

A New Model for Precipitation at Moving Interphase Boundaries

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A new model has been developed to describe precipitation in sheets at a moving, planar, interphase boundary. The model assumes that the diffusion profile developed by the growing sheet of interphase precipitates is similar to that resulting from the growth of a "pseudophase" of constant composition equal to the average solute concentration in the sheet. It assumes that the solute lost by the depleted layer during growth is equal to the excess solute found in the sheet. Nucleation of a new sheet is determined by the local solute concentration at migrating ledges at the interphase boundary. The model provides a theoretical rationale for correlations among parameters important in interphase precipitation and gives a coherent description of this phenomenon, upon which further understanding can be based.

I. INTRODUCTION

INTERPHASE precipitation has been categorized by Howell and Honeycombe^[1] as occurring at static and dynamic interfaces in many alloy systems. Typical examples of precipitation at static boundaries were cited as (a) the formation of θ' on GP zones in Al-Cu alloys;^[2,3] (b) σ phase nucleation on austenite/ferrite (γ/α) boundaries in duplex austenitic stainless steels;^[4] and (c) *in situ* transformation of carbides during the tempering of steels.^[5,6] Four examples of precipitation at dynamic interfaces were presented including (i) precipitation of alloy carbides at the γ/α interphase boundary;^[7,8,9] (ii) precipitation in carbon-free iron base alloys,^[10,11] (iii) nucleation at the bainite/austenite interface,^[12] and (iv) sympathetic nucleation.

The precipitation of alloy carbides or a metallic phase at the γ/α interphase boundary is of particular interest in this paper, since such precipitation may be associated with planar, semi-coherent interphase boundaries.^[13-16] Transmission electron microscopy studies have shown that sheets of precipitates, with uniform sizes and intersheet spacings, may be formed during isothermal transformation and can result in significant strength increases in these alloys.^[17-22] No quantitative analyses are available to predict the sizes and spacings of these precipitates in order to optimize the mechanical properties of such steels.

Roberts^[23,24] has proposed a solute depletion model for vanadium carbide (VC) in ferritic steels, in which VC precipitates nucleated homogeneously behind a migrating, nonfaceted γ/α boundary, depleting the surrounding ferrite of vanadium. Nucleation of a second sheet of precipitates could not then occur until the solute concentration behind the migrating interphase boundary had risen to a sufficiently high level. Roberts considered only the depletion zone that develops around an isolated spherical particle, and thus his analysis can not be applied to calculate the concentration profile produced by the uniformly distributed, closely spaced precipitates in an interphase precipitate sheet. Also, Roberts ignored the diffusivity discontinuity at

the interphase boundary. Further, it is difficult to reconcile Roberts' model with the ledge mechanism of ferrite growth and the planar sheets of VC precipitates frequently observed.

A solute balance approach is applied in the present model to precipitation at planar interphase boundaries. The model uses the solution for diffusion controlled planar growth obtained by Zener^[25] and investigates the hypothesis that the diffusion profile developed by the growing precipitate sheet is similar to that resulting from the growth of a "pseudophase" of constant composition equal to the average concentration of the precipitate sheet.

II. THE MODEL

The following premises are adopted in the new model for interphase precipitation, which is associated with the transformation of β phase to α phase with precipitates (P) formed at the α/β boundary:

1. The diffusion profile developed during growth of a sheet of interphase precipitates can be represented by that resulting from the growth of a "pseudophase" of constant solute concentration equal to the average concentration of the sheet.
2. The solute lost by the depleted layer during growth is exactly equal to the excess solute found in the precipitate sheet.
3. The average solute concentration in α at the interphase boundary is constant between nucleation events.
4. Nucleation of a sheet of precipitates occurs when the local solute concentration at migrating ledges at the interphase boundary reaches a critical value. (N.B. The intersheet spacing need not be equal to the ledge height.)
5. The nucleation event does not hinder the migration of the α/β interphase boundary, *e.g.*, by pinning the boundary.
6. The nucleation time for a sheet of precipitates is short once the critical solute concentration is reached at the ledges.
7. Precipitation has been completed but significant coarsening has not occurred.

