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Thermal Analysis on Planar Interface Stability in Solidification of Semitransparent Materials

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Significant melt undercooling may be developed in the melt in front of the solid/liquid interface during solidification of semitransparent materials because of internal radiative heat transfer with the environment. A nonequilibrium planar interface solidification model has been developed recently to permit the melt undercooling near the interface. A thermal analysis is presented for the stability of such a planar interface. An absolute stability theory derived by Ludwig for opaque materials has been employed as a first approximation. The stability theory takes into account the stabilizing effect of efficient cooling through the solid layer and defines a heat flux parameter to quantify the latent heat released that is transferred into the undercooled melt. When the variation of the heat flux parameter during solidification is calculated based on the present nonequilibrium planar interface model, the stability of the interface for given process conditions can be determined. The results suggest that, although the internal radiation leads to an undercooled melt in front of the interface, a planar interface can still be stable if there is strong external heat transfer. Based on this analysis, the physical mechanisms of the mushy-zone formation for the solidification of semitransparent materials have been presented.

Nomenclature

a = ratio of thermal diffusivities of liquid and solid
b = ratio of thermal conductivities of liquid and solid
c_p = specific heat, $J \cdot kg^{-1} \cdot K^{-1}$
D = thickness of the slab, m
G = temperature gradient on the liquid side of the interface, $K \cdot m^{-1}$
H = dimensionless enthalpy, $(h - c_p T_m)/c_p T_m$
H_R = convection–radiation parameter, $h_c/(\sigma T_m^3)$
h = enthalpy, $J \cdot kg^{-1}$
h_c = convective heat transfer coefficient, $W \cdot m^{-2} \cdot K^{-1}$
k = thermal conductivity, $W \cdot m^{-1} \cdot K^{-1}$
N = conduction–radiation parameter, $k/(4\sigma T_m^3 D)$
n = refractive index
q_r = radiative heat flux, $W \cdot m^{-2}$
$\bar{q}_r$ = dimensionless radiative heat flux, $q_r/(4\sigma T_m^4)$
S = dimensionless interface position, s/D
Ste = Stefan number, $c_p T_m/\lambda$
s = interface position, m
s_T = stability coefficient defined by Eq. (6)
T = absolute temperature, K
T_e = temperature of the environment, K
T_m = equilibrium freezing temperature, K
T_N = nucleation temperature, K
T_s = interface temperature, K
T_0 = initial temperature, K
t = time, s
V = interface velocity, $m \cdot s^{-1}$
V_a = absolute stability velocity, $m \cdot s^{-1}$
X = dimensionless coordinate, x/D
x = coordinate in direction across the slab, m
Y = dimensionless wave number of perturbation
α = thermal diffusivity, $m^2 \cdot s^{-1}$
β = extinction coefficient of the medium, m^{-1}
Γ = Gibbs–Thomson coefficient
η = heat flux parameter, Eq. (7)
θ = dimensionless temperature, T/T_m
θ_e = dimensionless environment temperature, T_e/T_m
θ_s = dimensionless interface temperature, T_s/T_m
κ_D = optical thickness of the layer, βD
λ = latent heat of fusion, $J \cdot kg^{-1}$
μ = dimensionless linear kinetics coefficient, $\rho c \mu_k/(4\sigma T_m^2)$
μ_k = linear kinetics coefficient, $m \cdot s^{-1} \cdot K^{-1}$
ρ = density, $kg \cdot m^{-3}$
σ = Stefan–Boltzmann constant, $5.6705 \times 10^{-8}\ W \cdot m^{-2} \cdot K^{-4}$
τ = dimensionless time, $(4\sigma T_m^3/\rho c_p D)t$
ω = single scattering albedo

Introduction

MELTING and solidification of semitransparent materials have wide engineering applications in the areas such as the design of certain latent heat-of-fusion thermal-storage systems,[1] crystal growth,[2] and laser processing of semiconductors and ceramics.[3,4] Because of volumetric radiation and absorbing of energy, phase change of semitransparent materials behaves very differently from that of opaque ones.

