

Asymptotic analysis of particle engulfment

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An asymptotic analysis is conducted on the interaction between an insoluble spherical particle and an advancing solid-liquid interface when the particle is in the near-contact region (gap thickness is much smaller than the particle's radius). The analysis considers only thermal effects in a pure substance. The interface equilibrium temperature includes the undercooling effects due to the front curvature and to the long-range intermolecular forces in the thin melt film behind the particle. An expression for the crystal-particle gap thickness is derived and used to calculate the threshold value for the front velocity V_T which separates pushing from engulfment. [S1063-651X(99)01211-8]

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I. INTRODUCTION

It is well known that when a particle interacts with a solidifying interface, it is either pushed or engulfed in the solid. Several experimental and theoretical studies have been performed for the purpose of quantifying the pushing or engulfment phenomenon [1–10]. These studies have demonstrated the existence of a critical value of the growth rate which separates pushing from engulfment. The dependence of this critical velocity on the physical and processing parameters has not been fully resolved and remains an intensive area of research. We refer the reader to Ref. [8] for an extensive review of the problem.

In this Brief Report, we allow one neutrally buoyant and insoluble spherical particle to be near enough the interface that the gap between the planar front and the nearest point on the sphere is much smaller than the radius of the particle. We shall conduct a small-gap asymptotic analysis [11] for the purpose of (i) determining the interface shape near the particle in the near-contact region (Fig. 1), whence determining the dependence of the minimum gap on the physical parameters and (ii) using the gap to calculate the forces that act on the particle since the calculated inner field provides the dominant forces on the particle. These forces consist of the viscous drag force, which opposes the movement of the particle, and the Van der Waals force which pushes the particle away from the interface. By balancing these two forces, we obtain an expression for the growth rate which separates pushing from engulfment.

II. MATHEMATICAL MODEL

We consider the directional solidification of a pure substance in such a way that the solidifying front is moving vertically upward with velocity V . We assume that an inert and neutrally buoyant spherical particle is located at a distance h^* from the planar solid-liquid interface. The system is described by the heat conduction equations in the liquid and solid phases and in the particle with the assumption of mass conservation. We consider an axisymmetric geometry with a moving cylindrical coordinate system which immobilizes the solid-liquid interface. With the vertical coordinate denoted by z^* and the radial coordinate, which is taken along the

solid-liquid interface, by r^* , the governing equations for the dimensional variables (denoted by $*$) are

$$\frac{\partial T^*}{\partial t^*} - V \frac{\partial T^*}{\partial z^*} = \kappa \frac{1}{r^*} \frac{\partial}{\partial r^*} \left(r^* \frac{\partial T^*}{\partial r^*} \right) + \frac{\partial^2 T^*}{\partial z^{*2}}, \quad (1)$$

where T^* and κ are the temperature and the thermal diffusion coefficient in the liquid. Similar equations hold for the temperature T_S^* in the solid, with κ_S in place of κ , and for

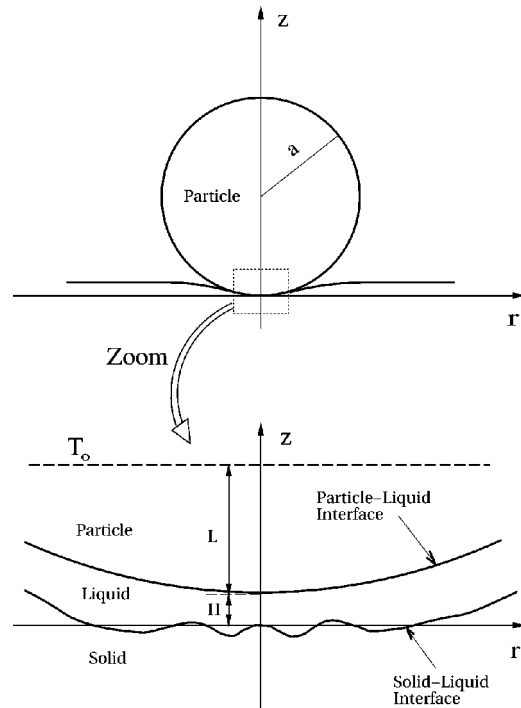


FIG. 1. Sketch of the near-contact region. Top: A spherical particle of radius a positioned near the solid-liquid interface. Note the deformation in the latter. This situation pertains to the case $\mu > 1$. Bottom: A magnification of the near-contact region shown in a box in the top figure. The waviness (slightly exaggerated here) in the solid front is of the order of the gap thickness. H is the depth of the melt film in dimensionless form and the reference temperature T_0 is measured at a distance L from the particle-melt boundary ($d = L + H$).

the temperature T_p^* in the particle, with κ_p in place of κ , κ_S , and κ_p being the thermal diffusion coefficients of the solid and particle, respectively.