The model can be applied to binary or pseudobinary systems where the precipitate and matrix phases can be treated as having constant compositions throughout the interphase precipitation reaction. The analysis applies only to systems where precipitate growth is controlled by the diffusion of one species.

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III. RESULTS

A. The Diffusion Profile

Zener^[25] has presented solutions for the multi-dimensional moving boundary diffusion problem. For one-dimensional growth of a plate, the solution can be represented by the equation

$$\frac{C^m - C^{mo}}{C^{mp} - C^{mo}} = \frac{\Phi(y/\sqrt{Dt})}{\Phi(s)} \quad [1]$$

where

$$\Phi(z) = \sqrt{\pi} \operatorname{erfc}(z/2) \quad [2]$$

$$y_p = s\sqrt{Dt} \quad [3]$$

and C^m = solute concentration in the matrix at position $y(t)$, C^{mo} = initial concentration of solute in the matrix, C^{mp} = concentration of solute at the precipitate/matrix interface (normally approximated by the equilibrium concentration), D = solute diffusivity in the matrix (α phase), t = time, y_p = forward growth distance of the plate. The parameter, s , is calculated from the equation

$$\Omega = \frac{C^{mo} - C^{mp}}{C^p - C^{mp}} = \frac{s\sqrt{\pi}}{2} \exp\left(\frac{s^2}{4}\right) \operatorname{erfc}\left(\frac{s}{2}\right) \quad [4]$$

where C^p is the concentration of solute in the plate. The concentration, C^m , is plotted schematically as a function of distance, y , at a time, t , in Figure 1(a).

Zener showed that a good approximation of Eq. [4] was given by the equation

$$s = \frac{\Omega}{\sqrt{1 - \Omega}} \quad [5]$$

Exact and approximate solutions, together with two asymptotic solutions derived by Zener, have been compared by Engberg, Hillert, and Odén.^[26] Their work is replotted in Figure 2.

The approximate solution, Eqs. [1] through [3] with Eq. [5], which is referred to as the quasi-stationary solution, assumes that (i) the depleted layer in front of the advancing planar precipitate has a definite thickness, y_p to L , as shown in Figure 1(b); (ii) the depleted layer between y_p and L has exactly lost the excess solute content found in the precipitate plate, and (iii) the concentration profile in the depleted layer is given at any moment by Fick's first law assuming steady state diffusion through the layer from L to y_p .

The application of these results to the solute balance model for the interphase precipitation reaction is straightforward. The sheet of interphase precipitates is replaced by a pseudophase with constant solute concentration, C^p , equal to the average concentration of the sheet. If the size and distribution of the precipitates is known along with (a) their composition and molar volume, and (b) the composition and molar volume of the matrix, then the solute concentration of the pseudophase, C^p , can be directly calculated. However, such information is not normally avail-