Most early efforts on modeling phase change of semitransparent materials focused on the mathematical treatment and the effects of introducing internal radiation (absorbing, emitting, and scattering) into a phase change model. Little attention was paid to the effects of internal radiation on the fundamental phase change characteristics.[5] It was assumed that there is a distinct solid/liquid interface separating the solid and liquid regions. Furthermore, a local thermodynamic equilibrium condition at the interface was employed, and the interface temperature is fixed at the equilibrium melting temperature.[6–9] Such models, however, result in significant solid overheating during melting or melt undercooling during solidification, although solid overheating and melt undercooling are inconsistent with the local equilibrium condition.

Chan et al.[5] postulated an isothermal mushy zone model to solve this paradox. Instead of a distinct solid/liquid interface, they proposed that there is a mushy zone that separates the pure solid and pure liquid regions. Within the mushy zone, the solid and liquid coexist, but their volume fractions may vary across the zone. Because no solid overheating or melt undercooling is allowed under

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an equilibrium condition, the entire mushy zone will be at the equilibrium melting temperature for pure materials. This model seems to work well for many cases and has been employed by various researchers for different problems.[10–12] The existence of a mushy zone in the melting of semitransparent materials is supported by an earlier observation of liquid pockets in ice exposed to light.[13] Recently, the present authors performed a systematic study of such an isothermal mushy zone model for pure semitransparent materials including internal emitting, absorbing, and scattering phenomena.[14]

The isothermal mushy zone model of Chan et al.[5] is an equilibrium model of phase change of semitransparent materials derived from pure thermodynamic considerations. The model would be valid as long as the rate of heat transfer is low and the speed of phase change is slow, so that the local equilibrium condition could be satisfied during the process. If the rate of heat transfer is high and a high interface speed results, the interface may deviate from local equilibrium, and the effect of phase change kinetics then becomes important. In addition, the isothermal mushy zone model provides no information about the geometric configuration or the morphology of the mushy zone. Accordingly, the present authors[15] have introduced melt undercooling and nonequilibrium kinetics of crystalline growth into the solidification model for semitransparent materials. The new model shows that, by permitting melt undercooling, a distinct solid/liquid interface may exist between the solid and liquid regions. The model calculations do show, however, that there is significant melt undercooling in front of the moving solid/liquid interface, due to internal radiation. Such melt undercooling may lead to the instability of the growing interface and the development of thermal dendrites, that is, a mushy zone.

In this paper, the solidification process of a semitransparent slab is formulated using the nonequilibrium planar interface model. The stability of the planar interface is examined, and the possibility of developing into a quasi-isothermal mushy zone is investigated. Based on the numerical results obtained, the physical mechanisms of mushy zone formation in a semitransparent material are discussed.

Mathematical Formulation of Nonequilibrium Solidification in a Semitransparent Slab

One-dimensional geometry, as shown in Fig. 1, is chosen for the sake of simplicity. The slab is initially at a uniform temperature T_0, which is above the freezing point of the material. The slab is made of a gray material that is emitting, absorbing, and isotropic scattering. The liquid slab, as well as the solid after solidification starts, will transfer heat directly into the environment through radiation. The slab is also cooled by external convection on the surface. The properties are independent of both temperature and phase. The energy transfer equation in the slab can be written as (e.g., see Chan and Hsu[16])

$$\frac{\partial H}{\partial \tau} = N\frac{\partial^2 \theta}{\partial X^2} - \nabla \cdot \bar{q}_r \qquad (1)$$

The radiative transfer equation to determine $\bar{q}_r$ for this geometry can be found elsewhere[14,15,17] and will not be repeated here. The enthalpy vs temperature relationship is given by

$$\theta = \begin{cases} H + 1 & \text{solid region,} \quad H < 0 \\ H + 1 - 1/Ste & \text{liquid region,} \quad H > 1/Ste \end{cases} \qquad (2)$$

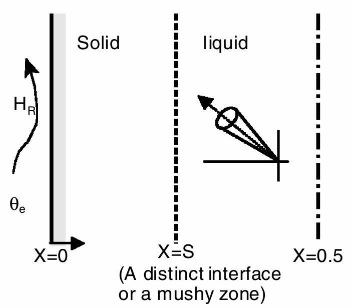

Fig. 1 Geometry and coordinates for a solidifying semitransparent slab.