At the particle-liquid boundary, $z^* = h^* + a - \sqrt{a^2 - r^{*2}} \equiv \mathcal{B}^*(r^*)$, the continuity of the temperature and of the heat flux yield $T^* = T_p^*$ and $\nabla(kT^* - k_p T_p^*) \cdot \mathbf{n} = 0$, where k and k_p are the thermal conductivities of the melt and the particle, respectively, \mathbf{n} denotes the normal to the particle-melt boundary that is pointing into the liquid and ∇ is the gradient vector. Furthermore, we will allow T_p^* to have some prescribed value T_0^* at some elevation $z^* = d^*$. If we neglect the kinetic and hydrodynamic effects, the solid-liquid interface equilibrium temperature is given by (see Ref. [8]) $T^* = T_S^* = T_m + \Delta T_{\text{curv}} + \Delta T_{dp}$, where ΔT_{curv} is the curvature undercooling given by the Gibbs-Thomson formula, $\Delta T_{\text{curv}} = [\alpha\Omega/\Delta H_f]\mathcal{K}$, where α is the crystal-melt interface energy, ΔH_f is the molar entropy of fusion, Ω denotes the molar volume of the liquid phase (taken to be the same as the molar volume in the solid phase), and \mathcal{K} is the mean curvature of the interface. The undercooling term ΔT_{dp} represents the change in the melting temperature due to the Van der Waals forces in the film separating the particle from the interface and is given by (see Ref. [8]) $\Delta T_{dp} = [\Omega/\Delta H_f][\mathcal{A}/6\pi g^3(r^*)]$, where \mathcal{A} is the Hamaker constant and $g(r^*)$ is the gap thickness, i.e., the depth of the film melt that is sandwiched between the particle and the solid-liquid interface. Far away from the interface, the temperature gradient in the solid phase is maintained at G_S^* .

When the particle enters the near-contact region, it induces pressure variations in the melt film. These pressure changes owe their existence to the disjoining pressure \mathcal{P}_{DP} and the pressure that is associated with the front curvature as a result of the Gibbs-Thomson effect \mathcal{P}_{GT} . For a spherical particle of radius a , the pressure variation that is induced by the front deformation, at the origin, is $\mathcal{P}_{GT} = \alpha\mathcal{K} = \alpha/2a$, while the disjoining pressure is given by $\mathcal{P}_{DP} = -\mathcal{A}/6\pi g^3(r^*)$. Following Ref. [8], we let l be the characteristic length scale over which the disjoining pressure \mathcal{P}_{DP} in the film balances the pressure due to the Gibbs-Thomson effect \mathcal{P}_{GT} , i.e., $|\mathcal{A}|/6\pi l^3 = \alpha/2a$, from which it follows that $l = [a|\mathcal{A}|/3\pi\alpha]^{1/3}$.

In the nondimensionalization of the equations and boundary conditions, we use l as lengthscale in the vertical direction and the radius of the particle a , as a lengthscale in the horizontal direction, l^2/κ as a scale for time and the fusion temperature of the pure substance T_m as a scale of temperature. The following equations in dimensionless form are obtained

$$\frac{\partial T}{\partial t} - v \frac{\partial T}{\partial z} = q \left[\epsilon^2 \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T}{\partial r} \right) + \frac{\partial^2 T}{\partial z^2} \right], \quad (2)$$

where $q = 1$ in the liquid phase, $q = \kappa_S/\kappa$ in the solid phase, and $q = \kappa_p/\kappa$ in the particle, and where $\epsilon = l/a$ and $v = lV/\kappa$.

