin and solvent. For the one-dimensional case, the movement of the free boundary is found in a short time asymptotic analysis. When including fluid flow and the levelling of the surface in a two-dimensional model, two small parameter cases are discussed. The first concerns the levelling by surface tension under the assumption of a small aspect ratio, where the thin film equation appears in the free boundary condition. Secondly the effect of a small perturbation of the flat free boundary shows different decay for long and short wavelength surface elevations.

Key words: free boundary, liquid coating, evaporation, Stefan problem

1 Introduction

A model is presented for the drying of a layer of liquid paint, consisting of a non-volatile resin and a volatile solvent. The drying process is assumed to be through diffusion and evaporation of the solvent, for which the model results in a Stefan type problem, so a free boundary problem similar to models for phase transitions. One part of this study is aimed to reduce the system of equations in a lubrication approximation, which will result in a diffusion problem for the solvent volume fraction, coupled to the thin film equation at the free boundary.

In Howison et al. [15] a mathematical model is derived for the drying of a paint layer using classical lubrication theory, including the effects of variable surface tension, viscosity, solvent diffusivity and solvent evaporation rate. A careful balance of the different parameters in the model allows the authors to

Email address: fliert@math.utwente.nl (B.W. van de Fliert).

derive a thin film model for the drying paint, also including evaporation of the volatile solvent. However, as is remarked by Howison et al., when the diffusion process is slow, say, at a later stage in the drying process or for a thicker film, the leading order distribution of the solvent across the layer is not uniform and their scaling arguments fail. It is this stage that we try to model here.

The motivation for including the flow of the solvent, is the intention to extend the study of the thin film equation in some way to viscoelastic materials. With a direct use of the constitutive equations corresponding to viscoelastic behaviour, the lubrication approximation does not supply a significant simplification. In the polymeric industry one adopts a model for so-called stress-driven diffusion in order to get a better understanding of the role of the viscoelasticity of the material. In such a model the stress build-up is related to changes in the composition of the material due to species diffusion [3,6,8]. Van de Fliert and Van der Hout [11] have included the stress-driven diffusion in a model for the drying of a layer of paint. This leads to a Stefan type model with a kinetic condition [12,13]. This model is presented in section 2 in higher space dimensions, however, still without the complicating effects of non-Newtonian flow. The aim of the present work is to show the stability properties of the Newtonian model with the free evaporation boundary. When starting from the viewpoint of the evaporation models, there is also a need to include fluid flow effects, in particular surface tension effects, which have not been taken into account in [11]. The need for surface tension in the Stefan type model is expected from the analogy of phase transitions in the supercooled Stefan problem. The Gibbs Thomson law or a kinetic law is needed to regularize the boundary condition [4,16–18,10,5]. It is not clear what is the equivalent of the Gibbs Thomson law for the model for species diffusion used for the drying paint. From an analogy of electrodynamic problems, the curvature is used in a flux condition for the model of drying liquid drops, used to explain the formation of coffee stains [7]. This means that in this coffee case, not the surface tension effects but the evaporation dependent on the curvature of the drop, supply the boundary conditions. We observe in the asymptotic analysis in section 4 that the effect of surface tension appears as a fourth order term.

We first present the model equations and then continue in three separate sections that can be read independently. In section 3 the actual analysis of the movement of the free boundary is reduced to the one-dimensional case, where surface tension effects play no role. The movement of the boundary is entirely determined by the evaporation and asymptotic techniques can be applied to find the time behaviour. A short time analysis is made using a Taylor expansion from the fundamental source solution. In section 4 a lubrication scaling is applied to reduce the model for small aspect ratio. In section 5 a stability analysis is done when small elevations appear on the paint-air interface, which shows a different decay behaviour for short and long wavelength perturbations.

