Modification of Ohnaka back diffusion equation

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Abstract. The first and simplest analytical model for a binary alloy came from Gulliver-Scheil, considering a constant partition coefficient at the interface during solidification without back diffusion in solid. Later, Brody-Flemings proposed an approximate analytical model, taking into account one dimensional diffusion of solute in solid during solidification in plate-like dendrite arms for parabolic growth. However, this model does not work well at high back diffusion values. Ohnaka modified this back diffusion parameter. The Ohnaka model works well at the low and high back diffusion values but it underestimates between them. In this paper, Voller-Ohnaka back diffusion model is modified further to predict concentration profiles more accurately than all others analytical models for the parabolic growth rate.

1. Introduction
During the solidification of an alloy, the solute rejected at the solid-liquid interface is redistributed in the dendritic structure, which is known as microsegregation. This controls the amount of interdendritic eutectic, the amount of inclusions, solidification temperature range, and hot tear, and also decreases mechanical and corrosion properties of final product.

The first attempts to predict microsegregation quantitatively were derived by Gulliver and Scheil and the equation is known as Gulliver-Scheil equation. It is assumed that there is complete mixing of solute in the liquid but no diffusion of solute in the solid can occur and local thermodynamic equilibrium exists at the solid-liquid interface, described by a constant equilibrium distribution coefficient (k) and negligible undercooling occurs at the tip. This equation is [1-2]:

\[ C_s = kC_o (1 - f)^{k-1} \]  
(1)

where:
- \( C_s \) = the solid composition (wt%),
- \( C_o \) = the average composition of an alloy (wt%),
- \( f \) = fraction of solid.

It predicts the worst segregation values. Since the introduction of the probe microanalysis technique in the late 1950s, it is shown that the assumption of no solid state diffusion is unjustified. Brody and Flemings recognised this and presented an analysis which quantifies the effect of solid state diffusion occurring between the Gulliver-Scheil and the lever rule cases. Their analytical solution for parabolic growth rate is [3-4]:

\[ C_s = C_o k (1 - (1 - 2\alpha k) f) \left( \frac{k-1}{1-2\alpha k} \right) \]  
(2)
where: \( \alpha = \left( \frac{D_s t_f}{L^2} \right) \) back diffusion parameter at the solid-liquid interface,
\( D_s \) the diffusion coefficient in solid (m\(^2\)/s),
\( t_f \) the solidification time (s),
\( L \) the half of the solidified distance (m).

In this equation the dimensionless parameter, \( \alpha \), determines the extent of diffusion in the solid. However, this treatment does not conserve solute, especially for fast diffusion elements, i.e. the applicability of this equation is limited to \( k\alpha < 0.1 \).

2. Back diffusion models
Since Brody-Flemings introduced this back diffusion parameter into the microsegregation equation, several researchers have attempted to modify back diffusion parameter to predict concentration profiles more accurately than the Brody-Flemings model.

Clyne and Kurz examined the influence of rapid solid state diffusion in solidification. They derived a relationship for large \( \alpha \) values. In their model the \( \Omega \) replaced by \( \alpha \) is [5]:

\[
\Omega = \alpha \left( 1 - \exp \left( -\frac{1}{\alpha} \right) \right) - 0.5 \exp \left( -\frac{1}{2\alpha} \right)
\]  

This equation works well at low and high \( \alpha \) values but not between them. Ohnaka has criticised their model and solved back diffusion equation approximately for the planar solidification [6]. He assumed that solute profiles in the solid can be expressed with a quadratic equation, then he obtained the following back diffusion equation.

\[
\Gamma = \frac{2\alpha}{1 + 2\alpha}
\]  

Comparing with the Kurz-Fisher equation he replaced \( 2\Omega \) with \( \Gamma \). His results showed that his equation can estimate concentration profiles better than the Brody-Flemings’s solution but it gives similar results to the Clyne-Kurz solution.

Voller suggested two different approaches for the back diffusion at the solid-liquid interface which are called the profile model and the parameter model. In the parameter model he replaced \( \beta \) with \( \Gamma \) in the Ohnaka’s model. His model for parabolic growth rate is [7]:

\[
\beta = \frac{2\gamma \alpha}{2\alpha + \gamma}
\]  

where \( \gamma \) is an integration factor, and

\[
\gamma = \frac{A\alpha k}{A\alpha k + 1}
\]  

where \( A \) is a constant.