The corresponding boundary conditions in dimensionless form are discussed in the following. Since we are focusing on the liquid region that is sandwiched between the particle and the solid-liquid interface, the upper boundary for the liquid region is given by $\epsilon z = (h^*/a) + 1 - \sqrt{1 - r^2}$, where

h^* is the distance between the planar interface and the nearest point on the particle, i.e., the minimum clearance between the particle and the solid front. The particle is assumed to be in the near-contact region so that $h^* \ll a$. We let $h^*/a = \epsilon H$ where $H = O(1)$. At this upper boundary, which we denote by $\mathcal{B}(r)$, the continuity of the heat flux and of the temperature imply $\nabla(T - \mu T_p) \cdot \mathbf{n} = 0$ and $T_p = T$, where μ is the ratio of the thermal conductivity of the particle to that of the liquid ($\mu = k_p/k$). In dimensionless form, the temperature at the interface $z = \eta(r, t)$ satisfies

$$T = T_S = 1 + \frac{\Gamma \epsilon}{\sqrt{1 + \epsilon^2 \eta_r^2}} \left[\frac{\eta_r}{r} + \frac{\eta_{rr}}{1 + \epsilon^2 \eta_r^2} \right] + \beta g^{-3}(r), \quad (3)$$

where $\eta(r, t)$ is the deviation of the interface from planarity, the subscript r denotes differentiation with respect to r , and $\Gamma = \alpha\Omega/l\Delta H_f T_m$, $\beta = \mathcal{A}\Omega/6\pi l^3 \Delta H_f T_m$. The parameter Γ describes the influence of the front curvature and β characterizes the effect of the pressure put forth by the Van der Waals forces. Note that a positive (negative) Hamaker constant implies $\beta > 0$ ($\beta < 0$).

An estimation of the order of magnitude of the various dimensionless parameters that appear in the equations and boundary conditions is now undertaken. Following Ref. [8] we consider a typical physical situation with $\mathcal{A} = 10^{-15}$ erg (1 erg = 10^{-7} J), $\alpha \approx 50$ erg/cm², and particle radius $a \approx 10^{-3}$ cm. Then the characteristic length scale is $l \approx 10^{-7}$ cm which yields $\epsilon \approx 10^{-4}$. For $V \approx 10^{-4}$ cm/s, $\kappa \approx 10^{-1}$ cm²/s, and $\Omega/\Delta H_f T_m \approx 3 \times 10^{-9}$ cm³/erg, we obtain $\Gamma \approx 0.13$, $\beta \approx 1.6 \times 10^{-4}$, and $v \approx 10^{-10}$. Hence appropriate scalings for the dimensionless parameters are $v = \epsilon^2 \hat{v}$, $\beta = \epsilon \hat{\beta}$, and $\Gamma = O(1)$ where the hat symbol denotes $O(1)$ quantities.

III. ANALYSIS

In this section we conduct an asymptotic analysis in the vicinity of the origin. For this purpose, we introduce the stretched coordinate $r = \sqrt{\epsilon} R$, a scaling that is suggested by the dimensionless location of the melt-particle boundary, i.e., $\epsilon z = \epsilon H + 1 - \sqrt{1 - r^2}$. The following system of equations and boundary conditions results:

$$\frac{\partial T}{\partial t} - \epsilon^2 \hat{v} \frac{\partial T}{\partial z} = q \left[\epsilon \frac{1}{R} \frac{\partial}{\partial R} \left(R \frac{\partial T}{\partial R} \right) + \frac{\partial^2 T}{\partial z^2} \right], \quad (4)$$

with similar equations in the solid phase and the particle. The upper boundary is now given by

$$z = H + \frac{R^2}{2} + \epsilon \frac{R^4}{8} + \dots = \mathcal{B}(R). \quad (5)$$

On $z = \mathcal{B}(R)$, the continuity of temperature and of the heat flux yields

$$\frac{\partial T}{\partial z} - \epsilon \frac{\partial \mathcal{B}}{\partial R} \frac{\partial T}{\partial R} = \mu \left[\frac{\partial T_p}{\partial z} - \epsilon \frac{\partial \mathcal{B}}{\partial R} \frac{\partial T_p}{\partial R} \right] \quad \text{and} \quad T = T_p. \quad (6)$$

The temperature in the solid and liquid phases at the interface $z = \eta(R, t)$ is given by

$$T = T_S = 1 + \frac{\Gamma}{\sqrt{1 + \epsilon \eta_R^2}} \left[\frac{\eta_R}{R} + \frac{\eta_{RR}}{1 + \epsilon \eta_R^2} \right] + \epsilon \hat{\beta} g^{-3}(R). \quad (7)$$