2 Derivation of the model equations

Let $\phi(\mathbf{x}, t)$ denote the solvent volume fraction, $0 \leq \phi \leq 1$, for which the solvent flux consists of a convective term and a diffusive term, $\mathbf{q}_\phi = \mathbf{q}\phi - D\nabla\phi$,

$$\phi_t + \mathbf{q} \cdot \nabla\phi = \nabla \cdot D\nabla\phi. \quad (1)$$

The paint domain $\Omega(t)$ may have a fixed boundary Γ_0 , at a substrate, where there will be no flux both of solvent and the total fluid,

$$D\nabla\phi \cdot \mathbf{n}_0 = 0, \quad \mathbf{q} \cdot \mathbf{n}_0 = 0. \quad (2)$$

The kinematic conditions at free surface $\Gamma(t)$, with evaporating solvent, are

$$\mathbf{q} \cdot \mathbf{n} = V_n + E(\phi), \quad (3)$$

$$\phi \mathbf{q} \cdot \mathbf{n} - D(\phi)\nabla\phi \cdot \mathbf{n} = \phi V_n + E(\phi), \quad (4)$$

where V_n is the speed of the free surface in the direction of the outward normal \mathbf{n} . We can rewrite (4), using (3), as $D\nabla\phi \cdot \mathbf{n} = -(1 - \phi)E(\phi)$.

Initially, the solvent concentration is $\phi(\mathbf{x}, 0) = \phi_0(\mathbf{x})$, $\mathbf{x} \in \overline{\Omega(0)}$, and we need to specify the initial domain $\Omega(0)$ with boundaries Γ_0 and $\Gamma(0)$. Finally, we need a functional relation for the diffusion coefficient $D(\phi)$ and the evaporation $E(\phi)$. In this paper we take the simple case of constant diffusion coefficient and rescale so that $D = 1$. Since E should satisfy $E(0) = 0$, $E(\phi) > 0$ for $0 < \phi \leq 1$ and it is likely that E is non-decreasing in ϕ , we will assume that, with evaporation constant K ,

$$E(\phi) = K\phi. \quad (5)$$

Assuming that the configuration is a thin layer of liquid paint, with on one side an impermeable substrate and with the upper surface free and moving due to the evaporation of solvent into air, we simply write $y - h(x, t) = 0$ at $\Gamma(t)$, so that $V_n = -h_t / \sqrt{1 + h_x^2}$, while $y = 0$ at the substrate Γ_0 . Then without the convection ($\mathbf{q} = 0$), the problem given by (1)-(5) is recognized as a generalized one-phase Stefan problem for ϕ and h . We first investigate the movement of the free boundary for this one-dimensional, non-convective model, then we include the velocity \mathbf{q} from a Stokes flow, to derive a lubrication model and perform a stability analysis.

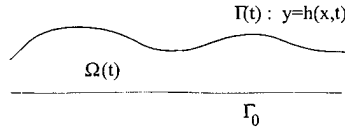


Fig. 1. Sketch of the two-dimensional paint domain

3 The one-dimensional model

When reduced to one dimension and without convection, the model for an evaporating layer is given by the following one-dimensional, one-phase generalized Stefan problem for volume fraction $\phi(y, t)$ and layer thickness $h(t)$.

$$\left\{ \begin{array}{ll} \phi_t = \phi_{yy}, & 0 < y < h(t), t > 0, \\ \phi_y = 0, & y = 0, t > 0, \\ \phi_y = -K\phi(1 - \phi), & \text{for } y = h(t), t > 0, \\ h_t = -K\phi, & y = h(t), t > 0, \\ h = h_0 > 0, & t = 0, \\ \phi = \phi_0(y) \geq 0, & 0 \leq y \leq h_0, t = 0. \end{array} \right. \quad (6)$$

We note that the boundary conditions in (6) do not only generalize the Stefan problem with kinetic undercooling, but also with a Stefan condition that does not conserve energy in the classical sense. The formal limit of this model in the case of fast evaporation, i.e. in the limit $K \rightarrow \infty$, is given by the Stefan problem with supercooling, with free boundary conditions $\phi = 0$ and $\phi_y = h_t$. In [12] it is shown that it is the proper limiting model for fast evaporation. Using that

$$h_1 = \int_0^{h(t)} 1 - \phi(y, t) dy = \int_0^{h_0} 1 - \phi_0(y) dy \quad (7)$$

which means that the volume of the non-evaporating material in the drying layer is conserved (and denoted by h_1), it is not difficult to show that $\phi \rightarrow 0$ and $h \rightarrow h_1$ as $t \rightarrow \infty$, given some conditions for ϕ_0 [12].

