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# A Solute Diffusion Model for Micro-Macroscopic Analysis of Columnar Dendritic Alloy Solidification

**Hoseon Yoo* and Charn-Jung Kim****

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A new solute diffusion model for columnar dendritic alloy solidification based on a two-phase expanding volume element is developed to refine the existing multiphase model. In particular, individual effect of dendrite arm coarsening and back diffusion is incorporated in the model equation separately from each other. This formulation not only leads to successful identification of the behind-the-scene cause of the uncertainty associated with the coarsening inherent in the multiphase model, but also yields more accurate prediction compared with available experimental and numerical data. Taking advantage of the analytical solution for the diffusion-controlled limit, the approximation of the parabolic concentration profile commonly adopted in solidification modelings is also justified legitimately. A sample calculation in the absence of back diffusion clarifies the effect of coarsening on the evolutions of the solid fraction along different cooling paths, in that the eutectic fraction with coarsening is always smaller than that without it, and a rapid cooling during the early stage of dendritic solidification is favorable for homogenizing the final composition of alloy microstructures. In addition to the capability of resolving fundamental solute transport mechanisms, the compactness and consistency retained in the formulation make the present model suitable for use in the coupled micro-macroscopic analysis of dendritic alloy solidification as a microscopic component.

**Key Words:** Solute Diffusion Model, Micro-Macroscopic Analysis, Multiphase Model, Back Diffusion, Dendrite Arm Coarsening or Ripening

## Nomenclature

$C$ : Concentration
$\bar{C}$ : Average concentration
$D$ : Mass diffusivity
$f$ : Mass fraction
$g$ : Volume fraction
$k_p$ : Equilibrium partition coefficient
$l$ : Diffusion length
$n$ : Coarsening exponent
$S$ : Interfacial area concentration
$s$ : Position of the solid-liquid interface
$T$ : Temperature
$t$ : Time
$X$ : Size of the expanding domain
$x$ : Coordinate

### Greek Characters

$\lambda_1$ : Primary dendrite arm spacing
$\lambda_2$ : Secondary dendrite arm spacing
$\rho$ : Density

### Subscripts

O : Initial state
eut : Eutectic
$f$ : End of solidification
$i$ : Interface
$l$ : Liquid phase
liq : Liquidus

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* Professor, Department of Mechanical Engineering, Soong Sil University, Seoul 156-743, KOREA

** Senior Research Scientist, Division of Mechanical/Control Systems, Korea Institute of Science and Technology (KIST), Seoul 136-791, KOREA

$s$ : Solid phase

## 1. Introduction

Modeling of transport phenomena occurring during dendritic alloy solidification has attracted considerable research attention in the past several decades. A variety of theoretical-numerical models have been developed to predict the fundamental characteristics of the alloy solidification such as microstructure formation, solute redistribution, cooling curve, channel and freckle formation, shrinkage, and so on (Rappaz, 1989; Viskanta, 1990). One of the most challenging problems involved in solidification modeling is the complex interaction between physical phenomena which take place on different length scales ranging from the grain size to the system level, as reviewed by Beckermann and Viskanta (1993).

Recently, a promising approach termed micro-macroscopic modeling has been proposed (Rappaz, 1989; Rappaz and Voller, 1990; Voller, 1993), in which microscopic phenomena such as back diffusion, dendrite arm coarsening, nucleation and undercooling are incorporated into macroscopic heat flow calculations in order to capture the microstructural properties of a solidifying alloy on the system scale. A key component in this approach is the solute diffusion model which provides the evolution of the local solid fraction for use in the macroscopic conservation equations, and thus plays the role of a linkage between the microscopic and macroscopic aspects of dendritic solidification (Voller, 1993). Since the solute diffusion process during columnar dendritic growth is influenced primarily by back diffusion, dendrite arm coarsening and dendrite tip undercooling (Kurz and Fisher, 1989), research efforts have been focusing on how to properly account for these effects in modeling. The sole effect of back diffusion in the solid has been well understood and incorporated in the solute diffusion model since the early studies, yielding not only the well-known lever rule and the Scheil equation but also some analytical and semi-analytical solutions such as those by Brody and Flemings (1966), Clyne and Kurz (1981), Ohnaka (1986), and Kobayashi (1988). A number of numerical models which can handle the combined effects of back diffusion and dendrite arm coarsening have also been developed (Roosz et al., 1986; Ogilvy and Kirkwood, 1987; Battle and Pehlke, 1990; Sundarraj and Voller, 1993) since the dominant role of coarsening during the solute redistribution was verified. On the other hand, the dendrite tip undercooling at the columnar front has not received research attention so much as back diffusion and/or dendrite arm coarsening, because its effect becomes significant under relatively restricted conditions, e. g., during rapid solidification processes (Giovanola and Kurz, 1990).

