The Structures Expected in a Simple Ternary Eutectic System: Part I. Theory

D. G. MCCARTNEY, J. D. HUNT, AND R. M. JORDAN

The structures which are to be expected during the steady state directional growth of alloys in a simple ternary eutectic system are discussed and it is predicted that for a particular velocity and temperature gradient five different structural regions can be observed. The structural regions of a two component system may be explained using a stability analysis and a competitive growth criterion, and it is suggested that these ideas may be extended to a three component system. Additional models have been proposed for the growth of a planar front two phase eutectic and single phase dendrites in a three component system.

THE purpose of this paper is to discuss the structures expected during the steady state directional growth of alloys in simple ternary eutectic systems such as that shown in Fig. 1. It is shown that the ideas developed to predict the ranges of structures in two component systems may be extended to predict the ranges found in three component systems.

Considering the simple ternary system of Fig. 1 and assuming that each of the three primary phases α, β and γ grow in a nonfaceted fashion, and nucleate easily, a number of different structural regions are then to be expected. The composition ranges of each of these for a fixed growth velocity and temperature gradient are shown schematically in Fig. 2, and referring to this figure:

Region 1—Near the pure component corners single phase growth occurs with a planar growth interface (*e.g.* Flemings[1]);

Region 2—Near the two component eutectic compositions two phase eutectic growth occurs, again with a macroscopically planar growth interface (*e.g.* Flemings[2]), and usually with a lamellar or rod-like structure.[3] The growth interface is shown schematically in Fig. 3(*a*);

Region 3—For alloys near the eutectic valley two phase eutectic cells or dendrites are expected with the third component appearing near the ternary eutectic temperature either as a thin layer of this phase or as a three phase eutectic layer around each cell depending on the volume fraction of the ternary eutectic. The growth interface is shown schematically in Fig. 3(*b*).

Region 4—Near the ternary eutectic composition a three phase eutectic structure is expected without any primary single phase or two phase eutectic (*e.g.* Kerr *et al*[4]). The three phase structure should grow with a (macroscopically) planar interface in a similar fashion to the pure single phase, and the pure two component eutectic. The growth interface is shown schematically in Fig. 3(*c*). Clearly the actual structure may be complex and not necessarily a simple lamellar or rod-like one.[5]

Region 5—For alloys away from the eutectic valleys single phase cells or dendrites are to be expected, followed by the structures described in regions 2, 3 and 4, and can be as follows:

a) Single phase dendrites can be followed by two

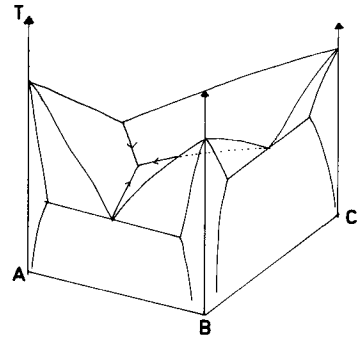

Fig. 1—Ternary eutectic phase diagram.

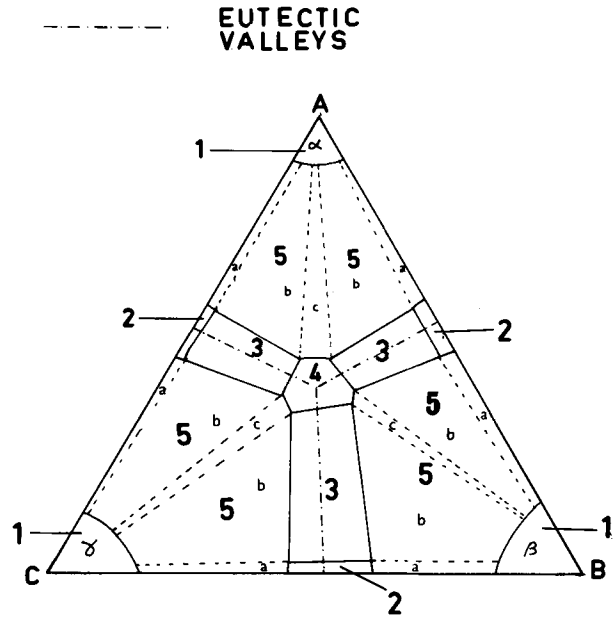

Fig. 2—Schematic representation of the composition boundaries of the various structural regions (dotted and solid lines) for a fixed velocity and temperature gradient. The numbers and letters refer to the regions described in the text.