We solve the steady-state version of the above problem by perturbation techniques using ϵ as the perturbation parameter. In order to keep the interface planar in the basic solution, we let the thermal conductance ratio μ deviate slightly from 1. Recall that $\mu > 1$ corresponds to a particle that is more conducting than the melt while $\mu < 1$ corresponds to a particle that is more insulating than the melt. Hence, we let $\mu = 1 + \epsilon \hat{\mu}$ where $\hat{\mu}$ is an $O(1)$ quantity that could be either positive or negative. This is done simply for the sake of simplifying the algebra. For $\mu - 1 = O(1)$, one has to consider a perturbation about a nonplanar interface. Let

$$[T, T_S, T_p, Z] = [\bar{T}, \bar{T}_S, \bar{T}_p, 0] + \epsilon [\theta, \theta_S, \theta_p, \eta] + O(\epsilon^2). \quad (8)$$

Then the $O(1)$ problem ($\epsilon = 0$) pertains to the case of a planar interface and is described by $\bar{T}(z) = (\Delta T/d)z + 1$, for $0 \leq z \leq H + R^2/2$, $\bar{T}_p = (\Delta T/d)(z - d) + T_0$, for $H + R^2/2 \leq z \leq d$, and $\bar{T}_S = G_S z + 1$, for $z \leq 0$, where $\Delta T = T_0 - 1$. Here $\Delta T/d$ is simply the temperature gradient across a layer consisting of the liquid film and a portion of the particle. The particle's presence does not affect the temperature profiles for two reasons: (i) at leading order the thermal conductivities of the melt and particle are equal so that the heat flow does not distinguish between melt and particle and (ii) the effect of the disjoining pressure does not appear at $O(1)$ but enters at the next order in the calculations, i.e., $O(\epsilon)$. Because the effect of the presence of the particle is not felt at the leading order of the calculations, the temperature profiles are all linear in the vertical coordinate. The ratio of the temperature difference ΔT over the depth d can be written as $\Delta T/d = (\Delta T_p + \Delta T_m)/(L + H)$, where H is the depth of the melt layer under the particle, $L = d - H$, ΔT_m , and ΔT_p are the dimensionless temperature changes over the depths H and L , respectively (see Fig. 1). In addition, the $O(1)$ contribution to the continuity of the heat flux at the particle-melt interface implies $\Delta T_p/L = \Delta T_m/H$, which when combined with the previous relation, yields an expression for the parameter d , namely, $d = H[1 + \Delta T_p/\Delta T_m]$. Next we proceed with the solutions to the $O(\epsilon)$ contributions for θ , θ_S , and θ_p . These expressions are not displayed in this report. The heat energy balance at the solid-liquid interface implies

$$\nabla [KT_S - T] \mathbf{n} = \mathcal{L} \left[\mathcal{V} + \frac{\partial \eta}{\partial t} \right], \quad (9)$$

where $K = k_S/k$ denotes the ratio of the thermal conductivity of the solid phase (k_S) to that of the liquid phase (k), $\mathcal{L} = \Delta H_f \kappa / k T_m$ and \mathcal{V} is the dimensionless growth rate. The leading order term of the latter is calculated from Eq. (9) to get $\mathcal{V} = KG_S - \Delta T/d = KG_S - \Delta T_m/H$. This expression for the dimensionless growth velocity is similar to the one corresponding to the directional solidification of a pure substance without particle with $\Delta T_m/H$ being the thermal gradient in the melt. The effect of the particle does not appear at this order of the

calculations. At $O(\epsilon)$ of Eq. (9) we obtain an evolution equation for the interface perturbation $\eta(R, t)$, namely,

$$\mathcal{L} \frac{\partial \eta}{\partial t} = - \frac{\Delta T}{d} \eta + \Gamma \left[\eta_{RR} + \frac{\eta_R}{R} \right] + \hat{\mu} \frac{\Delta T}{d} \left(H - d + \frac{R^2}{2} \right) + \hat{\beta} \left(H + \frac{R^2}{2} \right)^{-3}. \quad (10)$$

If we make the change of variable $x = R \sqrt{\Delta T/\Gamma d}$, then the steady state solution of Eq. (10) that is bounded at $x = 0$ and satisfies the symmetry condition at $x = 0$, i.e., $d\eta/dx(0) = 0$, is given by

$$\eta(x) = \hat{\mu} \left[H - d + \frac{2\Gamma d}{\Delta T} \right] + \frac{\hat{\mu} \Delta T}{2\Gamma d} x^2 + \frac{\hat{\beta} d}{\Delta T} \left[\int_x^\infty \frac{y K_0(y) I_0(x) dy}{[H + (\Gamma d/2\Delta T)y^2]^3} - \int_0^x \frac{y I_0(y) K_0(x) dy}{[H + (\Gamma d/2\Delta T)y^2]^3} \right], \quad (11)$$