Table 1 Numerical values of the parameters used

Parameters and symbols	Values
Ratios a and b in Eq. (6)	1
Kinetics coefficient μ in Eq. (4)	5
Refractive index, n	1.5
Conduction–radiation parameter, N	0.1
Convection–radiation parameter, H_R	0–2
Optical thickness, κ_D	1–20
Scattering albedo, ω	0–0.9
Stefan number, Ste	2
Initial temperature, T_0/T_m	1.05
Nucleation temperature, T_N/T_m	0.95
Environment temperature, $\theta_e = T_e/T_m$	0

The boundary conditions for Eq. (1) at $X = 0$ can be written as[15]

$$4N\frac{\partial \theta}{\partial X} = H_R(\theta - \theta_e) \qquad (3a)$$

and at $X = 0.5$,

$$\frac{\partial \theta}{\partial X} = 0 \qquad (3b)$$

Because of symmetry, only one-half of the slab needs to be analyzed with a symmetric condition at the centerline. Without loss of generality, the ambient temperature is assumed to be the same as the environment temperature.

A distinct planar solid/liquid interface is assumed to exist that separates the solid and liquid regions. Equation (1) is the governing energy equation for both solid and liquid phases with the corresponding physical properties. The melt undercooling is introduced at the interface, and the location of the interface is continuously tracked from the beginning of solidification based on the solidification kinetic relationship. As a first approximation, a linear crystalline growth kinetics model is adopted[18]:

$$V = \frac{dS}{d\tau} = \mu(1 - \theta_s) \qquad (4)$$

The linear kinetics coefficient of materials can be evaluated from solidification kinetics theories.[19]

In such a nonequilibrium model, solidification is assumed to be initiated at the surface of the slab at a prescribed nucleation temperature T_N that is below the equilibrium melting temperature of the material. Once the solidification begins, a planar interface is assumed to grow into the melt that may be undercooled. We assume a priori that the interface is stable, and the stability of the interface is then tested by comparing the interface velocity to the absolute stability velocity.

The detailed numerical solution procedures are omitted here to conserve the space. See Yao et al.[15] and Yao and Chung[17] for details on the solution of the radiative transfer equation[17] and the tracking of the interface.[15] The solidification of semitransparent materials depends on many parameters, even for such a simple geometry. It is not the intention of this paper to present a full-scale parametric study. Instead, only the effects of those parameters that are closely related to the morphology of the interface will be investigated. The parameters of particular interest include the optical thickness κ_D and the convection–radiation parameter H_R. The ranges of these two parameters are chosen from Siegel[20] and are given in Table 1.

Linear Stability Theory for a Planar Solidification Interface

It is well known that a planar interface may become unstable and develop into cellular or dendritic morphology. Extensive literature exists for interface stability analyses, and various criteria have been developed.[21–25] It is found that, for a planar interface growing into a melt, the instability of the interface is caused primarily by solute partitioning (for alloys) and a negative temperature gradient in the melt.[22,23] For pure materials, the only driving force for interface instability is the negative temperature gradient due to the melt undercooling ahead of the interface. In other words, if the temperature gradient in the melt is positive, the planar interface will always be stable; if the temperature gradient becomes negative, that is, the latent heat is transferred into the melt from the interface, the interface may become unstable. Analyses [21,23,24] have found that a planar solidification front may still be stable in such a case if the interface velocity satisfies the condition $V > V_a$, where V_a is the absolute stability velocity. For a pure opaque material without the constitutional effect, from a linear stability analysis,[21,24] the absolute stability velocity can be written as

$$V_a = s_T \alpha \lambda / (\Gamma c_p) \tag{5}$$

where the coefficient s_T is a function introduced by Ludwig to include the effect of cooling through the solid phase and is expressed by

$$s_T = \max\left\{\frac{4b}{Y^2}\left[\frac{\eta(-1+\sqrt{1+Y^2})+(\eta-1)(a+\sqrt{a^2+Y^2})}{-a+b+\sqrt{a^2+Y^2}+b\sqrt{1+Y^2}}\right]\right\}_{\eta,a,b} \tag{6}$$