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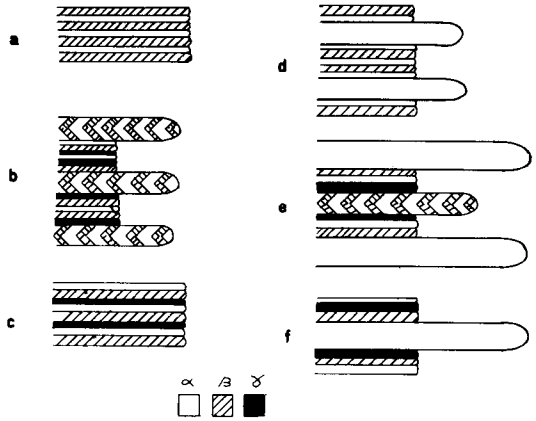

Fig. 3—Schematic growth interfaces in a ternary system: (*a*) two phase eutectic, (*b*) two phase cells plus ternary eutectic, (*c*) three phase eutectic, (*d*) single phase cells plus binary eutectic, (*e*) single phase cells, two phase cells, and ternary eutectic, and (*f*) single phase cells plus ternary eutectic.

phase eutectic growing with a planar interface in region 5(*a*). The growth interface is shown schematically in Fig. 3(*d*).

b) Single phase dendrites can be followed firstly by two phase cells and finally by three phase eutectic in region 5*b*. The growth interface is shown schematically in Fig. 3(*e*).

c) Single phase dendrites can be followed by three phase eutectic in region 5*c*. The growth interface is shown schematically in Fig. 3(*f*) although in practice, near the pure component corners, the residual liquid may freeze as a single or two phase liquid layer around the single phase cells.

TWO COMPONENTS

In a two component system the limit of planar single phase growth has been shown by stability analysis[6] to be given to a good approximation assuming equal thermal conductivities in the solid and liquid by the constitutional undercooling condition. That is for planar interface growth

$$G > m\left(1 - \frac{1}{K}\right)\frac{V}{D}C_\infty \qquad [1]$$

where G is the temperature gradient, m the liquidus slope, k the distribution coefficient, V the growth velocity, D the liquid diffusion coefficient and C_∞ the alloy composition.

The eutectic range, defined as the composition range over which eutectic growth occurs without a primary phase being present, depends on growth velocity and temperature gradient. It has been shown[7,8] that the complex dependence of this two phase eutectic range on growth velocity and temperature gradient can be explained in terms of a competitive growth condition. That is single phase cells or dendrites will only be present with the eutectic when the dendrites grow at a higher temperature than the eutectic. The critical condition is thus when the dendrite tip temperature T_{ID} equals the eutectic growth temperature T_{IE}. Defining the eutectic undercooling $\Delta T_E = T_E - T_{IE}$ (where T_E is the equilibrium eutectic temperature) and the dendrite undercooling $\Delta T_D = T_o - T_{ID}$ (where T_o is the alloy liquidus temperature) gives $\Delta T_D - \Delta T_E = T_o - T_E$ at the critical condition. Noting from the phase diagram that $T_o - T_E = m(C_\infty - C_E)$ where C_E is the eutectic composition and defining the eutectic range on one side of the phase diagram as $\Delta C = (C_\infty - C_E)$ gives

$$\Delta C = (\Delta T_D - \Delta T_E)/m. \qquad [2]$$

The eutectic undercooling is given by $\Delta T_E = A V^{1/2}$ where A is a constant which may be obtained from Ref. 9. The dendrite undercooling was given by Burden and Hunt[10-12] as

$$\Delta T_D = \frac{GD}{V} + 2\left[-\frac{2aV}{D}\left(m(1-K)C_\infty + \frac{KGD}{V}\right)\right]^{1/2} \qquad [3]$$

Thus the eutectic range may be calculated.

THREE COMPONENTS

It is suggested that lines defining the limits of the regions in three component systems can be predicted by using similar reasoning.

i) The limit of region 1, *viz.* that of planar single phase growth, will be given by the constitutional undercooling condition modified for three components. That is

$$G > m_{jB}\frac{dC_B}{dx} + m_{jc}\frac{dC_c}{dx} \qquad [4]$$

where $j = \alpha, \beta$ or γ phases, $m_{jB} = (\partial T/\partial C_B)_{C_c}$ and $m_{jC} = (\partial T/\partial C_C)_{C_B}$ are the slopes of the liquidus surfaces, and subscript B and C refers to components B and C. Continuity of matter at the interface requires

$$D_B\frac{dC_B}{dx} = -V\left(\frac{1}{k_{jB}} - 1\right)C_{\infty B}$$

and

$$D_C\frac{dC_C}{dx} = -V\left(\frac{1}{k_{jC}} - 1\right)C_{\infty C}$$

so that the condition for stability is

$$G > m_{jB}\frac{V}{D_B}\left(\frac{1}{k_{jB}} - 1\right)C_{\infty B} - m_{jC}\frac{V}{D_C}\left(\frac{1}{k_{jC}} - 1\right)C_{\infty C} \qquad [5]$$