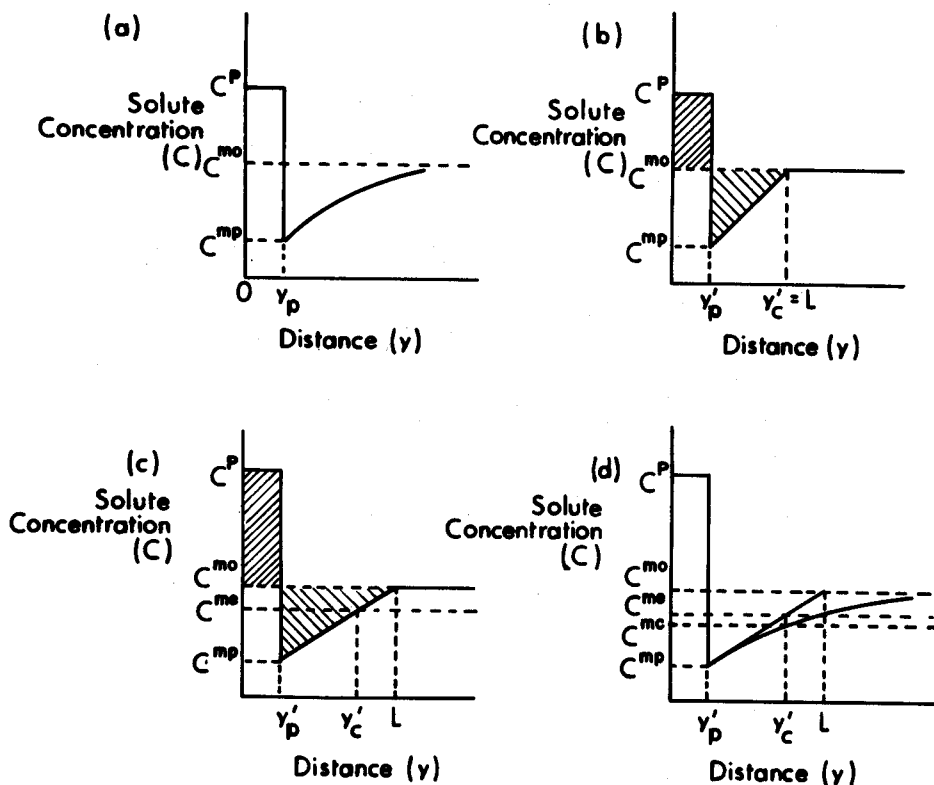


Fig. 1—Schematic representation of the concentration profile produced by the growth of a plate precipitate or pseudophase: (a) actual profile; (b) approximate linear profile produced by the growth of a plate precipitate according to the "quasi-stationary" approximation; (c) approximate linear profile for the case where the nucleation of a new sheet of interphase precipitates does not occur at the boundary of the depleted layer; and (d) actual profile and approximate linear profile for the case where $C^{mc} < C^{mo}$, indicating the relationship of C^{mc} to C^{mo} .

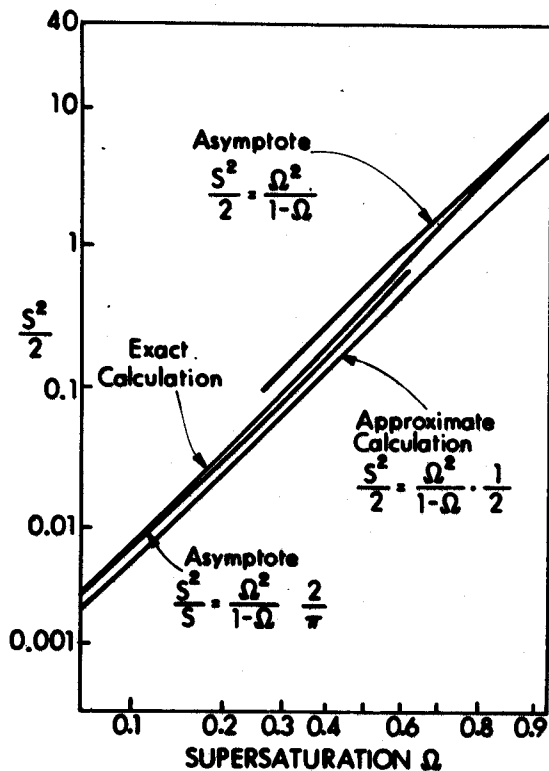


Fig. 2—The growth rate of a planar precipitate as a function of the supersaturation.