where the maximum (max{ }) is taken over all values of Y (from zero to infinite). The heat flux parameter η, defined by Ludwig,[24] represents the fraction of latent heat released at the interface that flows into the undercooled melt,

$$\eta = -Gk/(V\lambda) \tag{7}$$

Because the temperature gradient G on the melt side is negative when the melt is undercooled, a minus sign is added in Eq. (7) to make η positive in such cases. For a given material, a and b are fixed, and thus, s_T is only a function of η. The value of η depends on heat transfer conditions and varies during solidification. Figure 2 shows typical variations of s_T with η for three values of equal a and b. The ratios of thermal properties of liquid and solid phases (a and b) have only a minor effect on s_T. Note that Fig. 2 is different from that given originally by Ludwig.[24] (It was found that Ludwig's original derivation for s_T has two errors that are corrected here.) Figure 2 shows that when η is smaller or equal to 0.5, s_T is zero. In other words, a planar interface growing into an undercooled melt is stable if more than one-half of the latent heat released at the interface could be transferred away from the solid side. Therefore, by examining the value of η during solidification, one can determine whether the planar interface is stable and whether a mushy zone could be formed. A smooth planar interface could be preserved even if the melt in front of the interface is undercooled, as long as η is small. Physically, this requires a high rate of external heat transfer so that a sufficient amount of latent heat released at the interface is transferred through the solid region. This is because an efficient cooling through a large positive temperature gradient in the solid compensates the destabilizing effect of a negative temperature gradient in the melt at the interface.

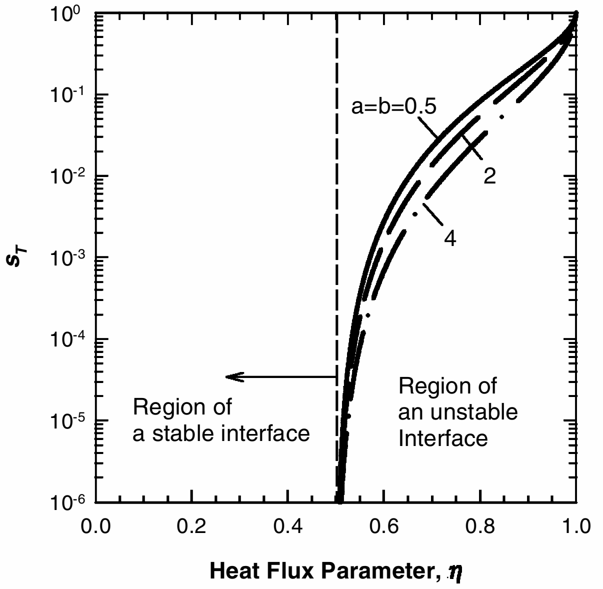

Fig. 2 Coefficient s_T in Eq. (6) as a function of heat flux parameter η for various values of a and b.

The preceding linear stability criteria, represented by Eqs. (5–7), were derived for opaque materials. It is found that the internal radiative heat transfer in a semitransparent material will affect the interface stability,[25] although no analytical relationship has been derived. As a first approximation, the preceding criterion will be used to calculate the absolute stability velocity for a semitransparent material. By doing this, one assumes that the main effect of the internal radiative heat transfer on the interface stability is through the temperature field, that is, through the heat flux parameter η. For example, changing the internal radiative properties will result in a variation of the interface heat transfer condition and, therefore, a different value of η. By monitoring the variation of the value of η throughout the solidification process, we can examine the stability of the planar interface. When the value of η at a given location becomes smaller than 0.5, the absolute stability velocity of the interface approaches zero, based on Eqs. (5) and (6), and the planar interface should be stable. Otherwise, for a large and positive value of η, the interface will become unstable, and a columnar dendritic structure would be developed. In the following discussion, therefore, the heat flux parameter η will be first obtained for any given radiative conditions by solving the one-dimensional planar interface model, as discussed in the preceding section. Then the stability of the planar interface is examined by comparing the interface velocity with the absolute stability velocity at the corresponding η. If the interface velocity is higher than the calculated stability velocity, then the planar interface is stable. Otherwise, the solidification should take place in a dendritic mode.