More recently, Beckermann and coworkers have developed the so-called multiphase micro-macroscopic model using the volume averaging technique, which, they claimed, could effectively incorporate nearly all of the microscopic mechanisms present in dendritic solidification on different length scales including nucleation, growth kinetics and dendrite morphology. Basic concept, detailed derivation procedure and some typical results of the multiphase model can be found in references (Ni and Beckermann, 1990; Wang and Beckermann, 1993a), and thus are not repeated here. Depending on the extent of simplifications, various versions of the model equations have been derived (Wang and Beckermann, 1993a) and applied to selected cases of interest successfully (Schneider and Beckermann, 1995a; 1995b). They have also demonstrated that the model equations reduced to the lever rule and the Scheil equation for the limiting cases of back diffusion (Wang and Beckermann, 1993b). At present, the multiphase model seems to be one of the most flexible and sophisticated among diverse solidification models reported in the open literature. Nevertheless, careful investigation on the model equations still leads to a problem about incorporation of dendrite arm coarsening phenomena in the solute diffusion model. Specifically, the effect of coarsening is reflected in the solid-phase solute balance equation of the multiphase model only by the interfacial area concentration which constitutes the phase interaction term together with mass diffusion coefficient, diffusion length and interfacial concentration difference between the adjacent phases. The apparent problem is that the phase interaction term becomes zero when the back diffusion is absent. Therefore, in such circumstances, the coarsening which already proved to affect the solute diffusion process significantly is forced to be neglected in the analysis. However, neither the evidence of interrelation that the coarsening does not take place in the absence of back diffusion has been reported, nor another means except the interfacial area concentration by which the coarsening can be incorporated in the solute diffusion model are found. It is an established fact that the coarsening results from the local remelting of dendrite arms due to the curvature or surface energy effect (Flemings, 1974) and appears to be independent of back diffusion.

The aim of this work is not only to clarify the uncertainty associated with the coarsening in the solute balance equation of the multiphase model, but also to present an alternative solute diffusion model suitable for micro-macroscopic analysis of dendritic solidification in which the coarsening as well as the back diffusion is rigorously incorporated. In developing the new model, some simplifications are introduced within the extent of fundamental characteristics of the microscopic transport mechanisms being resolved. The developed model is discussed formally in comparison with the multiphase model first, and then is validated quantitatively by comparing the predicted results using it for a specific case with available data from the experiment as well as the numerical simulation. In addition, the parabolic concentration profile in the solid which has been commonly adopted in the solute diffusion modelings (Ohnaka, 1986; Ni and Beckermann, 1990) is reexamined to confirm its validity. Finally, the effect of coarsening on the evolution of the local solid fraction along different cooling paths during dendritic solidification is investigated as an application of the present model.

## 2. Problem Definition

In order to specify the problem involved in the multiphase model, the solute balance equation in the solid needs to be considered. Before doing it, the basic features of the multiphase approach are briefly addressed here to understand its framework. The multiphase model is essentially intended to account for the different microscopic length scales of physical significance existing in a dendritic structure as many as possible in the macroscopic calculations. According to Wang and Beckermann (1993a), this has been accomplished by considering not only the physical solid and liquid phases but also phases associated with different length scales such as the interdendritic and extradendritic liquids separately and averaging the field variables of each phase over an appropriate-sized volume element. The macroscopic (or volume averaged) conservation equations for each phase are derived from the microscopic counterparts in such a way that the phase interaction term resulting from the volume averaging process is expressed as a function of the characteristic lengths of each phase such as the interfacial area concentration and the diffusion length. The characteristic lengths of each phase can be specified by the topology of the microstructure concerned.

