Interface evolution of a particle in a supersaturated solution affected by a far-field uniform flow*

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The effect of far-field uniform flow on the morphological evolution of a spherical particle in a supersaturated solution affected by a far-field uniform flow is studied by using the matched asymptotic expansion method. The analytical solution for the interface shape, concentration field, and interface velocity of the particle growth shows that the convection induced by the far-field uniform flow facilitates the growth of the spherical particle, the upstream flow imposed on the particle enhances the growth velocity of the interface when the flow comes in, the downstream flow lowers the growth velocity of the surface when the flow goes out, and the interface morphology evolves into a peach-like shape.

Keywords: convection, interfaces, growth models, crystal morphology

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1. Introduction

Understanding the convection effect in crystallization is essential for the accurate control of microstructure formation.[1,2] The convection driven by mechanical or electromagnetic stirring determines the morphological evolution of the particles and is beneficial for the enhancement of the mechanical and physical properties of the final products. Unlike the conventionally solidified dendritic microstructure, the globular crystals can directly nucleate from the stirred melt and grow spheroidally into large size ones.[3] The upstream flow imposed on the spherical crystal enhances the growth velocity of the interface when the flow is incoming.[4–7] In the stirred melt, a great number of pear-like or peach-like spherical crystals with nanometer sizes significantly improve the grain refinement and the mechanical properties of the solidified material, such as tensile strength and elongation.[7] Though various experimental observations and numerical simulations explicitly suggest the significant effect of the convection on the interface microstructure formation, theoretical investigations of the morphological evolution of interface during crystallization in the stirred melt and solution are still very limited due to the complicated effects of convection.[2,8–17] Hence, there is a strong need for the common analytical method to predict the experimentally observed particle morphologies developed under different flow conditions. A feasible way is to model the growth of spherical particles in the complex flow of the stirred liquid as a singularly perturbed free boundary problem and find its asymptotic solution by using the matched asymptotic expansion method. In this paper, we extend our studies[18–20] to the effect of the far-field uniform flow on the morphological evolution of a spherical particle in a supersaturated solution. By employing the matched asymptotic expansion method, we find the asymptotic solution in the entire solution region. With the numerical calculation of the obtained solution, we show the interface morphologies and growth velocities of the spherical particle growing in the convective solution under different far-field uniform flow conditions.

2. Mathematical model

We consider the diffusion-controlled and convection-controlled growth of a spherical precipitate particle of initial radius r_0 in a uniformly supersaturated solution. It is assumed that far from the sphere the solute concentration is C_∞ and the fluid velocity is induced by the downward far-field uniform flow $\boldsymbol{U}_\infty$. For nondimensionalization, we use the dimensionless variables, the length and the time are scaled by r_0 and $r_0^2(C_S - C_0)/(D_L \Delta C)$, respectively, where D_L is the solute diffusion coefficient in the solution, C_0 is the equilibrium concentration at the flat interface, C_S is the concentration in the crystal, and ΔC is the solute concentration gradient, $\Delta C = C_\infty - C_0$. For simplicity, C_S is assumed to be a constant. It is assumed that the solution is very dilute and the impurity rejection from the interface is negligible. We denote the convection velocity, pressure, and solute concentration of the solution by $\boldsymbol{U}$, P, and C, respectively, which are scaled respectively by D_L/r_0, $(C_S - C_0)\rho_L V^2/\Delta C$, and ΔC, where ρ_L is the solution density. In the spherical coordinates (r, θ, φ) with the origin located in the center of the particle in the ambient supersaturated solu-

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tion, the interface of the sphere is written as $r = R(\theta, \varphi, t)$. After all the bars over their heads are dropped, the governing equations for the convection velocity, pressure, and solute concentration of the solution are expressed in the dimensionless forms as

$$\nabla \cdot \boldsymbol{U} = 0, \tag{1}$$

$$\varepsilon \frac{\partial \boldsymbol{U}}{\partial t} + \varepsilon(\boldsymbol{U} \cdot \nabla)\boldsymbol{U} = -\nabla P + S_{\mathrm{c}} \Delta \boldsymbol{U}, \tag{2}$$

$$\varepsilon \frac{\partial C}{\partial t} + \varepsilon(\boldsymbol{U} \cdot \boldsymbol{\nabla})C = \Delta C, \tag{3}$$

where $\varepsilon = (C_\infty - C_0)/(C_{\mathrm{S}} - C_0)$, $S_{\mathrm{c}} = \upsilon / D_{\mathrm{L}}$, and υ is the kinematical viscosity.