3.1 Small time behaviour of the boundary

The small time behaviour of the free boundary will be obtained here from a formal asymptotic analysis, based on a Taylor expansion and an embedding as

in [14]. An embedding result for the classical Stefan problem and more general melting problems was given by Boley [2,1]. We will not show the validity of the analysis, but only give a comparison with some numerical calculations, as shown in figures 3.1 and 3.1 below. We will find that for a solution pair (ϕ, h) of (6), for small time the free boundary $h(t)$ behaves as

$$h(t) = h_0 - K\phi_0(h_0)t + \frac{4K}{3\sqrt{\pi}} [K\phi_0(h_0) \{1 - \phi_0(h_0)\} + \phi_0'(h_0)] t^{3/2} + \mathcal{O}(K^2t)^2. \tag{8}$$

We first write the solution ϕ as if it were defined on the whole real axis instead of only for $0 \leq y \leq h(t)$, with the use of the fundamental solution

$$\begin{aligned} \mathcal{K}(y, t; \xi, \tau) &= \frac{1}{2\sqrt{\pi}(t - \tau)^{1/2}} \exp -\frac{(y - \xi)^2}{4(t - \tau)} \quad \text{for } \tau \leq t, \\ \phi(y, t) &= \int_{-\infty}^0 \mathcal{K}_0 f_1(\xi) d\xi + \int_0^{h_0} \mathcal{K}_0 f_2(\xi) d\xi + \int_{h_0}^{\infty} \mathcal{K}_0 f_3(\xi) d\xi. \end{aligned} \tag{9}$$

with $\mathcal{K}_0 = \mathcal{K}(y, t; \xi, 0)$. Here $f_2 = \phi_0$ is the initial condition of the original problem, but the functions f_1 and f_3 are unknown and need to be determined from the boundary conditions at $y = 0$ and $y = h(t)$. We are interested in the behaviour for small t and expect a similarity-like behaviour, so we define new variables

$$\nu = 2\sqrt{t}, \quad p = \frac{y - \xi}{\nu}.$$

This choice can be motivated by the fact that we want to use a Taylor expansion in ν and by using the new variable $\nu \sim t^\alpha$ it is observed that $\alpha = 1/2$ provides integer powers in ν , with equal powers for h_t and ϕ_y . If we write $\Phi(y, \nu) = \phi(y, t)$, $H(\nu) = h(t)$, then at $y = H(\nu)$:

$$\begin{aligned} \Phi(H(\nu), \nu) &= \frac{1}{\sqrt{\pi}} \left\{ \int_{(H(\nu))/\nu}^{\infty} f_1(H(\nu) - \nu p) e^{-p^2} dp + \right. \\ &\quad \left. \int_{(H(\nu)-h_0)/\nu}^{H(\nu)/\nu} f_2(H(\nu) - \nu p) e^{-p^2} dp + \int_{-\infty}^{(H(\nu)-h_0)/\nu} f_3(H(\nu) - \nu p) e^{-p^2} dp \right\}, \end{aligned}$$

By differentiating (9) with respect to y we get a similar expression for $\Phi_y(y, t)$. A Taylor expansion in ν is now used to match the two conditions at $y = H(\nu)$. If we write $H(\nu) = h_0 - A_1\nu - A_2\nu^2 + \mathcal{O}(\nu^3)$, then due to the free boundary

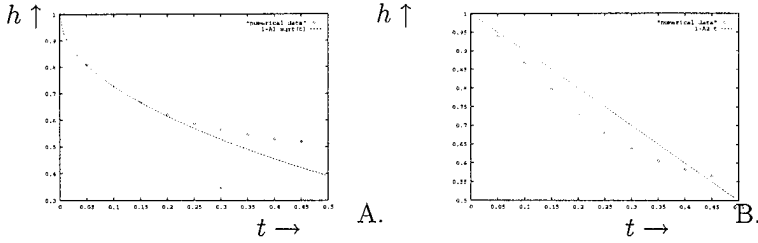


Fig. 2. The thickness $s(t)$ as a function of time for the Stefan problem; A. initial data $\phi_0 = \frac{1}{2}$, $h_0 = 1$ and B. $\phi_0(y) = 1 - y$, $h_0 = 1$.