Now, restricting our attention to the solute diffusion in the solid phase under consideration, the volume-averaged solute balance equation for a typical control (or averaging) volume shown in Fig. 1(a) is expressed, in the present notation, as

$$\frac{d}{dt}\left(\rho_s g_s \bar{C}_s\right) = \rho_s C_{si}\frac{dg_s}{dt} + \frac{\rho_s D_s S_s}{l_s}\left(C_{si} - \bar{C}_s\right) \quad (1)$$

where $S_s$ and $l_s$ denote the interfacial area concentration, i. e. the solid-liquid interfacial area per unit volume, and the diffusion length the definition of which can be found in Wang and Beckermann (1993a), respectively. The interfacial solid concentration $C_{si}$ is related to the liquid concentration, under the local equilibrium at the interface, as $C_{si} = k_p C_l$. In deriving Eq. (1), solute transport due to either diffusion or convec-

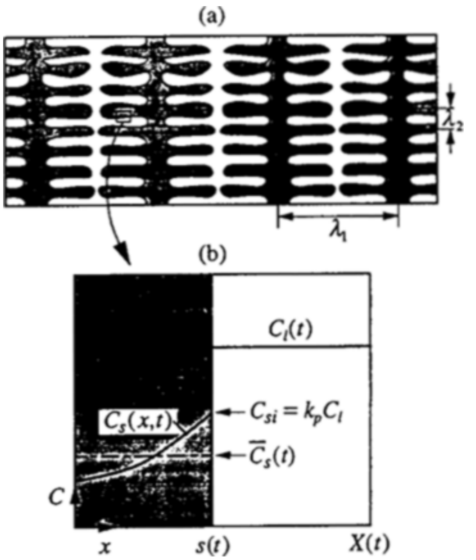

**Fig. 1** Representative volume elements used for describing (a) the macroscopic, and (b) the microscopic solidification characteristics.

tion on the macroscopic scale has been neglected. It is apparent in Eq. (1) that the interfacial area concentration $S_s$ is the only term capable of implying the effect of dendrite arm coarsening. However, note that, in the absence of back diffusion, i.e. $\rho_sD_sS_s/l_s=0$, the model equation by no means accounts for the coarsening, as mentioned in Introduction. Nevertheless, such an important problem has never been pointed out in the literature yet.

From the physical point of view, the dendrite arm coarsening is regarded as affecting the overall solute diffusion process in the following two ways. One is that it decreases the interfacial area concentration of an averaging volume, thereby making the volume-averaged diffusive flux across the interface (the back diffusion) smaller. The other is that it increases the secondary dendrite arm spacing $\lambda_2(t)$, which, in effect, brings forth the reduction of either the solid volume fraction $g_s$ at the same phase change rate or the average concentration in the solid phase $\bar{C}_s$. The former is anyhow reflected in the model by the second term on RHS of Eq. (1), whereas the latter is not apparently. Therefore, at a glance, the problem at issue may be caused by neglecting only the second effect of coarsening in the formulation of the multiphase model. However, the true source of the problem originates from the other fact, which will be clarified in detail later.

For the sake of the comparative discussion followed, let us specify $S_s$ and $l_s$ in terms of the microscopic characteristic lengths and simplify Eq. (1). Assuming the well-mixed interdendritic and extradendritic liquids (the separate handling of them is not directly relevant to the present problem), and defining $X(t)=\lambda_2(t)/2$, a simple one-dimensional platelike geometry of the secondary dendrite arms yields (Wang and Beckermann, 1993a)

$$S_s=1/X \tag{2}$$

For such a geometry with the parabolic concentration profile in the solid (Ohnaka, 1986), the diffusion length $l_s$ is readily related to the interface position $s$ as (Wang and Beckermann, 1993a)

$$l_s=s/3 \tag{3}$$

It is further assumed that the solid density $\rho_s$ is independent of time (but may depend on the concentration), which is valid for many dilute alloys of practical interest (Poirier et al., 1991). Then, Eq. (1) reduces to

$$\frac{d}{dt}\left(g_s\bar{C}_s\right)=C_{si}\frac{dg_s}{dt} + \frac{3D_s}{sX}\left(C_{si}-\bar{C}_s\right) \tag{4}$$

## 3. Modeling

It is desired that a new solute diffusion model not only accounts for the effect of dendrite arm coarsening completely, but also is suitable for coupling with the macroscopic heat flow calculations. To this end, introduced is the postulate that the solute diffusion characteristics of the macroscopic control volume can be represented, in an average sense, by those of a microscopic volume element the size of which is half a secondary dendrite arm spacing $\lambda_2(t)$ (see the box in Fig. 1 (a)). The volume element adopted here differs substantially from that for the microscopic conservation equations of each phase in the above-mentioned multiphase model in that the present one consists of not a single phase but two phases and expands with time. This type of volume element is not a new one but has been commonly used in the microsegregation modelings, e. g., by Battle and Pehlke (1990). Although it has not been Proved at this stage that the introduced postulate is physically realistic, few attempt has been made to facilitate the solute diffusion model based on it for use in the micro-macroscopic analysis (Voller, 1993).