At the interface, the total mass conservation, the tangential non-slip condition, and the solute conservation condition hold

$$\boldsymbol{U} \cdot \boldsymbol{n} = 0, \quad \boldsymbol{U} \cdot \boldsymbol{\tau} = 0, \tag{4}$$

$$(C - C_{\mathrm{S}})U_{\mathrm{I}} = -\nabla C \cdot \boldsymbol{n}, \tag{5}$$

where U_{I} is the growth velocity of the interface, and $\boldsymbol{n}$ and $\boldsymbol{\tau}$ are the unit normal and tangent vectors to the interface, respectively. It is assumed that the dilute solution theory holds; the equilibrium concentration at the interface is determined by the capillarity condition

$$C_{\mathrm{I}} = -\Gamma K, \tag{6}$$

where C_{I} is the concentration at the interface, K is the twice local mean curvature (taken to be positive for a spherical crystal), and Γ is a capillary parameter, $\Gamma = \gamma C_0 \Omega / (r_0 R_{\mathrm{G}} T \Delta C)$, in which γ is the interfacial free energy, Ω is the increment of precipitate volume per mole of added solute, R_{G} is the gas constant, and T is the absolute temperature.

Far from the surface of the spherical crystal, the far-field conditions hold, as $r \to \infty$,

$$\boldsymbol{U} \to -U_\infty \boldsymbol{k}, \tag{7}$$

$$C \to 1, \tag{8}$$

where U_∞ is a dimensionless parameter scaled by D_{L}/r_0, and $\boldsymbol{k}$ is the unit vector to the northern pole in the rectangular coordinates.

Finally, the initial condition holds, in which the initial condition for the interface at time $t = 0$ is

$$R(\theta, \varphi, 0) = 1. \tag{9}$$

In the above, for simplicity, the physical properties of the material are assumed to be constant and the buoyancy effect is neglected.

3. Asymptotic solution

We restrict our attention to the case[21] in which $|C_\infty - C_0| \ll |C_{\mathrm{S}} - C_0|$, it follows that $\varepsilon = (C_\infty - C_0)/(C_{\mathrm{S}} - C_0) << 1$. We take ε as a small parameter. It implies that the solution is very dilute and the impurity rejection from the interface is negligible. By using the matched asymptotic expansion we seek the asymptotic solution in the entire solution region as $\varepsilon \to 0$,

$$\psi \sim \psi_0 + \varepsilon \psi_1 + \varepsilon^2 \psi_2 + \cdots, \tag{10}$$

$$C \sim C_{\mathrm{B}0} + \varepsilon C_{\mathrm{B}1} + \varepsilon^2 C_{\mathrm{B}2} + \cdots, \tag{11}$$

$$R \sim R_{\mathrm{B}0} + \varepsilon R_{\mathrm{B}1} + \varepsilon^2 R_{\mathrm{B}2} + \cdots, \tag{12}$$

where ψ is the stream function of the flow field $\boldsymbol{U}$. We further expand each order approximation in Eqs. (10)–(12) into a series of spherical harmonics. Substituting Eqs. (10)–(12) into Eqs. (1)–(9) and equating coefficients of like powers of ε, we obtain the equations and boundary conditions for each order approximation.

When $\partial/\partial t = \mathrm{O}(1)$, we have the leading order approximation

$$\psi_0 = \frac{1}{2}\left(-\frac{R_{\mathrm{B}0}^3}{2r^3} + \frac{3R_{\mathrm{B}0}}{2r} - 1\right) r^2 \sin^2\theta, \tag{13}$$

$$C_{\mathrm{B}0} = 1 - (R_{\mathrm{B}0} - 2\Gamma)\frac{1}{r}, \tag{14}$$

and $R_{\mathrm{B}0}$ is the solution of the ordinary differential equation

$$\frac{\mathrm{d}R_{\mathrm{B}0}}{\mathrm{d}t} = \frac{R_{\mathrm{B}0} - 2\Gamma}{R_{\mathrm{B}0}^2 (C_{\mathrm{S}} - C_{\mathrm{R}})}, \tag{15}$$