Numerical Results and Discussion

The solidification process of a semitransparent slab is determined using the preceding non-equilibrium planar interface model. Figure 3 shows typical temperature distributions at three time levels corresponding to the interface locations at $S = 0.05$, 0.25, and 0.45 for $\kappa_D = 5$, $N = 0.1$, $\omega = 0$, and $H_R = 1$. It can be seen that, at each time level, a temperature peak exists at the solid/liquid interface. The melt undercooling and the peaked temperature distribution around the interface result from internal radiative heat transfer. Note that the melt ahead of the interface is subject to radiative cooling during the entire solidification process due to internal radiative heat exchange

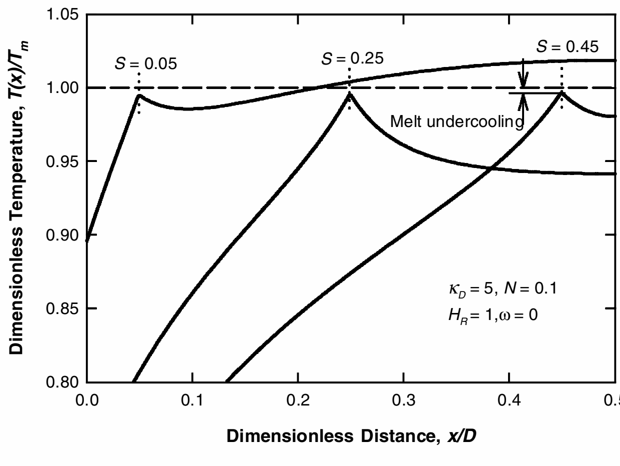

Fig. 3 Temperature distributions at the times when the interface is located at $S = 0.05$, 0.25, and 0.45 for $\kappa_D = 5$, $N = 0.1$, $\omega = 0$, and $H_R = 1$, predicted using the nonequilibrium planar interface model.

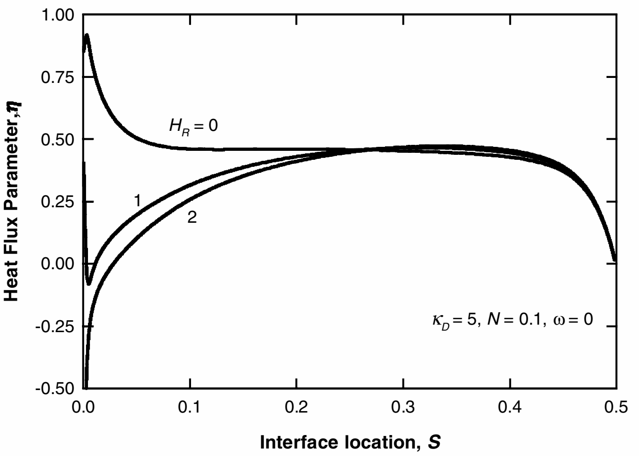

Fig. 4 Heat flux parameter as a function of the interface location for $\kappa_D = 5$, $\omega = 0$, and $N = 0.1$ under various external convective conditions.

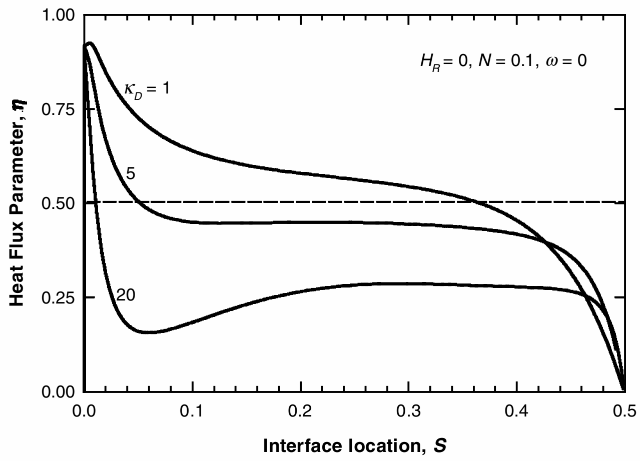

Fig. 5 Heat flux parameter vs interface location with optical thickness as a parameter for pure radiative cooling with $N = 0.1$, $\omega = 0$, and $H_R = 0$.

between the melt and the solid and between the melt and the environment. The latent heat released at the interface is carried out not only from the solid region through both radiation and heat conduction, but also from the undercooled melt by internal radiation. The latter results in a negative temperature gradient on the melt side of the interface. Because the latent heat goes into both the solid and liquid regions, the flux parameter η is larger than zero.