Referring to the enlarged plot of the microscopic volume element in Fig. 1(b) and the simplifications adopted in deriving Eq. (4), the integral form of the solute balance in the solid phase is expressed as (Ohnaka, 1986; Kurz and Fisher, 1989)

$$\frac{d}{dt}\left(s\bar{C}_s\right)=C_{si}\frac{ds}{dt}+D_s\frac{\partial C_s(s,t)}{\partial x} \tag{5}$$

where $s(t)$ denotes the solid-liquid interface position. When the same parabolic concentration profile in the solid as in Eq. (4) is invoked, Eq. (5) takes the form of

$$\frac{d}{dt}\left(s\bar{C}_s\right)=C_{si}\frac{ds}{dt}+\frac{3D_s}{s}\left(C_{si}-\bar{C}_s\right) \tag{6}$$

Note that Eq. (6) has been derived under almost the same assumptions and is cast in the apparently similar form with Eq. (4) although the approaches differ from one another.

In order to examine the distinction between the multiphase and the present solute diffusion models and to pick up the origin of the problem described before formally in advance of the validation of the developed model, Eq. (6) is rearranged in terms of the solid volume fraction defined by (see Fig. 1(b))

$$g_s=s/X \tag{7}$$

Then, the resulting equation is expressed as

$$\frac{d}{dt}\left(g_s\bar{C}_s\right)=C_{si}\frac{dg_s}{dt}+\left[\frac{3D_s}{sX}+g_s\left(\frac{1}{X}\frac{dX}{dt}\right)\right](C_{si}-\bar{C}_s) \tag{8}$$

Although the only difference between Eqs. (4) and (8) is an additional term $\left(\frac{1}{X}\frac{dX}{dt}\right)$ in Eq. (8), the term, which can be interpreted physically as the expansion rate of a secondary dendrite arm spacing, manifests the problem associated with coarsening in the solute diffusion process. First of all, contrary to Eq. (4), Eq. (8) explicitly includes the effect of coarsening via the volume expansion rate term even in the absence of back diffusion. The more important point is that Eq. (8) coincides with Eq. (4) when the dendrite arm spacing is fixed, i.e.$X(t)=$const. This fact implies, in a theoretical sense, that the effect of coarsening was not partly, as described in conjunction with Eq. (1), but completely ignored in the multiphase model presuming that the present model is perfectly valid. In practice, however, it is not likely that the effect of coarsening have been completely neglected in the reported results by the multiphase model, not only because the time-dependent secondary dendrite arm spacings instead of the fixed ones have been used for the coefficient of the second term on RHS of Eq. (4) (Wang and Beckermann, 1993b), but also because the present model may not be a perfect one. These aspects can be estimated only by rigorous model validation. In Eq. (4), even when a more sophisticated geometric model for the secondary dendrite arm structure is adopted to specify $S_s$ and $l_s$ instead of Eqs. (2) and (3), only a numerical factor will be changed in the coefficient of the second term on RHS. As for the two effects of coarsening on the solute diffusion, the first and the second terms in the square bracket in Eq. (8) correspond to them in sequence. The above observations lead to the conclusion that the present model accounts for the effect of coarsening more properly than the multiphase model at least in a formal view, apart from the ultimate validity of the model equation itself. Also, it is obvious that Eq. (4) is merely a subset of Eq. (8). Meanwhile, it is meaningful to note that the framework of computational procedure developed for the multiphase model (e. g., Wang and Beckermann, 1993b) can be readily applied to the present case with only minor changes due to the addition of the volume expansion rate term.

For evaluating the solid volume fraction and the average solid concentration at the prescribed temperature with the aid of Eq. (7), the following overall solute balance for the two-phase expanding volume element and initial conditions are needed

$$\rho_s\frac{d}{dt}\left(g_s\bar{C}_s\right)+\rho_l\frac{d}{dt}\left(g_lC_l\right)=0 \tag{9}$$

$$g_s=0;\ \bar{C}_s=C_{si};\ C_l=C_0 \text{ at } t=0 \tag{10}$$

That is, Eqs. (8), (9) and (10) constitute a set of governing equations for the present solute diffusion model, where the macroscopic solute transport is assumed to be suppressed. Thermodynamic relations pertinent to the phase equilibrium, density dependency on the concentration and coarsening of the secondary dendrite arms should be supplemented for the closure of modeling. The solid mass fraction corresponding to the solid volume fraction is calculated by