which obeys the initial condition (9), where $C_{\mathrm{R}} = 2\Gamma / R_{\mathrm{B}0}$ is the concentration on the sphere unaffected by the far-field uniform flow. Equation (15) demonstrates the symmetrical growth or decay of the spherical particle without the inclusion of the far-field flow. If $R_{\mathrm{B}0} > 2\Gamma$ and $\mathrm{d}R_{\mathrm{B}0}/\mathrm{d}t > 0$, then the crystal particle grows, whereas if $R_{\mathrm{B}0} < 2\Gamma$ and $\mathrm{d}R_{\mathrm{B}0}/\mathrm{d}t < 0$, the crystal particle decays. Thus $\mathrm{d}R_{\mathrm{B}0}/\mathrm{d}t = 0$ represents the critical state of growth with the critical value $R_{\mathrm{B}0} = 2\Gamma$, which corresponds to the dimensional critical radius of the nucleus $R_* = 2\gamma C_0 \Omega / (R_{\mathrm{G}} T \Delta C)$. When the initial radius is taken to be larger than the dimensional critical radius of the nucleus R_*, we obtain the leading order approximation of the interface $R_{\mathrm{B}0}$ in the implicit function form

$$\begin{aligned} t \;=\; & 4\Gamma^2 (C_{\mathrm{S}} - 1) \ln \frac{R_{\mathrm{B}0} - 2\Gamma}{1 - 2\Gamma} + \frac{1}{2} C_{\mathrm{S}} (R_{\mathrm{B}0}^2 - 1) \\ & + 2\Gamma (C_{\mathrm{S}} - 1)(R_{\mathrm{B}0} - 1). \end{aligned} \tag{16}$$

The first-order approximations are derived as follows:

$$\begin{aligned} C_{\mathrm{B}1} \;=\; & \frac{A_{0,0}}{r} + d_{0,0} - \frac{r}{2}\frac{\mathrm{d}R_{\mathrm{B}0}}{\mathrm{d}t} + \left[\frac{A_{1,0}}{r^2} - U_\infty (R_{\mathrm{B}0} - 2\Gamma)\right. \\ & \left. \times \left(\frac{R_{\mathrm{B}0}^3}{8r^3} + \frac{3R_{\mathrm{B}0}}{4r} - \frac{1}{2}\right)\right] \cos\theta, \end{aligned} \tag{17}$$

$$R_{\mathrm{B}1} \;=\; g_{0,0}(t) + g_{1,0}(t)\cos\theta, \tag{18}$$

where

$$
\begin{aligned}
A_{0,0} &= -\frac{(R_{\mathrm{B}0}-4\Gamma)}{R_{\mathrm{B}0}}g_{0,0}(t)+\frac{1}{2}R_{\mathrm{B}0}^2\frac{\mathrm{d}R_{\mathrm{B}0}}{\mathrm{d}t}-R_{\mathrm{B}0}d_{0,0},\\
A_{1,0} &= -(R_{\mathrm{B}0}-2\Gamma)g_{1,0}(t)+\frac{3U_\infty}{8}R_{\mathrm{B}0}^2(R_{\mathrm{B}0}-2\Gamma),\\
g_{0,0} &= \frac{4\Gamma^2(2C_{\mathrm{S}}-3)(C_{\mathrm{S}}-C_{\mathrm{R}})}{C_{\mathrm{S}}^4(R_{\mathrm{B}0}-2\Gamma)}\ln\frac{(C_{\mathrm{S}}-C_{\mathrm{R}})R_{\mathrm{B}0}}{C_{\mathrm{S}}-2\Gamma}\\
&\quad+\frac{(C_{\mathrm{S}}-C_{\mathrm{R}})}{(R_{\mathrm{B}0}-2\Gamma)}[F_1(R_{\mathrm{B}0})-F_1(1)]\\
&\quad-\frac{4U_\infty\Gamma^3(C_{\mathrm{S}}-1)(C_{\mathrm{S}}-C_{\mathrm{R}})}{C_{\mathrm{S}}^4(R_{\mathrm{B}0}-2\Gamma)}\ln\frac{(C_{\mathrm{S}}-C_{\mathrm{R}})R_{\mathrm{B}0}}{C_{\mathrm{S}}-2\Gamma}\\
&\quad+\frac{U_\infty(C_{\mathrm{S}}-C_{\mathrm{R}})}{6C_{\mathrm{S}}^3(R_{\mathrm{B}0}-2\Gamma)}[F_2(R_{\mathrm{B}0})-F_2(1)],\\
g_{1,0}(t) &= \frac{3U_\infty}{16}(R_{\mathrm{B}0}^2-1),
\end{aligned}
$$

in which $d_{0,0}=U_\infty(R_{\mathrm{B}0}-2\Gamma)/2$ is a matching constant determined by matching with the outer solution, $F_1(R_{\mathrm{B}0})$ and $F_2(R_{\mathrm{B}0})$ are expressed respectively as