As mentioned earlier, a negative temperature gradient in the melt right in front of the interface may suggest an unstable interface, depending on the magnitude of the flux parameter, η. Figure 4 shows the variation of η during the solidification process of the same semitransparent slab shown in Fig. 3, but under three different external convection conditions: $H_R = 0$, 1, and 2. It can be seen from Fig. 4 that, for $H_R = 1$ and 2, the values of the heat flux parameter are always less than 0.5, which implies that a stable planar interface exists according to Fig. 2 and Eq. (5). When external convection is stronger, for example, $H_R = 1$ and 2, η is negative in the early stage of the solidification process, as a result of a positive temperature gradient at the interface on the melt side. However, this temperature gradient becomes negative when the interface advances farther from the surface. In the absence of external convection $H_R = 0$, Fig. 4 shows that η is large at the beginning but decreases quickly to a value below 0.5 as the interface moves forward. Therefore, in this case, the planar interface is unstable during the early period of time after the onset of solidification, and a mushy zone made of columnar thermal dendrites would be formed at this stage. As solidification takes place, however, the mushy zone may disappear, and the planar interface is stabilized in the later stage of solidification.

The effect of the kinetics coefficient μ has also been examined, and it is found that increasing kinetic coefficient μ from 5 to 100 raises the values of η slightly in the early stage of solidification and that it does not affect the overall trend regarding the stability of the planar interface. Therefore, in the following discussion, a fixed value of kinetics coefficient, that is, $\mu = 5$, will be used.

Effects of the optical thickness on the stability of the interface are illustrated in Fig. 5, which shows the variation of the heat flux parameter as a function of the interface location for three values of optical thickness $\kappa_D = 1$, 5, and 20. Only the results for $H_R = 0$ are shown because the interface is most likely unstable in this case, as discussed earlier. It can be found that the unstable region is enlarged when the optical thickness is decreased. For example, for $\eta > 0.5$, the interface has to pass, approximately, $X = 0.01$, 0.05, and 0.36 for the optical thick nesses $\kappa_D = 20$, 5, and 1, respectively.

Figure 6 shows the effect of the levels of isotropic scattering on the variation of the heat flux parameter, η during solidification for both $\kappa_D = 5$ and $\kappa_D = 20$. The convection coefficient H_R is again set equal to zero, and the conduction parameter is 0.1. Figure 6 shows a strong destabilizing effect of scattering on interface stability, in particular for the case with small optical thickness. For example, in the case of $\kappa_D = 5$ without scattering, η is larger than 0.5 for less than

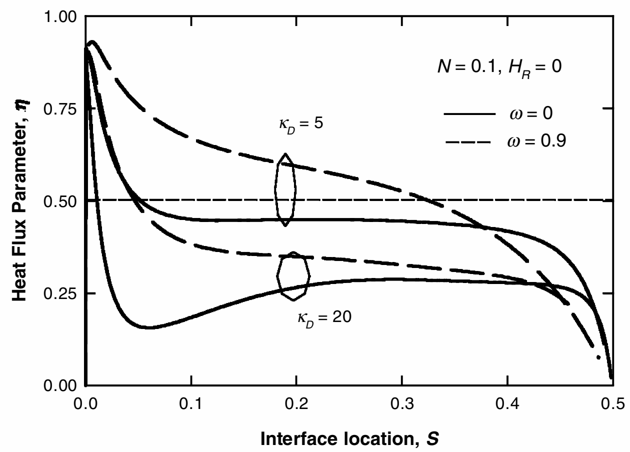

Fig. 6 Heat flux parameter vs interface location with $\omega = 0.9$ and $\omega = 0$ (without scattering). $\kappa_D = 5$ and 20, $N = 0.1$, and $H_R = 0$.