where $I_0(x)$ and $K_0(x)$ are the modified Bessel functions of order zero of the first and second kind, respectively. Equation (11) implies that

$$\eta(0) = \hat{\mu} \left[H - d + \frac{2\Gamma d}{\Delta T} \right] + \frac{\hat{\beta} d}{\Delta T} \int_0^\infty \frac{y K_0(y) dy}{[H + (\Gamma d/2\Delta T)y^2]^3}, \quad (12)$$

which, with the condition $d\eta/dx(0) = 0$, provides the initial conditions that are needed to alternatively solve the steady version of Eq. (10) for $\eta(x)$ numerically. These calculations are beyond the scope of this report. The knowledge of the gap thickness, namely,

$$g(x) = H + \frac{\Gamma d}{2\Delta T} x^2 + \epsilon \left[\left(\frac{\Gamma d}{\Delta T} \right)^2 \frac{x^4}{8} - \eta(x) \right], \quad (13)$$

allows the calculations of the forces acting on the particle. In this report, we consider the simple case of using the gap thickness at the origin to calculate the force balance on the particle. Thus, we are in fact ignoring the interface distortions. The inclusion of the latter will require the integration of Eq. (11) and is beyond the scope of this Brief Report. The gap thickness at the origin is given by $g(0) = H - \epsilon \eta(0)$, which, upon using Eq. (12) and the definition of d , can be written as $g(0) = \Psi H - \Phi/H^2$, where

$$\Psi = \left\{ 1 - \epsilon \hat{\mu} \left[1 - \left(1 + \frac{\Delta T_p}{\Delta T_m} \right) \left(1 - \frac{2\Gamma}{\Delta T} \right) \right] \right\}, \quad (14)$$

$$\Phi = \frac{\epsilon \hat{\beta}}{\Delta T_m} \int_0^\infty \frac{y K_0(y) dy}{[1 + (\Gamma/2\Delta T_m)y^2]^3}. \quad (15)$$

The minimum gap thickness at the origin is calculated by minimizing $g(0)$ with respect to H . We find that $g(0)$ has a minimum at $H_c = [-2\Phi/\Psi]^{(1/3)}$ if $\Phi < 0$ (which corresponds to $\hat{\beta} < 0$ or equivalently a positive disjoining pres-

sure), and has no minimum if $\Phi \geq 0$. For $\hat{\beta} < 0$, the minimum gap thickness at the origin is $g_m = (-3/2)(\Phi\Psi^2)^{(1/3)}$.

Now that an expression for the gap thickness is obtained, we turn our attention to the calculation of the forces that act on the particle. Given our assumption of a neutrally buoyant particle and by neglecting any buoyancy due to the thermal expansion of the fluid, the particle is then subjected to only two forces (assumed opposing). The first one is the repelling Van der Waals force [8], $F_+ = \mathcal{A}a/6g_m^{*2}$, where the asterisk indicates the dimensional minimum thickness, and the second one is the compressing Stokes force which accounts for the presence of the interface and derived by Brenner [12]. Given that $g_m^* \ll 1$, we have $F_- = 6\pi\lambda Va^2/g_m^*$, where λ is the melt's viscosity, and V is the velocity of the solid-liquid interface. A force balance, $F_- = F_+$, then yields an expression for the growth rate V_T that separates pushing from engulfment, i.e., $V_T = |\mathcal{A}|/[36\pi\lambda a g_m^*]$.

Using the definitions of H , ϵ , the length scale l , Γ , $\hat{\beta}$, and $\hat{\mu}$, V_T is then expressed in dimensional form. For instance, in the limit of vanishing disjoining pressure ($\hat{\beta}=0$), V_T is given by

$$V_T = |\mathcal{A}| \mathcal{G} \left[36\lambda [\Delta T_m^* + (1-\mu)\Delta T_p^*] \pi a + 6\pi(1-\mu)\lambda \left(\frac{12(3\pi)^{1/3}(\alpha)^{4/3}\Omega}{|\mathcal{A}|^{1/3}\Delta H_f} \right) a^{2/3} \right]^{-1}, \quad (16)$$

where $\mathcal{G} = \Delta T_m^*/h$ is the dimensional thermal gradient in the melt, $\Delta T_m^* = T_m \Delta T_m$, and $\Delta T_p^* = T_m \Delta T_p$. Equation (16) isolates the thermal conductivity effect on V_T . The derivation of a similar expression for V_T , which includes the disjoining pressure effect, is complicated by the presence of the integral term in Eq. (15), and is not considered here.

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