$$f_s=\rho_sg_s/(\rho_sg_s+\rho_lg_l) \tag{11}$$

The eutectic fraction, which denotes the unsolidified liquid fraction when the temperature or the liquid attains the eutectic state, is one of the meaningful results obtainable in the analysis of solute diffusion problem. From the combination of Eqs. (8) and (9), it can be easily deduced that the liquid fraction decreases with increasing back diffusion and/or dendrite arm coarsening rate during dendritic solidification. This implies that the accumulated effects of primary parameters on the overall solute diffusion process can be represented by the eutectic fraction at the end of solidification (Sarreal and Abbaschian, 1985). Furthermore, the eutectic fraction is of practical importance in alloy solidification, because the solute concentration of the eutectic solid is much higher than that of the dendritic solid in the final product (Kurz and Fisher, 1989). Accordingly, it has been commonly used as an index of the solute redistribution.

## 4. Validation and Application

So far, a new solute diffusion model based on the two-phase expanding volume element has been developed and formally discussed on its properties in comparison with the solute balance equation of the multiphase model. Obviously, the present model appears to be better than the multiphase model as far as the dendrite arm coarsening is concerned. The task to be undertaken is the validation of the developed model, which is followed by an application.

The present model is validated by comparing the predicted eutectic volume fractions with available data for a specific case. Directional solidification of an Al-4.9% Cu alloy is selected for comparison, not only because experimental data for the wide range of the cooling rates have been reported by Sarreal and Abbaschian (1985), but also because a sophisticated numerical simulation has been performed (Sundarraj and Voller, 1993). Moreover, numerical data for the calculation including thermophysical properties, phase equilibrium diagram, density model, and coarsening model have been well established for this system, which are documented in the works of Sundarraj and Voller (1993), Yoo (1996a), and elsewhere.

Figure 2 shows the predicted eutectic volume fractions with respect to the cooling rate by both the present and the multiphase models in comparison with those from the experiment as well as the numerical simulation. In order to focus our attention on the dendrite arm coarsening at issue, the effect of dendrite tip undercooling which creates

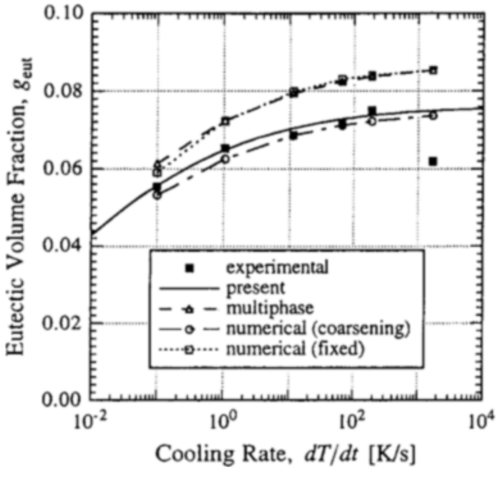

**Fig. 2** Comparison of the predicted eutectic volume fractions based on the present and the multiphase models with data from the experiment and numerical simulation for the directional solidification of an Al-Cu alloy.

another facit of the problem (see Flood and Hunt, 1987; Giovanola and Kurz, 1990; Wang and Beckermann, 1993b) has been excluded in all models. This, however, causes the common disagreement between the experiment and all predictions at high cooling rate where the dendrite tip undercooling predominates, in Fig. 2. Except such an extreme condition, the present predictions favorably agree with both of the experimental and the numerical data qualitatively and quantitatively over the wide range of the cooling rate. In contrast, the multiphase model, despite being based on almost the same simplifications, considerably overpredicts the experimental data, the present and numerical results. Since the only difference between the present and the multiphase models is the volume expansion rate term, the overprediction obviously originates from the improper incorporation of the coarsening phenomena. This fact is consistent with the close agreement between the multiphase model predictions and the numerical results for the case of fixed arm spacings. In consequence, the fundamental postulate introduced at the beginning of the present modeling can be said to hold from the physical reality. In regard to the computational efforts, only a set of ordinary differential equations, Eqs. (8) ~ (10), is solved in the present model, whereas partial differential equations should be analyzed in the numerical simulation (Sundarraj and Voller, 1993). This sort of facile character enables the present model to fit the micro-macroscopic analysis of alloy solidification as a microscopic component.