able, so C^p will be calculated here by equating the two cross-hatched areas shown in Figure 1(b) at the instant when a new sheet is nucleated. This procedure, which assumes that the increased solute in the pseudophase is provided from a depleted layer between the position of the pseudophase boundary, y'_p , and the α/β interphase boundary, y'_c , at the time, t'_c , when the next sheet is nucleated, gives the equation

$$(C^p - C^{mo})y'_p = \frac{1}{2}(C^{mo} - C^{mp})(L - y'_p) \quad [6]$$

where L is the position of the boundary of the depleted layer, and in this case is equal to y'_c . The parameter C^{mp} is used to designate the solute concentration in the matrix at the pseudophase/matrix boundary. This, of course, is also equal to the solute concentration in the matrix in the sheet of interphase precipitates represented by the pseudophase. This solute concentration can often be approximated by the concentration of solute in the α phase in equilibrium with the precipitates. Equation [6] can be solved for the unique composition of the pseudophase, C^p , giving

$$C^p = \frac{1}{2}(C^{mo} - C^{mp})\left(\frac{y'_c}{y'_p} - 1\right) + C^{mo} \quad [7]$$

This equation is appropriate for the case where no solute is transported by diffusion across the interphase boundary. It should be noted that if solute balance is applied in this manner, the layer $0 \leq y \leq L$, Figure 1(b), has an average solute concentration equal to C^{mo} . Hence, no long range diffusion fields develop in front of the advancing interphase interface.

B. Boundary Concentration

If solute is transported across the interphase boundary and the diffusivities of the solute are approximately the

same in both phases, then the nucleation of a new sheet of interphase precipitates may not occur at the boundary of the depleted layer. In this case, it is convenient to define an effective matrix concentration $C^{me} < C^{mo}$ (Figure 1(c)), which allows the depleted layer to extend beyond y'_c , the position where the next sheet is nucleated, and gives

$$L - y_p = \frac{C^{mo} - C^{mp}}{C^{me} - C^{mp}}(y_c - y_p) \quad [8]$$

In this case, the pseudophase will have a larger concentration of solute than if $C^{me} = C^{mo}$ (Figure 1(b)). The pseudophase concentration is given by the equation

$$C^p = \frac{(C^{mo} - C^{mp})^2}{2(C^{me} - C^{mp})} \left(\frac{y'_c}{y'_p} - 1\right) + C^{mo} \quad [9]$$

where y'_c and y'_p are the positions of the interphase and pseudophase boundaries at the time, $t = t'_c$, when the next sheet is nucleated. The parameter Ω is given by the equation

$$\Omega = \left(\frac{1}{2} \frac{C^{mo} - C^{mp}}{C^{me} - C^{mp}} \left(\frac{y'_c}{y'_p} - 1\right) + 1\right)^{-1} \quad [10]$$

The forward growth distance of the pseudophase is given by

$$y_p = s\sqrt{Dt} \cong \frac{\Omega}{\sqrt{1-\Omega}} \sqrt{Dt} \quad [11]$$

where $t =$ time. The distance between the α/β interphase boundary and the pseudophase boundary, $y_c - y_p$, is given by the equation

$$y_c - y_p = \left(\frac{C^{me} - C^{mp}}{C^{mo} - C^{mp}}\right) 2\sqrt{1-\Omega} \sqrt{Dt} \quad [12]$$

The position y_c , where the concentration equals C^{me} for the linear concentration profile of the quasi-stationary solution, is given by the equation

$$y_c = \left(\frac{C^{me} - C^{mp}}{C^{mo} - C^{mp}} 2\sqrt{1-\Omega} + \frac{\Omega}{\sqrt{1-\Omega}}\right) \sqrt{Dt} \quad [13]$$

The average concentration of solute in α at the α/β interphase boundary, C^{mc} (i.e., the actual average α concentration at the position $y_c = y'_c$, and time $t = t'_c$, where and when the next sheet nucleates), is given for the quasi-stationary solution by the equation

$$\frac{C^{mc} - C^{mo}}{C^{mp} - C^{mo}} = \frac{\operatorname{erfc}(y'_c/2\sqrt{Dt'_c})}{\operatorname{erfc}\frac{\Omega}{2\sqrt{1-\Omega}}} \quad [14]$$

where $y'_c/\sqrt{Dt'_c}$ is given by Eq. [13].