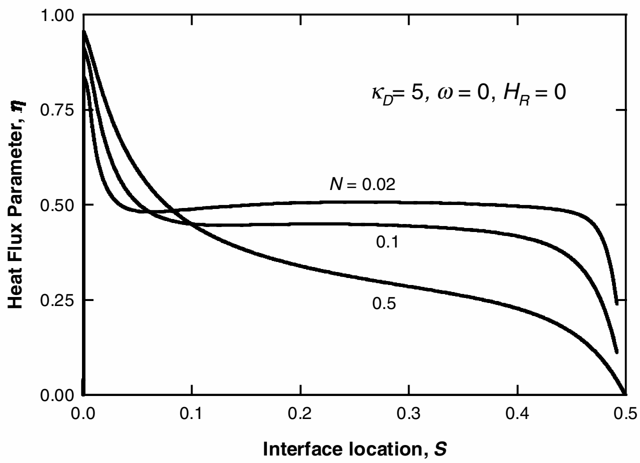

Fig. 7 Heat flux parameter vs interface location for three conduction–radiation parameter $N = 0.002, 2, 0.1$, and 0.5. $\kappa_D = 5$, $\omega = 0$, and $H_R = 0$.

10% of the thickness. When scattering is introduced with $\omega = 0.9$, η becomes larger than 0.5 for more than 60% of the slab. Therefore, the planar interface is less stable, and a mushy solidification front is more favored in the case of strong scattering.

Effect of the conduction–radiation parameter N on interface stability is illustrated in Fig. 7, which shows the variation of the heat flux parameter for $N = 0.02$, 0.1, and 0.5. Previous calculations[15] showed that reducing N leads to a more sharply peaked temperature distribution at the interface, which suggests that reducing N leads to a less stable interface. This is true for the later stages of solidification, as shown in Fig. 7. When the solid layer becomes thicker than about 0.1, reducing N leads to an increased heat flux parameter η, that is, a less stable interface. Surprisingly, at the very beginning of the solidification process, the effect of N on η is reversed, and a smaller value of η is obtained for a smaller N. As one can see in Fig. 7, when the interface location is less than 0.1, the values of η for $N = 0.5$ is much larger than that for $N = 0.1$ and $N = 0.02$, which indicates that a larger N leads to an instable planar interface at the beginning of the process. Such an inverse effect of N on the interface stability in different time periods of solidification can be understood by inspecting the definition of the heat flux parameter η [Eq. (7)], which shows that $\eta \propto Gk$. At the beginning of solidification, the temperature gradient G in the liquid due to radiation is relatively small, and η is primarily determined by the thermal conductivity k. Reducing N indicates a smaller k and, therefore, a smaller η, that is, a more stable planar interface. As solidification goes on, the internal radiation slowly builds up a larger temperature gradient in the melt in front of the interface, and η is then dominated by the temperature gradient term G. Under such conditions, reducing N leads to a large value of G and, therefore, a larger η, which results in a less stable interface.

Further Discussion on Physical Mechanisms of Mushy Zone Formation

A conclusion that can be drawn from the aforementioned results is that solidification of a semitransparent material may take place with a stable planar interface; even the melt in front of the interface is undercooled. No mushy zone will form under such conditions. In solidification terminology, the mushy zone is referred to as the area where the crystalline solid coexists with the liquid during solidification.[26] Two different geometric morphologies of the mushy zone can be observed, depending on the initial crystalline nucleation conditions. A widely spread mushy zone will be formed when individual nuclei are distributed in the undercooled melt and grow into grains, as shown schematically in Fig. 8a. This kind of solidification process is also called equiaxed growth. Equiaxed solidification may also take place if catalytic agents such as impurities exist in the bulk melt.[27] In equiaxed solidification, the melt between the crystals is undercooled, and the degree of undercooling depends on crystalline growth kinetics and the rate of external heat transfer. In many cases, however, the undercooling is very small, and a local equilibrium condition can be assumed.

Under a directional solidification condition where a positive temperature gradient exists across the melt, crystalline phase may nucleate on the surface instead of in the bulk melt. In this case, solid grows from the surface into the bulk melt opposite to the direction of heat flow. If the solidification velocity is slow, as in most of single crystal growth processes,[28] a planar solid/liquid interface appears, separating the solid and liquid regions, and no mushy zone is formed. For alloy systems at a moderate solidification velocity, however, constitutional undercooling will develop in front of the interface, which eventually leads to a cellular or dendritic solidification, that is, formation of a mushy zone made of columnar cells or dendrites. For pure materials, if the temperature gradient in the liquid is positive, that is, the melt is superheated, the planar solid/liquid interface is always stable. If the melt in front of the interface is undercooled, then a negative temperature gradient develops there, and the interface may become unstable and develop into thermal cells or dendrites, which leads to a different type of mushy zone shown in Fig. 8b.