$$
\begin{aligned}
F_1(R_{\mathrm{B}0}) &= \Big[2\Gamma C_{\mathrm{S}}^2(2C_{\mathrm{S}}-3)R_{\mathrm{B}0}^2-C_{\mathrm{S}}^3R_{\mathrm{B}0}^3-8\Gamma^2C_{\mathrm{S}}(C_{\mathrm{S}}-2)R_{\mathrm{B}0}\\
&\quad-16\Gamma^3(C_{\mathrm{S}}-1)\Big]\Big/\Big[2C_{\mathrm{S}}^4R_{\mathrm{B}0}(R_{\mathrm{B}0}-2\Gamma)\Big],\\
F_2(R_{\mathrm{B}0}) &= C_{\mathrm{S}}^2R_{\mathrm{B}0}^3-3\Gamma C_{\mathrm{S}}(C_{\mathrm{S}}-1)R_{\mathrm{B}0}^2-12\Gamma^2(C_{\mathrm{S}}-1)R_{\mathrm{B}0}.
\end{aligned}
$$

When proceeding to the higher-order terms, we obtain the asymptotic solution in Eqs. (10)–(12). It is noticed that the solution obtained in Eqs. (10)–(12) approaches some special functions as $t\to 0$, which does not satisfy the initial conditions given beforehand. Due to the presumption of $\partial/\partial t=\mathrm{O}(1)$ and $\varepsilon\to 0$, the solution for the concentration field is only valid in the time interval $[\mathrm{O}(\varepsilon),+\infty]$. In fact, during the early time there is a transient modification for the flow field and the concentration field. In order to explain the early-time behavior, we introduce time inner variable $\tau=t/\varepsilon$ and find the 'inner' solution in the sense of time. Similar to Chen *et al.*'s [18,20] analysis, by solving a classical initial-boundary value problem, it is proved that the obtained solution satisfies the initial conditions required for the growth system of the particle.

It can be seen from Eq. (17) that the first-order solution for the concentration field does not satisfy the vanishing condition at the far field. Actually, the first-order solution is derived from the first-order equations and boundary conditions, which include the viscosity effect but neglect the inertia effect shown by the inertial term $(\boldsymbol{U}\cdot\nabla)\boldsymbol{U}$ in Eq. (2). Since the viscosity effect dominates near the interface, whereas the inertia effect dominates far from the interface, the first-order solution in Eqs. (17) and (18) for the flow field is valid only near the interface. In order to eliminate the superficial paradox, we separate the entire special region into two sub-regions: one is the inner region that is near the interface of the spherical particle, and the other is the outer region that is far from the interface. In the outer region, we take the Oseen approximation, i.e., $(\boldsymbol{U}\cdot\boldsymbol{\nabla})\boldsymbol{U}\approx -U_\infty(\boldsymbol{k}\cdot\boldsymbol{\nabla})\boldsymbol{U}$. By introducing outer variable $\rho=\varepsilon r$ we transform Eqs. (1)–(3) with boundary conditions (7)–(9) into the 'outer' problem $\{\psi^{(\mathrm{o})},C^{(\mathrm{o})}\}$. By seeking the asymptotic expansion of $\psi^{(\mathrm{o})}$ and $C^{(\mathrm{o})}$ as $\varepsilon\to 0$, we find the outer solution that satisfies conditions (7) and (8) at the far field

$$\psi^{(\mathrm{o})}=-\frac{U_\infty\rho^2}{2\varepsilon^2}\sin^2\theta+\frac{1}{\varepsilon}\psi_1^{(\mathrm{o})}+\cdots, \tag{19}$$

$$C^{(\mathrm{o})}=1+\varepsilon C_1^{(\mathrm{o})}+\cdots, \tag{20}$$

where

$$
\begin{aligned}
\psi_1^{(\mathrm{o})} &= \frac{3S_{\mathrm{c}}}{2}R_{\mathrm{B}0}(1-\cos\theta)\left(1-\mathrm{e}^{-U_\infty\rho(1+\cos\theta)/2S_{\mathrm{c}}}\right),\\
C_1^{(\mathrm{o})} &= \mathrm{e}^{-U_\infty\rho\cos\theta/2}\sum_{n=0}^{\infty}B_n(t)\sqrt{\frac{\pi}{U_\infty\rho}}\mathrm{K}_{n+1/2}\\
&\quad\times\left(\frac{U_\infty\rho}{2}\right)P_n(\cos\theta),
\end{aligned}
$$

with $B_n(t)$ being the unknown functions of t, and $\mathrm{K}_{n+1/2}$ the modified Bessel function of the second kind. Further, we take the first terms in Eqs. (19) and (20) and match the outer solution in Eqs. (19) and (20) with the inner solution in Eqs. (10) and (11) respectively to determine the unknown functions

$$d_{0,0}=\frac{U_\infty}{2}(R_{\mathrm{B}0}-2\Gamma),\quad B_0(t)=-\frac{U_\infty}{\pi}(R_{\mathrm{B}0}-2\Gamma),\quad \ldots. \tag{21}$$