He calculated \( A=4 \) in this parameter by comparing Kobayashi’s exact results with his suggested equation. These two equations can be simplified as follows:

\[
\beta = \frac{2\alpha}{1 + 2\alpha + cor}
\]
Comparing to Ohnaka’s back diffusion model he only added cor (cor=(1/2k)) to the denominator part of the Ohnaka’s equation. Voller showed that his model can predict very closely the maximum concentration of Kobayashi’s model. However, the prediction of this model for an amount of eutectic and therefore, for concentration profiles is not very close to the exact model as shown in this paper. In order to improve his back diffusion model, a fitting function is introduced to his equation as given below:

\[
cor = \frac{f^{10+2f^2}}{2k}
\]  

(8)

As you can see in equations 7 and 8, back diffusion parameter is not constant but is a function of k, \(\alpha\), and f. cor is not an integration factor anymore as Voller suggested but is a correction variable for back diffusion model.

On the other hand, Ganesan and Poirier wrote a numerical programs for the planar dendritic solidification with the parabolic growth rate. Then comparing this numerical model with Brody-Flemings analytical model for large number of \(\alpha\) and k values and using a statistical package they developed an alternative back diffusion equation for parabolic growth rate. In their model the \(\mu\) replaced by \(\alpha\) is [8]:

\[
\mu = \frac{\exp\left(-\frac{Af}{(ak)^\alpha}\right)}{2 + \frac{Cf}{\alpha^\beta}}
\]  

(9)

where A, B, C, and D are constants and A=0.1523, B=0.4331, C=1.1093, and D=0.9367. Similarly, their results showed that back diffusion is not constant during solidification but is a function of \(\alpha\), k, and f. It is a good agreement with the present suggestion.

3. Results and comparisons

Ohnaka, Voller, Ganesan-Poirier, and present modified back diffusion equations are plotted as a function of fraction of solid for two different \(\alpha\) values (\(\alpha=0.1\) and \(\alpha=1\)) in the figure 1. It can be easily recognized that modified back diffusion model decreases gradually from Ohnaka back diffusion model to Voller back diffusion model after f=0.6 and it is equal to the Voller back diffusion model at f=1, therefore it does not affect its prediction of maximum concentration. Ganesan-Poirier model shows similar behavior but its prediction is slightly less than the present curve.

The prediction of amount of eutectic structure of several analytical models are compared to each other in figure 2-4 for \(C_{eu}/C_o=3, 6.5\) and 12 values, respectively. As you can see in the figure 2, the prediction of amount of eutectic of the present analytical model (with modified back diffusion parameter) is very close to the Kobayashi’s results [9] whereas, the prediction of Ohnaka’s model underestimates them. On the other hand, Voller model overestimates the amount of the eutectic structure. The estimations of Wang-Beckermann model [10] is also very close to the exact solution of Kobayashi at the low \(C_{eu}/C_o\) values.

For \(C_{eu}/C_o=6.5\), all available analytical models in literature are compared with the exact analytical model of Kobayashi in the figures 3a and 3b. The behaviour of all analytical models are almost the same as the previous figure. Kurz-Fisher and Ohnaka models underestimate eutectic values, whereas, Voller’s model overestimates them. The prediction of Wang-Beckermann model at low partition coefficients are very close to the exact model, however, increasing k values it overpredicts the amount of eutectic because this model becomes infinite when the fraction of solid approaches to one. Ganesan-Poirier estimates very closely to the exact model of Kobayashi. The prediction of
Nastac-Stefanescu model [11] has its own unique profiles because no growth rate is dictated in the assumption of this model.

**Figure 1.** Comparisons of back diffusion models as a function of fraction of solid for two different $\alpha$ values ($k=0.2$, $\alpha=0.1$ and $\alpha=1$).

**Figure 2.** Prediction of eutectic structure as a function of back diffusion parameter for several models (for $C_{eu}/C_o=3$).
Figure 3. a and b. Prediction of eutectic structure as a function of back diffusion parameter for several models (for $C_{eu}/C_o=6.5$).

Figure 4. Prediction of eutectic structure as a function of back diffusion parameter for several models (for $C_{eu}/C_o=12$).

Some models which are very close to the exact solution are compared to each other only for $C_{eu}/C_o=12$. As shown in the figure 4, the prediction of present analytical and Ganesan-Poirier models are very close to the prediction of exact solution of Kobayashi whereas the Wang-Beckermann model overestimates it with increasing $k$ values.
4. Conclusion
Voller-Ohnaka back diffusion equation is modified and compared with other analytical models. It is shown that the prediction of the amount of eutectic structure using the modified back diffusion equation is very close to the exact solution of Kobayashi model. The calculation of the present model is much easier than the exact Kobayashi equation.

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