This equation may be rearranged to describe the position where the solute concentration equals C^{mc} , which according to the model is also equal to the position of the α/β interphase boundary, y_c , and is given by

$$y_c = 2\sqrt{Dt} \operatorname{erf}^{-1}\left(1 - \frac{C^{mc} - C^{mo}}{C^{mp} - C^{mo}} \operatorname{erfc}\frac{\Omega}{2\sqrt{1-\Omega}}\right) \quad [15]$$

Equations [13] and [15] establish a relationship, between y'_c/y'_p and C^{mc} , where y'_p is the forward growth distance of the pseudophase at $t = t'_c$. For a specific C^{mc} , y'_c/y'_p is the root of the equation

$$\frac{C^{me} - C^{mp}}{C^{mo} - C^{mp}} 2\sqrt{1 - \Omega} + \frac{\Omega}{\sqrt{1 - \Omega}} = 2 \operatorname{erf}^{-1} \left(1 - \frac{C^{mc} - C^{mo}}{C^{mp} - C^{mo}} \operatorname{erfc} \frac{\Omega}{2\sqrt{1 - \Omega}} \right) \quad [16]$$

A schematic plot of Eq. [14] indicating the relationship of C^{mc} to C^{me} is given in Figure 1(d). In the case where the depleted layer extends beyond the interphase boundary, the average solute content of the pseudophase and the depleted layer is not equal to C^{mo} . Thus a long range diffusion profile will develop in front of the advancing α/β boundary and may cause instabilities to occur.

Equations [8] through [16] can be applied to the case where no solute is transferred across the interphase boundary by setting C^{me} equal to C^{mo} .

C. Temperature Dependence of the Intersheet Spacing and Average Boundary Velocity

Equations [13] and [15] can be written in the form

$$y_c = A\sqrt{Dt} \quad [17]$$

where y_c is the position of the interphase boundary and A is a function of C^{mc} , C^{me} , C^{mp} , C^{mo} , and y'_c/y'_p . The boundary moves with a fixed concentration C^{mc} and its position fixes the amount of solute contributed by the depleted layer to the pseudophase. The precipitate sheet spacing is given by the equation

$$y'_c = A\sqrt{Dt'_c} = At'_c{}^{1/2} D_0^{1/2} e^{-\Delta H_p/2RT} \quad [18]$$

where D is the diffusivity of solute and ΔH_p is the activation energy for solute diffusion.

The average velocity of the α/β boundary is given by the equation

$$\bar{v}_b = \frac{y'_c}{t'_c} = A \frac{D^{1/2}}{t'_c{}^{1/2}} = At'_c{}^{-1/2} D_0^{1/2} e^{-\Delta H_p/2RT} \quad [19]$$

A plot showing schematically the position of the interphase boundary as a function of time during growth of the i^{th} precipitate sheet is given in Figure 3.

IV. DISCUSSION

The new model predicts relationships among the variables important in the interphase precipitation reaction. Li and Todd have applied the new model to interphase precipitation in Fe-C-V steels and have concluded that it provides a satisfactory interpretation of the observed correlations.^[16] In this section, data for a specific Fe-C-V steel and isothermal annealing temperature are analyzed so that several features of the model can be demonstrated. Also, a test for the model is proposed.

Batte and Honeycombe^[8] have measured the intersheet spacing and intersheet width to be 30 nm and 15 nm, respectively, for an Fe-0.2C-1.04V-0.023Nb alloy austenitized at 1200 °C and isothermally aged at 825 °C. The concentration of vanadium in this alloy is 9.67×10^{20} atoms cm^{-3} , which is set equal to C^{mo} . The precipitate is taken to be stoi-