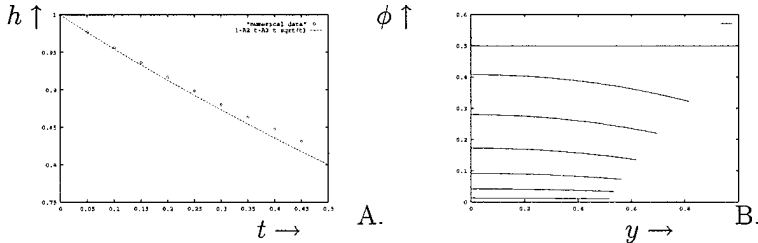


Fig. 3. A. The thickness $h(t)$ as a function of time for problem (6), with the first two terms in the approximation. B. The volume fraction $\phi(y, t)$ as a function of y for $y \in [0, h(t)]$ at the values $t = 0.5m$, $m = 1..10$. Initial data $\phi_0 = \frac{1}{2}$, $h(0) = 1$, $K = 1$. The shorter lines at larger times indicate the retracting boundary.

condition $h_t = -K\phi$, the powers of ν can only be matched if $A_1 = 0$. Then from the condition $\phi_y = -K\phi(1 - \phi)$ we find $f_3(h_0) = f_2(h_0) = \phi_0(h_0)$. The Taylor expansion for Φ thus reduces to

$$\Phi(H(\nu), \nu) = \phi_0(h_0) + \frac{\nu}{2\sqrt{\pi}} \{f'_3(h_0) - f'_2(h_0)\} + \mathcal{O}(\nu^2).$$

The condition $\Phi_y = -K\Phi(1 - \Phi)$ at $y = H(\nu)$ then yields, with $f'_2(h_0) = \phi'_0(h_0)$, that $f'_3(h_0) = -2K\phi_0(h_0) \{1 - \phi_0(h_0)\} - \phi'_0(h_0)$, and we conclude that

$$A_1 = 0, \quad A_2 = -\frac{K}{4}\phi_0(h_0), \quad A_3 = -\frac{K}{6\sqrt{\pi}} [K\phi_0(h_0) \{1 - \phi_0(h_0)\} + \phi'_0(h_0)].$$

This gives the behaviour in (8).

Completely similarly, for the solution of the supercooled Stefan problem, being the limiting problem for $K \rightarrow \infty$, we find the expected short-time behaviour

$$h(t) = h_0 - 2A\sqrt{t} + \mathcal{O}(t) \tag{10}$$

where A is a constant depending on $\phi_0(h_0)$ only.

4 A thin film approximation

In Howison et.al. [15] where the thin film equation is derived including evaporation, it is argued that the validity of the model depends on the scales of diffusion and convection. When the lateral diffusion of solvent as measured by the dimensionless diffusion coefficient is very small, the leading order distribution of solvent across the layer is not uniform and as a consequence the variables will depend on y - as well as x and t . It is exactly this parameter regime where $D \sim \varepsilon^2$ (with ε the aspect ratio of the thin film) that will be used below. We deal here with the simplest case of constant viscosity μ and surface tension τ . Naturally, since the composition of the paint changes during the drying process, the physical coefficients are likely to vary with the volume fraction of the solvent. One may expect it to be a simple extension of the model by taking the coefficients as functions of the volume fraction ϕ only. However, in order to be able to make a reduction in the equations by a lubrication scaling, it is required that $\mu = \mu(x, t)$ only at leading order (so independent of y , the scaled coordinate). During the drying process, this is not to be expected, unless the volatile and non-volatile fluids have the same viscosities. If the assumption is not made, however, the reduction of equations is ended and one has to concentrate on solving the full system numerically. The assumption $\mu(x, t)$ is a large restriction on the validity of the model, and in fact we might as well assume that the viscosity is constant. It is explicitly not a restriction for the model in [15] because of the uniform volume fraction over the thin layer in their scalings. We will also neglect gravity and external effects. As is argued in [15], gravity and surface tension gradients are important in the practical situation of a drying paint, with the Bond and Marangoni number of size comparable to other non-dimensional numbers included in this model. The model is quite easily extended to include these and other effects.

4.1 Lubrication scalings

We denote by E_0 a typical initial evaporation value, L and U are typical length and velocity scales and we define the aspect ratio by $\varepsilon = h_0/L$. See [15] for a discussion on the different choices for U . We are interested in the situation where the effects of surface tension and diffusion are comparable, and we choose $U = D/\varepsilon^2 L$, which will scale the diffusion coefficient to 1. The evaporation coefficient is non-dimensionalised as $E = E_0 e(\phi)/\varepsilon U$ and the inverse capillary number is $C = \varepsilon^3 \tau/\mu U$.