Therefore, by matching the inner solution with the outer one, we can obtain the uniformly valid asymptotic solution in the entire region. When we find the asymptotic solution to the first-order approximation, the asymptotic solution has the error of $\mathrm{O}(\varepsilon^2)$. According to the theory of solidification, the solute transfer occurs mainly near the vicinity of the interface. The concentration field and the interface morphology of the spherical particle near the interface are expressed respectively as

$$C=1-(R_{\mathrm{B}0}-2\Gamma)\frac{1}{r}+\varepsilon C_{\mathrm{B}1}+\mathrm{O}(\varepsilon^2), \tag{22}$$

$$R=R_{\mathrm{B}0}+\varepsilon[g_{0,0}(t)+g_{1,0}(t)\cos\theta]+\mathrm{O}(\varepsilon^2). \tag{23}$$

The growth velocity of the spherical particle is expressed as

$$\frac{\partial R}{\partial t}=I(t)+\frac{3\varepsilon U_\infty(R_{\mathrm{B}0}-2\Gamma)}{8R_{\mathrm{B}0}(C_{\mathrm{S}}-C_{\mathrm{R}})}\cos\theta+\mathrm{O}(\varepsilon^2), \tag{24}$$

where $I(t)$ denotes the part of symmetrical growth

$$
\begin{aligned}
I(t) &= \frac{\mathrm{d}R_{\mathrm{B}0}}{\mathrm{d}t}-\frac{\varepsilon}{R_{\mathrm{B}0}^2(C_{\mathrm{S}}-C_{\mathrm{R}})}\left[\left(1-2\Gamma\frac{\mathrm{d}R_{\mathrm{B}0}}{\mathrm{d}t}\right)g_{0,0}(t)\right.\\
&\quad\left.+R_{\mathrm{B}0}^2\frac{\mathrm{d}R_{\mathrm{B}0}}{\mathrm{d}t}-\frac{U_\infty R_{\mathrm{B}0}(R_{\mathrm{B}0}-2\Gamma)}{2}\right].
\end{aligned}
$$

It can be seen from Eqs. (23) and (24) that in the upstream front of the particle ($0<\theta<\pi/2$), if $R_{\mathrm{B}0}>2\Gamma$, then $\mathrm{d}g_{1,0}(t)/\mathrm{d}t>0$ and $\partial R/\partial t>I(t)$; if $R_{\mathrm{B}0}<2\Gamma$, then $\mathrm{d}g_{1,0}(t)/\mathrm{d}t<0$ and $\partial R/\partial t<I(t)$; in the downstream rear

$(\pi/2 < \theta < \pi)$, the situation is entirely contrary. The maximum velocity influenced by the far-field flow is

$$V_{\max} = \frac{3\varepsilon U_\infty (R_{B0} - 2\Gamma)}{8R_{B0}(C_S - C_R)}.$$

The maximum distance influenced by the far-field flow is

$$\begin{aligned} d_{\max} = & -\frac{4\varepsilon U_\infty \Gamma^3 (C_S - 1)(C_S - C_R)}{C_S^4 (R_{B0} - 2\Gamma)} \ln \frac{(C_S - C_R)R_{B0}}{C_S - 2\Gamma} \\ & + \frac{\varepsilon U_\infty (C_S - C_R)}{6C_S^3 (R_{B0} - 2\Gamma)} [F_2(R_{B0}) - F_2(1)] \\ & + \frac{3\varepsilon U_\infty}{16}(R_{B0}^2 - 1). \end{aligned}$$