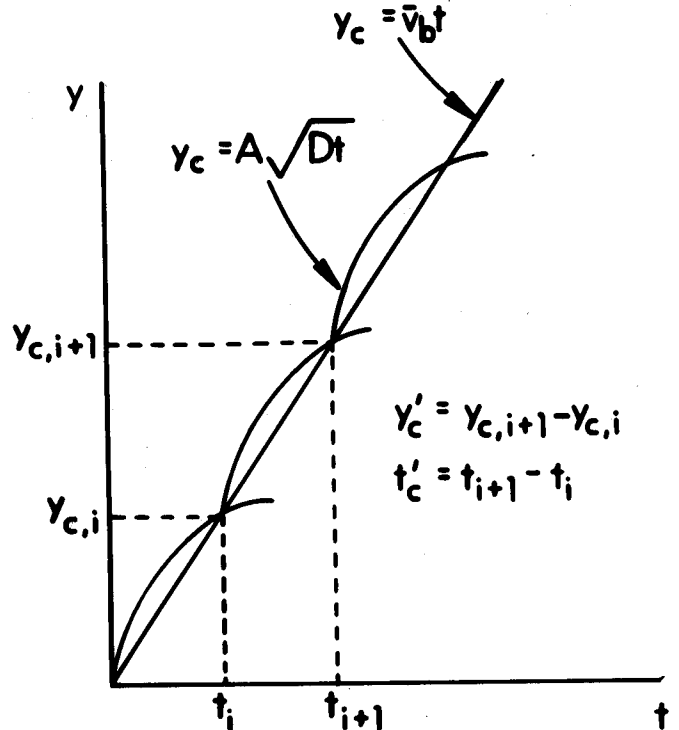


Fig. 3—Position of the interphase boundary shown schematically as a function of the time for nucleation and growth of the i^{th} precipitate sheet.

chiometric VC throughout the precipitation reaction. The concentration of vanadium at the pseudophase boundary is thought to be close to that in equilibrium with VC and, hence, to be very small. Accordingly, in the calculations C^{mp} is set equal to zero.

At 825 °C the diffusivity of vanadium in ferrite is approximately 300 times greater than in austenite. Thus, the case where no solute is transferred by diffusion across the interphase boundary, *i.e.*, $C^{me} = C^{mo}$, applies. The diffusivity of vanadium is much less than that of carbon in both ferrite and austenite so vanadium diffusion controls precipitate growth.

A relationship between the intersheet spacing (y'') and y'_c , and the intersheet width (y'_p) and y'_p is illustrated in Figure 4. Figure 4(a) shows a pseudophase layer nucleated at the origin that has grown to a thickness y'_p in a time t'_c . Also shown is a sheet of nuclei at y'_c , where the next pseudophase layer has just nucleated. According to Eq. [6] solute balance requires that

$$\frac{C^p}{C^{mo}} = \frac{1}{2} \left(\frac{y'_c}{y'_p} + 1 \right) \quad [20]$$

Figure 4(b) shows two pseudophase layers and the solute concentration distribution after all precipitation is completed. Residual solute due to the concentration variation from $C^{mp} = 0$ at y'_p to C^{mc} at y'_c has caused additional growth to occur at the ends of the precipitates forming the pseudophase layers. In this case, solute balance requires that

$$\frac{C^p}{C^{mo}} = \frac{y''}{y'_p} \quad [21]$$

If the composition of the pseudophase is constant, which is a basic premise of the model, then (noting that $y'' = y'_c$)