For a Newtonian, viscous and incompressible fluid in a two-dimensional layer where $0 < x < 1$ and $0 < y < h(x, t)$, we have for the leading order velocities

$\mathbf{q} = (u, v)$ and pressure p , the equations of classical lubrication theory:

$$u_x + v_y = 0, \quad p_x = u_{yy}, \quad p_y = 0,$$

with boundary conditions at $y = 0$ $u = 0, v = 0$ and at $y = h(x, t)$

$$ph_x + u_y = -Ch_{xx}h_x, \quad -p = Ch_{xx}, \quad h_t + uh_x = v - E,$$

This is now coupled to the diffusion equation for the volume fraction of solvent ϕ , in leading order (with D of order ε^2)

$$\phi_t + \phi_x u + \phi_y v = \phi_{yy},$$

with at $y = h(x, t)$ $\phi_y = -(1 - \phi)E$, and at $y = 0$ $\phi_y = 0$.

As usual in a lubrication analysis, one solves for u, v and p to obtain an equation for h , the well known thin film equation with evaporation term $E(\phi) = K\phi$. In the convection-diffusion equation we need explicitly the expressions for the velocities; writing $u = \psi_y, v = -\psi_x$, we can summarize the leading order lubrication model for $\phi(x, y, t)$ and $h(x, t)$

$$\left\{ \begin{array}{ll} \phi_t + \phi_x \psi_y - \phi_y \psi_x = \phi_{yy}, & 0 < y < h(x, t), \\ \psi = -Ch_{xxx}(\frac{1}{6}y^3 - \frac{1}{2}hy^2), & 0 < y < h(x, t), \\ \phi_y = 0, & \text{for } y = 0, \\ \phi_y = -K(1 - \phi)\phi, & y = h(x, t), \\ h_t + \frac{C}{3}(h^3 h_{xxx})_x = -K\phi, & y = h(x, t), \end{array} \right. \quad (11)$$

for $t > 0$ and appropriate x . These equations are to be supplemented by initial conditions and boundary conditions in x .

5 Stability of the unsteady flow

We consider a linear perturbation analysis of the one-dimensional solution for the full free boundary problem in section 2, coupled to Stokes flow (so not for the reduced problem in section 4, but we will comment on the small aspect ratio situation later). We start with assuming that the boundary is of the form

$$h(x, t) = H^0(t) + \delta H^1(t) \cos kx + \mathcal{O}(\delta^2)$$

for some small initial amplitude δ . We would now like to define $\phi = \Phi^0 + \delta\Phi^1 \cos kx + \mathcal{O}(\delta^2)$, and similarly for ψ , but since naturally ϕ is defined on $Q_T(h) = \mathbb{R} \times (0, h(x, t)) \times (0, T]$, while Φ^i is defined on $Q_T(H^0)$ (defined similarly), we write symbolically

$$\begin{aligned} \phi(x, y, t) &\sim \Phi^0(y, t) + \delta\Phi^1(y, t) \cos kx + \mathcal{O}(\delta^2), \\ \psi(x, y, t) &\sim \delta\Psi^1(y, t) \sin kx + \mathcal{O}(\delta^2). \end{aligned}$$

Of course the same dilemma is encountered when prescribing the initial data ϕ_0 and we need to assume that a proper ϕ_0 can be formulated on $Q_T(H^0)$ with the same total solvent mass. We now use a Taylor expansion around $H^0(t)$, which causes additional higher order terms in the free boundary conditions. Since \mathbf{q} , like ψ , is of the order $\mathcal{O}(\delta)$, the convection does not appear in the leading order equations for the volume fraction. The equations for Φ^0, H^0 are given by the one-dimensional Stefan problem (6) of section 3.

Since we have now a flow defined on a fixed domain, the Ψ^1 in these equations, which is directly related to the vertical velocity, can be found independent of H^1 and Φ^1 . After elimination of the pressure p from the Stokes equations,

$$p_x = \psi_{xxy} + \psi_{yyy}, \quad p_y = -\psi_{xxx} - \psi_{xyy},$$

we solve the bi-laplacian $\Delta^2\Psi^1 \sin kx = 0$ on the domain $Q_T(H^0)$, giving

$$\Psi^1 = (P + Qy)e^{ky} + (R + Sy)e^{-ky}.$$

Then we can solve for the pressure (up to a constant):

$$p^1 = -2k \cos kx(Qe^{ky} + Se^{-ky}).$$

Satisfying the boundary conditions, at $y = 0$ $\Psi^1 = \Psi^1_y = 0$, and at $y = H^0(t)$

$$\Psi^1_{yy} - \Psi^1_{xx} = 0, \quad -p^1 - 2\Psi^1_{xy} \cos kx = -C\kappa/\delta = -CH^1k^2 \cos kx,$$

gives the four conditions

$$\begin{aligned} P + R &= 0, \quad 2Pk + Q + S = 0, \\ (2Pk + 2Q + 2QkH^0)e^{kH^0} + (-2Pk - 2S + 2SkH^0)e^{-kH^0} &= 0, \\ (2Pk + 2QkH^0)e^{kH^0} + (2Pk - 2SkH^0)e^{-kH^0} &= CH^1k. \end{aligned}$$