Recently, Yoo (1996b) has presented an analytical solution to the solute diffusion problem for a certain limiting case, named the diffusion-controlled limit, where the back diffusion is absent, the equilibrium partition coefficient is constant, and the densities of the solid and liquid are equal and constant. Under the last condition, the volume and mass fractions are identical, i.e. $f_s = g_s$. The solution is known to be

$$f_s = \frac{1}{1-k_p}\left(1-\frac{C_0}{C_l}\right) - \frac{k_p}{(1-k_p)^2}\frac{{C_l}^{-1/(1-k_p)}}{X(t)}\int_0^t X(t)\,{C_l}^{k_p/(1-k_p)} \times \left(1-\frac{C_0}{C_l}\right)\frac{dC_l}{dt}dt. \qquad (12)$$

Equation (12) reduces to the well-known Scheil equation,

$$f_s = 1-(C_0/C_l)^{1/(1-k_p)} \qquad (13)$$

when the dendrite arm spacing is fixed, i.e. $X(t) = \text{const}$. Noting that the Scheil equation has been derived directly from the multiphase model, Eqs. (4), (9) and (10), under the same conditions with Eq. (12) (Wang and Beckermann, 1993b), the improper incorporation of coarsening in the multiphase model becomes evident once again.

The parabolic concentration profile in the solid, a representative among the simplifications involved in solidification modeling, is worthy of rigorous verification, although it has been regarded as a commonly acceptable approximation since the work of Ohnaka (1986). Since Eq. (12) is completely free from the type of the concentration profile, the validity of the approximation can be estimated by comparing the present prediction for the diffusion-controlled limit with Eq. (12). The predicted timewise evolution of the microscopic interface position which is very sensitive to the variation of the solid concentration is compared with the analytical solution for $D_s = 0$, $k_p = 0.14$ and $\rho_s = \rho_l$ in Fig. 3. Although considered here is only a limiting case, the excellent agreement between them suffices to validate the approximation. The present verification which is

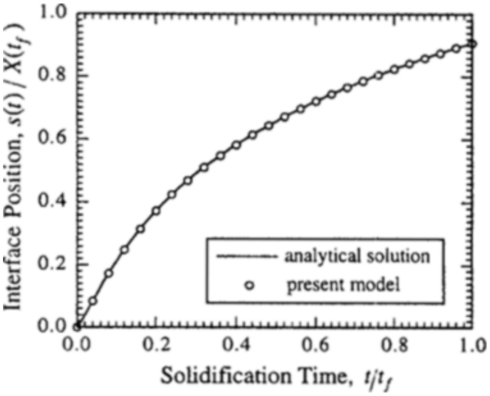

**Fig. 3** Comparison of the timewise variation of the interface position between the present model prediction and the analytical solution for the diffusion-controlled limit.

attempted for the first time would provide the theoretical background for using the parabolic concentration profile in solidification modelings.

In the presence of back diffusion, the solute diffusion process depends strongly on the local solidification time, i.e. the cooling rate without regard to the dendrite arm coarsening, thereby resulting in, for example, different eutectic fractions as shown in Fig. 2 (Sarreal and Abbaschian, 1985; Kurz and Fisher, 1989). On the other hand, in the absence of back diffusion, it has long been recognized that the solute redistribution is independent of the local solidification time, since the Scheil equation, which relates the local solid fraction to the liquid or interfacial concentration and provides the universal microsegregation curve (Kurz and Fisher, 1989), has been exclusively invoked to evaluate the results such as the eutectic fraction. The multiphase model must produce the same results, because it reduces to the Scheil equation for the diffusion-controlled limit. In reality, the above-mentioned fact in the absence of back diffusion is true only when the dendrite arm spacings are fixed. However, the coarsening always accompanies dendritic solidification. That is, the local solute redistribution may depend on the cooling path even for the prescribed solidification time.

As an application example of the present model, the timewise evolutions of the solid fraction along different cooling paths are investigated. In order to concentrate on the effect of dendrite arm coarsening and to prevent the results from being smeared by the other effects, calculations have been performed for zero back diffusion and for the same solidification time ($t_f = 100$ s for all cases under consideration) using the same numerical data as those for Fig. 2. Figures 4 and 5 depict the variations of the solid fraction during dendritic solidification with and without coarsening, respectively, each for three cooling patterns: square root, linear and quadratic temperature-time relations. Since the overall solidification behaviors are influenced essentially by temperature variation, similar trends between Figs. 4 and 5 appear to be reasonable. That is, considerable differences between the solid fraction curves corresponding to different cooling patterns at the same time are found in both of Figs. 4 and 5. From the physical point of view, in other words, considering that all the solute rejected from the solid phase due to the solubility difference piles up in the liquid phase in the absence of back diffusion, it is likely that the three cases with the same solidification time yield the same solid fractions at the same eutectic temperature or concentration. In practice, however, the foregoing statement proves to be true only when the dendrite