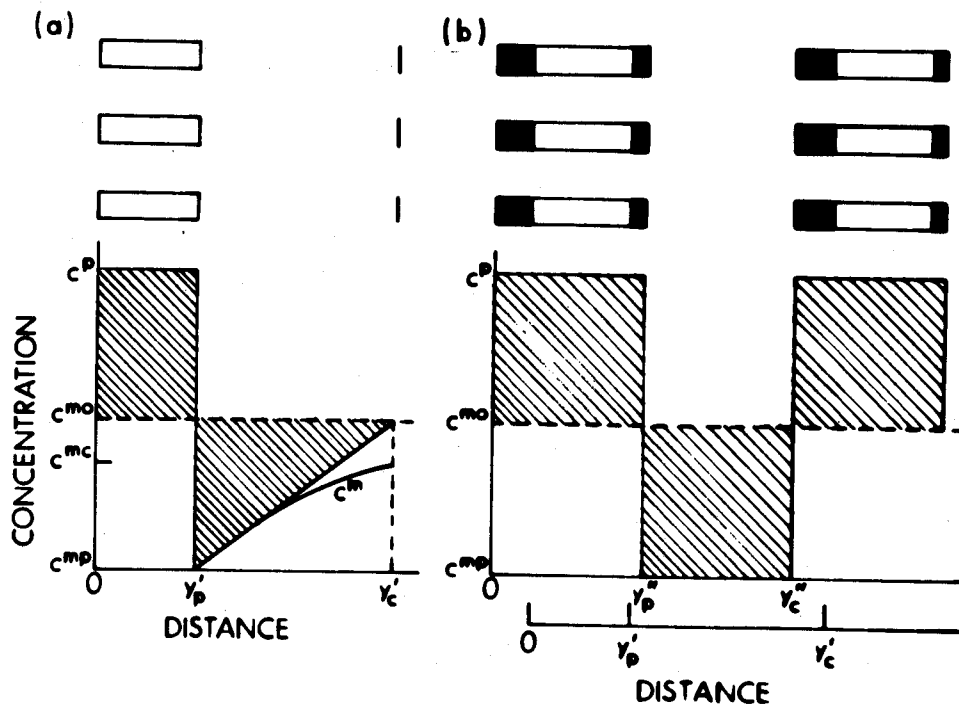


Fig. 4—Diagram showing the solute concentration distribution and the relationship of y'_c to y'_c and y''_p to y'_p ; (a) at $t = t'_c$, the instant a new pseudophase layer is nucleated; and (b) after precipitation is completed (shaded regions of precipitates show where additional growth has occurred due to the residual solute).

$$\frac{y'_c}{y''_p} = \frac{1}{2} \left(\frac{y'_c}{y'_p} + 1 \right) \quad [22]$$

Thus if y''_c and y''_p are measured to be 30 nm and 15 nm, then y'_c and y'_p must be 30 nm and 10 nm, respectively.

With the values for y'_c and y'_p , a value for Ω can be calculated using Eq. [10].

$$\begin{aligned} \Omega &= \left[\frac{1}{2} \left(\frac{y'_c}{y'_p} - 1 \right) + 1 \right]^{-1} \\ &= \left[\frac{1}{2} \left(\frac{30}{15} - 1 \right) + 1 \right]^{-1} = 0.500 \end{aligned}$$

The pseudophase composition is given by Eq. [9]

$$\begin{aligned} C^p &= \frac{1}{2} (C^{mo} - C^{mp}) \left(\frac{y'_c}{y'_p} - 1 \right) + C^{mo} \\ &= \frac{1}{2} (9.67 \times 10^{20}) \left(\frac{30}{10} - 1 \right) + 9.67 \times 10^{20} \\ C^p &= 19.3 \times 10^{20} \text{ atoms cm}^{-3} \end{aligned}$$

The intersheet repeat period can be calculated using Eq. [3] and setting $D = 1.22 \times 10^3 \text{ nm}^2 \text{ s}^{-1}$

$$t'_c = \frac{y_p'^2 (1 - \Omega)}{D \Omega^2} = \frac{(10)^2 (1 - 0.500)}{(1.22 \times 10^3) (0.500)^2} = 0.163 \text{ s}$$

According to this value, the average velocity of the interphase boundary is $y'_c/t'_c = 30/0.163 = 184 \text{ nm s}^{-1}$. The critical supersaturation, $C^{mc} = 7.58 \times 10^{20} \text{ atoms cm}^{-3}$.