Solving these equations for the coefficients P to S , we find that the velocity at $y = H^0(t)$ satisfies

$$v(x, H^0, t) = \frac{1}{2}C\delta H^1 k \frac{kH^0 - \sinh kH^0 \cosh kH^0}{k^2 H^{02} + \cosh^2 kH^0} \cos kx. \tag{12}$$

See figure 5 for a plot of this velocity. We observe that for $H^0 > 5/k$, the value of v is nearly independent of $H^0(t)$,

$$v(x, H^0, t) \approx -\frac{1}{2}Ck\delta H^1 \cos kx. \tag{13}$$

Using this in the equation for H^1 , we find for H^1 the following exponential decay

$$H_t^1 = -\frac{1}{2}kCH^1 - K(\Phi^1 + H^1\Phi_y^0),$$

evaluated at $y = H^0(t)$ and with initial value $H^1(0) = 1$. At first inspection we conclude that for short time, the disturbance may grow, since $\Phi_y^0 < 0$ at $y = H^0$. This depends on the values of the wavenumber k (assumed large here) and C . Even when the disturbance grows initially, it depends on the definition whether we call this unsteady basic flow stable or unstable [9]. When assuming that all quantities are uniformly bounded, we observe that for large time the disturbance will always die out. This can be seen from the fact that $\Phi^0 \rightarrow 0$ in time, since Φ^0 satisfies (6) independent of H^1 and Φ^1 . This implies that the forcing terms in the equations for Φ^1 vanish. With the given function $H^0(t)$ from (6) we argue similarly that $\Phi^1 \rightarrow 0$ for large time, while $H^0 \rightarrow h_1$. This implies an exponential decay of $H^1 \sim \exp(-Ckt/2)$ for short wavelengths (short relative to the inverse layer thickness).

For small values of H^0 , i.e. $H^0 < 1/k$ we can approximate v by

$$v(x, H^0, t) \approx -\frac{1}{3}Ck^4\delta H^1 H^{03} \cos kx.$$

It can be checked that this is also the approximation that is found from the thin film equation, when substituting $h(x, t) = H^0(t) + \delta H^1(t) \cos kx$. Now H^1 satisfies

$$H_t^1 = -\frac{1}{3}k^4CH^{03}H^1 - K(\Phi^1 + H^1\Phi_y^0),$$

and for small k the disturbance will indeed grow initially, but again with $\Phi^0 \rightarrow 0$ and $\Phi^1 \rightarrow 0$ for large time, we find $H^1 \sim \exp(-Ck^4h_1^3t/3)$ for these long wavelengths or thin layers.

Although we cannot directly conclude stability of the unsteady flow from this analysis, we observe that both long and short wavelength perturbations will die out in the long run, assuming that the linearization remains applicable. Since a separation of variables is not suitable due to the non-autonomous boundary conditions, global estimates are needed to define a proper criterion for stability [9].

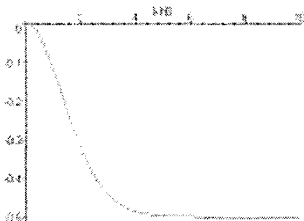


Fig. 4. Velocity $v/(C\delta H^1 k \cos kx)$ as a function of kH^0 .

6 Conclusion

We have restricted ourselves here to the derivation of a model for the drying of a paint film in a specific parameter regime. One motivation was to get an idea of the role of surface tension in regularizing the Stefan problem which is found from diffusion of species rather than phase transitions, for which the Gibbs Thomson law is used. Three small parameter cases were discussed: first the small time asymptotics for the movement of the interface, secondly the derivation of a model for a small aspect ratio situation (a thin layer lubrication model) and finally a small amplitude perturbation of the flat paint-air interface. Visco-elastic effects have not been included and these will be addressed in future work.

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