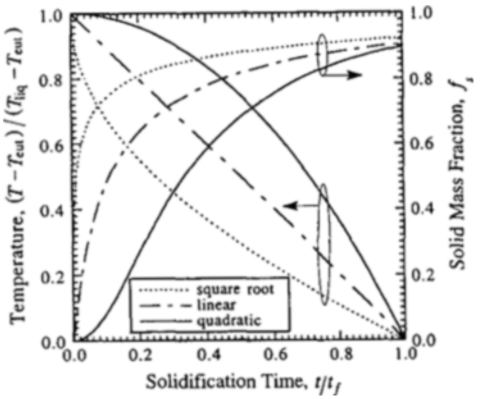

**Fig. 4** The evolutions of the solid mass fraction along different cooling paths during dendritic solidification with coarsening in the absence of back diffusion.

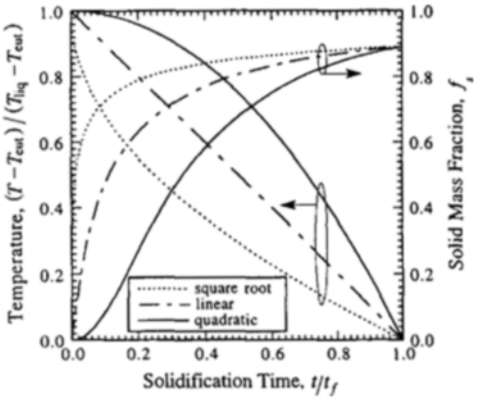

**Fig. 5** The evolutions of the solid mass fraction along different cooling paths during dendritic solidification without coarsening in the absence of back diffusion.

arm spacing is fixed, but not when the coarsening takes place. That is, at the end of solidification, the solid fraction of each case in Fig. 5 coincides with each other, whereas those in Fig. 4 are distinct from one another. The quantitative differences between the final solid fractions in Fig. 4 are relatively small but non-negligible, since they characterize the effect of coarsening on the dendritic growth in the absence of back diffusion. This argument can be illustrated more clearly by the microsegregation curve, i. e. the liquid concentration-solid mass fraction diagram. Figure 6 shows four microsegregation curves, three of which correspond to each case of Fig. 4, respectively, and the last one represents the case of fixed arm spacings. Here, it should be noted that all cases without coarsening, e. g., three cases in Fig. 5, turned out to reduce to the same curve regardless of the cooling pattern on the liquid concentration-solid mass fraction plane. This can be deduced *a priori* from the Scheil equation, since Eq. (13) expresses a unique relation between the solid fraction and the liquid concentration, and is independent of time.

Two observations can be made from Fig. 6. First, the effect of coarsening grows significant as solidification proceeds, resulting in the eutectic fraction always smaller than that for the fixed spacings at the final state. For the case of non-zero back diffusion, the qualitatively consistent results with this finding have already been presented in Fig. 2 (note the distinction existing between the present and the multiphase model predictions), although the quantitative difference in the eutectic fraction between with and without coarsening should be affected by the solidification time or the cooling rate. Second, imposing a rapid cooling rate during the early stage of dendritic solidification is an efficient method for reducing the final eutectic fraction under the same solidification time. According to the established coarsening models (Kirkwood, 1985; Roosz *et al.*, 1986; Battle and Pehlke, 1990; Mortensen, 1991), the dendrite arm spacing is approximately proportional to $t^{-n}$, where the exponent $n$ ranges approximately from 0.29 to 0.33. Since the liquid volume, i. e. the dendrite arm spacing, remains small during the early solidification, the rapid cooling in this period effectively decreases the actually solidified portion at relatively low concentration, in spite of high values of the solid fraction. Consequently, such a cooling pattern may contribute to reduce the microsegregation or to homogenize the final composition of the alloy microstructures. However, care should be taken to apply this method in the presence of back diffusion, because it also retards the back diffusion, thereby decreasing the solid concentration. The higher the average solid concentration, the smaller is the final eutectic fraction.

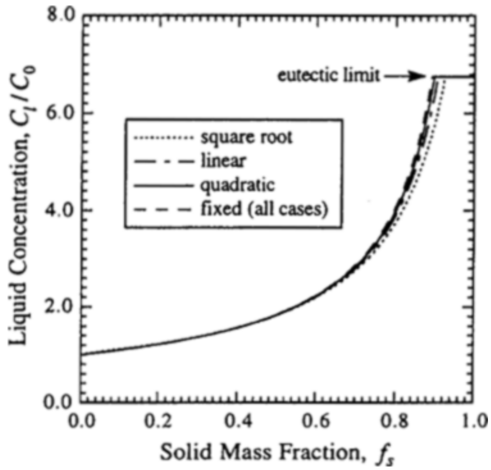

**Fig. 6** Microsegregation curves corresponding to three cooling paths in Fig. 4, and the case of fixed arm spacing in the absence of back diffusion.