Figure 1 shows that in the upstream front of the particle, the convective flow induced by the far-field flow makes the growing interface of the spherical particle further grow and inhibit the growth in the downstream direction. As the far-field uniform flow increases, the velocity of the interface increases, and then the interface protrudes into the solution. Figure 2 shows that the velocity of the interface in the upstream front is greater than that in the downstream rear as the far-field uniform flow increases. Figure 3 shows that compared with the situation without the far-field uniform flow, the interface of the spherical particle enhances its growth velocity near the surface where the convective flow comes in the upstream direction and lowers the growth velocity near the surface where the convective flow goes out, and the interface evolves into a peach-like shape. Thus the convective flow facilitates the growth of the interface. It is consistent with the results of Galenko *et al.*[4], Herlach and Galenko,[5] Chen and Chen,[11] Wang *et al.*,[17] and Chen *et al.*[18–20]

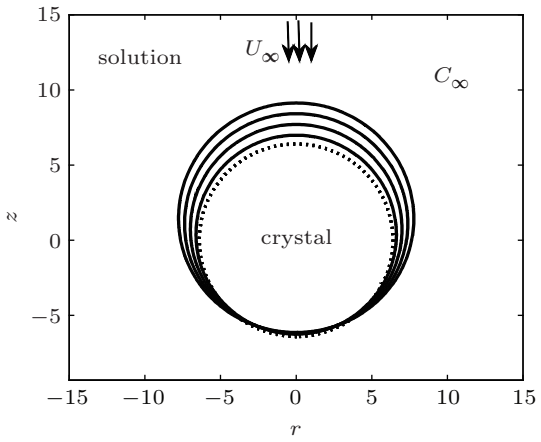


Fig. 1. Cross-section of the interface of the particle at time $t = 20$ for different values of the far-field uniform flow $U_\infty = 0.0, 0.20, 0.40, 0.60, 0.80$ (from inside to outside), with $\Gamma = 0.25$, $C_S = 1$, and $\varepsilon = 0.25$.

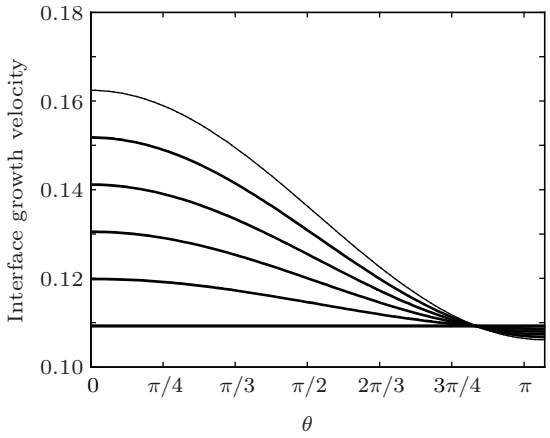


Fig. 2. Interface velocities at time $t = 20$ for different values of the far-field uniform flow $U_\infty = 0, 0.10, 0.20, 0.30, 0.40, 0.50$ (from down to up at the left side), with $\Gamma = 0.25$, $C_S = 1$, and $\varepsilon = 0.25$.

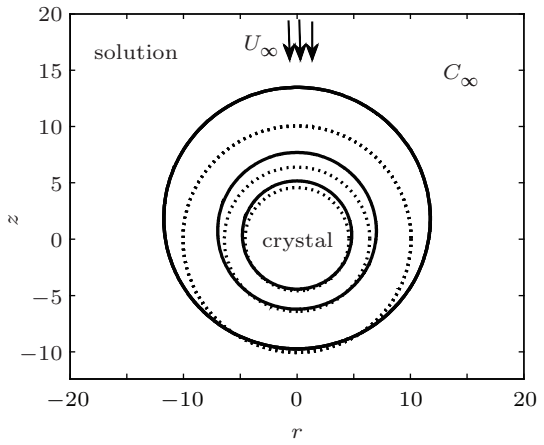


Fig. 3. Interface shapes of the particle at different times $t = 10, 30, 50$ (from down to up), where the nearest dashed lines are respectively the interface shapes without the far-field uniform flow, with $U_\infty = 0.40$, $\Gamma = 0.25$, $C_S = 1$, and $\varepsilon = 0.25$.

4. Conclusion

We have obtained the asymptotic solution for the interface and concentration field of particle growth in a supersaturated solution affected by a far-field uniform flow by using the method of matched asymptotic expansion. The analytical solution shows that the far-field uniform flow facilitates the growth of the spherical particle and the interface morphology protrudes into the solution. The upstream flow imposed on the particle enhances the growth velocity of the interface when the flow comes in, and the downstream flow lowers the growth velocity of the surface when the flow goes out. The interface morphology evolves into a peach-like shape. The pattern formations of the spherical particle under different far-field uniform flows obey the same mechanism as those in the convective pure melt and alloy.[16–20]

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