Clearly, it is possible, knowing C^{mo} , y'_c , and y'_p to evaluate model variables such as C^p , t'_c , and C^{mc} . Evaluating these variables may give valuable insights regarding inter-

phase precipitation. However, the ultimate goal of a theory of the interphase precipitation reaction is to predict relationships between measurable quantities such as y'_c and y''_p and other measurable quantities.

One approach to developing such a theory is to measure or predict t'_c and either C^{mc} or C^p . If C^{mc} is known for a specific alloy composition and isothermal transformation temperature, then y'_c/y'_p can be determined by solving Eq. [16]. This establishes the solute concentration (by Eq. [9]) of the pseudophase that will consume all of the solute from the depleted zone produced by the moving boundary. Alternatively, if C^p is measured, then y'_c/y'_p can be determined by Eq. [9]. If t'_c is known, then y'_c can be calculated using Eq. [18]. According to the model, C^p and C^{mc} are both constant during growth of the pseudophase. Unfortunately, experimental measurement or theoretical calculations of both these parameters would appear to be difficult.

A more feasible approach to test the validity of the model involves measurement of the average interphase boundary velocity, \bar{V}_b , in a volume where the interphase precipitate sheet spacing, $y'_c = y''_c$, is known. According to Eq. [19], $t'_c = y'_c/\bar{V}_b$; consequently

$$A = \left(\frac{\bar{V}_b y'_c}{D} \right)^{1/2} \quad [23]$$

For the case of alloys such as the Fe-C-V steels, where no solute is transported by diffusion across the interphase boundaries and the concentration of solute in equilibrium with the precipitate is small, $C^{mc} = C^{mo}$ and $C^{mp} \cong 0$. Equations [13] and [15] give

$$A = \frac{2 - \Omega}{\sqrt{1 - \Omega}} \quad [24]$$

For the case where $C^{mo} = C^{me}$, Ω is a function only of y'_c/y'_p , see Eq. [10]. Accordingly, y'_c/y'_p and hence (by Eq. [22]) y''_p can be predicted from measurements of \bar{V}_b and $y'_c = y''_c$, which provides a critical test of the theory.

V. CONCLUSIONS

The new model provides a basis for understanding correlations among parameters important in interphase precipitation. For a specific alloy, it can be employed to relate interphase precipitate sheet spacing and width, vanadium diffusivity, and average interphase boundary velocity. If the average interphase boundary velocity is measured, and the diffusivity and intersheet spacing are known, then the sheet width can be predicted. Alternatively, if the sheet spacing and width are measured and the diffusivity is known, then the interphase boundary velocity can be predicted. The experimental verification of such predictions appears feasible and should provide a critical test of the model.

LIST OF SYMBOLS

A	constant
C^m	solute concentration in the matrix at position $y(t)$
C^{mc}	average solute concentration in the α phase at the interphase boundary ($y_c = y'_c$) at the time $t = t'_c$, when the next sheet nucleates
C^{me}	effective concentration of solute in the α matrix defined for the case where solute can be transported across the interphase boundary
C^{mo}	initial concentration of solute
C^{mp}	concentration of solute at the precipitate/matrix interface or the pseudophase/matrix interface
C^p	concentration of solute in the pseudophase
D	solute diffusivity in the matrix (α phase)
D_0	diffusivity coefficient
ΔH_p	activation energy for solute diffusion
L	position of the boundary of the depleted layer given by the "quasi-stationary" approximation
Ω	supersaturation
R	gas constant
t	time
t'_c	critical time between pseudophase sheet nucleation events
T	temperature
\bar{V}_b	average velocity of the interphase boundary
y_c	position of the advancing α/β interphase boundary
y'_c	position of the advancing α/β interphase boundary at time t'_c when the next pseudophase sheet is nucleated
y''_c	intersheet spacing ($= y'_c$)
y_p	forward growth distance of the pseudophase sheet
y'_p	forward growth distance of the pseudophase sheet at time t'_c when the next pseudophase sheet is nucleated
y''_p	final interphase precipitate sheet width

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