For semitransparent materials, significant melt undercooling may develop before crystalline nucleation because of volume cooling by internal radiation.[15] If strong catalytic agents are distributed in the melt, bulk nucleation may take place with little melt undercooling, which results in equiaxed solidification with a quasi-isothermal mushy zone. Such a solidification process can, therefore, be described fairly accurately by the isothermal mushy zone model proposed by Chan et al.[5] For many cases in solidification, however, walls or surfaces in the system also act as nucleation agents, and

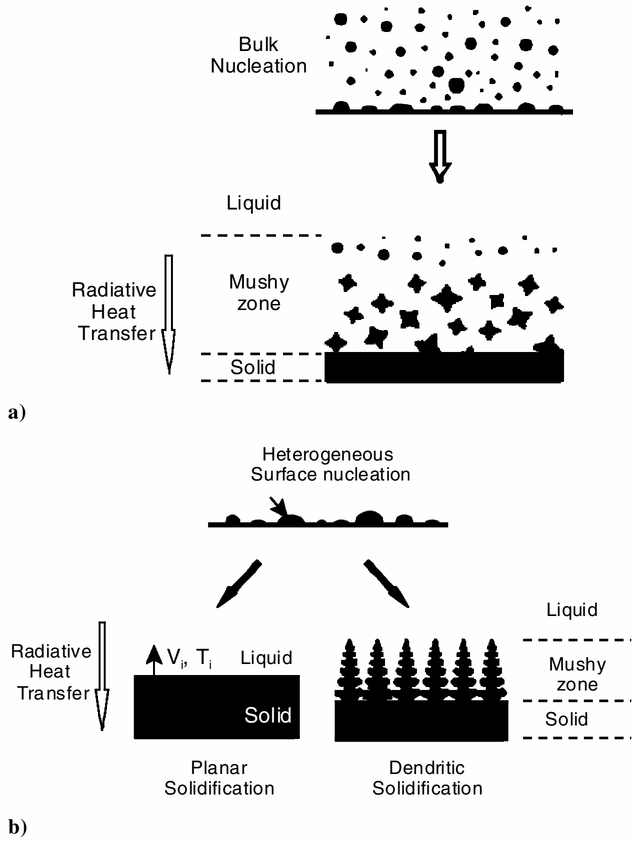

Fig. 8 Solidification, where corresponding mushy zones have two different geometric configurations: a) equiaxed solidification due to bulk nucleation in the melt and b) columnar solidification due to heterogeneous surface nucleation.

crystalline phase will nucleate on these surfaces. If heat is transferred out of the system through these surfaces, the directional solidification condition is satisfied. In this case, a stable planar interface may develop for pure substances, and the mushy zone will develop for multicomponent systems when solute partitioning causes interface instability. For pure semitransparent materials, the negative temperature gradient ahead of the interface due to radiative supercooling may also lead to an unstable interface and the development of thermal cells or dendrites as discussed earlier.

Conclusions

A nonequilibrium planar interface model is employed to analyze the stability of the planar interface during solidification of a one-dimensional semitransparent slab subject to both internal radiative cooling to the cold environment and convective cooling at the surface. The stability of the interface is examined against a linear stability criterion that takes into account the stabilizing effect of a positive temperature gradient in the solid. The results suggest that, although internal radiation leads to an undercooled melt in front of the interface, a planar interface can still be stable if there is strong external heat transfer. Both reducing the optical thickness and increasing internal scattering destabilize the interface. Increasing the conduction–radiation parameter destabilizes the interface in the early stage of solidification but becomes a stabilizing effect in the later stage of solidification. An unstable interface may eventually develop into a columnar dendritic mushy zone, which may be modeled by the isothermal mushy zone model developed by Chan et al.[5]

Acknowledgments

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