Noting that the local solid fraction of the control volume predominates the heat transport mechanism due to the release of the latent heat of fusion in the macroscopic calculations, the coarsening which affects the local solid fraction-concentration (or temperature) relation even in the absence of back diffusion should be carefully taken into account in the micro-macroscopic analysis of dendritic alloy solidification. In this sense, the present solute diffusion model in which the dendrite arm coarsening as well as back diffusion is rigorously incorporated will be useful for determining proper values of the local solid fraction. The coupled micro-macroscopic analysis for a simple directional casting using the present model is under way, the results of which are to be presented soon.

## 5. Conclusions

Motivated by an overlooked uncertainty in the solute balance equation of the existing multiphase model, a new solute diffusion model for columnar dendritic alloy solidification has been developed in this work. The present model was primarily aimed at not only accounting for the key mechanisms pertinent to the solute redistribution on a microscopic scale rigorously, but also facilitating the micro-macroscopic analysis of dendritic solidification under the same level of simplifications with the existing models. The final results obtained throughout this work can be summarized as follows.

Based on a two-phase expanding volume element representing the coarsening secondary dendrite arm spacings and with the aid of the parabolic concentration profile in the solid phase, a compact-form model equation for the solute diffusion process has been successfully derived. The model equation was so formulated that individual effect of the dendrite arm coarsening and the back diffusion could be incorporated separately from each other. Compared with the multiphase model, the volume expansion rate term which characterizes the effect of coarsening is additionally included in the present equation. This formal comparison suffices to identify that the problem inherent in the multiphase model originates from the improper incorporation of coarsening possibly, during the volume averaging process or the formulation of constitutive relations. It can also be deduced from the model equation *per se* that the present model is more refined than the multiphase at least in describing the coarsening phenomenon.

The model validation has been accomplished by comparing the predicted eutectic volume fractions by both the present and the multiphase models with available data from the experiment as well as the sophisticated simulation for the well-known directional solidification of an Al-Cu alloy. While the present predictions agree closely with both the experimental and the numerical data over the wide range of the cooling rate, the multiphase model considerably overpredicts both of them. Rather, the results from the multiphase model nearly coincide with the simulated data for the fixed arm spacings. It has been further confirmed that these behaviors persist for the diffusion-controlled limit. Under such circumstances, the multiphase model yields the Scheil equation directly, whereas the analytical solution which has relied on the same approach with this work reduces to the same consequence only in case of the fixed arm spacings. From these observations, it can be justified that not only the present model is physically more realistic, but also the behind-the-scene cause of the problem in the multiphase model is really associated with the coarsening. In addition, a legitimate background for using the parabolic concentration profile in solidification modelings has been established. That is, the timewise variation of the microscopic interface position predicted by the present model for the diffusion-controlled limit proved to agree indistinguishably with the above-mentioned analytical solution which is free from the type of concentration profile.

The practical utility of the present model has been effectively demonstrated by an example, where the effect of coarsening in the absence of back diffusion on the evolutions of the local solid fraction along three different cooling patterns has been investigated under the same solidification time. A number of physically meaningful facts have been revealed about the role of coarsening. The liquid concentration-solid fraction diagrams with coarsening appear to depend considerably on the cooling pattern, whereas those without coarsening coincide with each other, yielding a unique microsegregation curve. In result, all the eutectic fractions with coarsening are always smaller than the unified value for the fixed arm spacings at the end of dendritic solidification. This implies that the solute diffusion models, such as the multiphase model, in which the effect of coarsening is improperly taken into account, tend to underpredict the final eutectic fraction of practical importance. Another feature found is that the eutectic fraction decreases with accelerating the cooling rate at early stages of dendritic solidification. Accordingly, it is possible to control the microsegregation of the final solidification products by imposing the appropriately prescribed cooling patterns. However, the dependence of the eutectic formation on the cooling path in the presence of back diffusion needs further studies.

In conclusion, noting that the present model proved to account for the fundamental solute diffusion mechanisms properly and was expressed in a compact form suitable for use within the existing framework of computational procedure, the present model is expected to be feasible for the micro-macroscopic analysis of columnar dendritic alloy solidification as a microscopic component.

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