ii) In the past it has usually been assumed without justification that the limit of the planar two phase eutectic (the line between regions 2 and 3, Fig. 2) was given by the constitutional undercooling condition with respect to the third component. This can be justified as follows: in Appendix A it is shown that the undercooling below the two component (A and B) eutectic temperature in a three component (A, B and C) system for planar two phase ($\alpha\beta$) eutectic growth for small $C_{\infty C}$ is given by

$$\Delta T_E = A'V^{1/2} + M_c C_{\infty C}/k_C \qquad [6]$$

where $M_c = dT/dC_C^L$ is the slope of the eutectic valley (assumed constant) and $C_C^E = C_{\infty C}/k_C$ is the average

liquid composition at the interface, $k_C = (k_{\alpha C}s_\alpha)/(s_\alpha + s_\beta) + (k_{\beta C}s_\beta)/(s_\alpha + s_\beta)$ is the weighted eutectic distribution coefficient for component C.

The first term on the right hand side of Eq. [6] is similar to that for a two component eutectic. A' is almost a constant and marginally dependent on the value of $C_{\infty C}$ through M of Eq. [A8a] and [A8b]. The second term on the right hand side of Eq. [6] is similar to that for planar interface growth of a single phase in a two component system where component C is the impurity. A perturbation analysis can thus be carried out in an identical fashion to Mullins and Sekerka[6] for component C ahead of the eutectic assuming A' constant and using an averaged surface energy term. An identical result is obtained except that the denominator will have an additional term due to the first term on the RHS of Eq. [6]. This additional term merely decreases the rate of change in amplitude of the perturbation. The wavelength of the instability will similarly be of the same size as that for the single phase since here A' is effectively a constant, in contrast to Ref. 13. The condition for stability is thus given approximately by the constitutional undercooling condition.

That is, for a planar two phase eutectic interface the line between regions 2 and 3 is given by

$$G > M_c\left(1 - \frac{1}{k_C}\right) C_{\infty C} \frac{V}{D_C} \qquad [7]$$

iii) In Appendix B it is shown, following the order of magnitude model proposed by Burden and Hunt[11] for two component cells or dendrites, that the undercooling ΔT_D for cells or dendrites in a three component system is given by

$$\Delta T_D = \frac{DG}{V} + 2\sqrt{2}\left\{-\frac{aV}{D}\left[m_{\alpha B}(1 - k_{\alpha B})C_{\infty B} + \frac{k_{\alpha B}G_B D}{V} + m_{\alpha C}(1 - k_{\alpha C})C_{\infty C} + \frac{k_{\alpha C}G_C D}{V}\right]\right\}^{1/2}. \qquad [8]$$

This is identical to Burden and Hunt's expression except that an additional $m_{\alpha C}(1 - k_{\alpha C})C_{\infty C} + k_{\alpha C}G_c D/V$ appears as the effect of the third component.

The extent of the planar two phase eutectic range (the line between regions 2 and 5) can be predicted from a competitive growth condition as for a two component using Eqs. [6], [8] and [2].

iv) When the planar interface two phase eutectic breaks down to a cellular two phase eutectic the undercooling will not be given by Eq. [6]. Instead as cells first form the temperature of the cell tip will rise (due to radial rejection of solute C) then decrease again with increasing velocity. By direct analogy with single phase cells or dendrites[11] an expression for the two phase cells or dendrites can be proposed.

In a two component system the dendrite temperature is given by Eq. [3], where the undercooling is measured from the bulk liquidus temperature.[10-12] Modifying this equation by referring the undercooling to the pure metal melting temperature assuming a straight liquidus line gives

$$\Delta T_D^1 = -mC_\infty + \frac{GD}{V} + 2\left[-\frac{2aV}{D}\left(m(1 - K)C_\infty + \frac{KGD}{V}\right)\right]^{1/2}. \qquad [9]$$

The analogous expression for the eutectic cells might be expected to be of the form

$$\Delta T_E = -M_c C_{\infty C} + \frac{GD}{V} + 2\left[-\frac{2aV}{D}\left(M_c(1 - K)C_{\infty C} + \frac{k_C DG}{V}\right)\right]^{1/2} + A^1 v^{1/2}. \qquad [10]$$

Equation [10] reduces to Eq. [6] at the critical condition for planar cell breakdown when inequality[7] becomes an equality. A plot of Eq. [10] at constant gradient is shown schematically in Fig. 4.

In principal Eq. [10] could be compared with Eq. [8] using the competitive growth condition to give the line between region 3 and 5*b*.

v) Assuming a simple interpenetrating three phase lamellar structure such as that shown schematically in Fig. 3(*c*) a theoretical model for three phase growth could be derived in a similar fashion to that derived by Jackson and Hunt[9] for two phase growth. It seems probable that the undercooling ΔT_T would be related to the velocity in a similar fashion to that for the two phase structure

$$\Delta T_T = A''\sqrt{V} \qquad [11]$$

where A'' is a constant depending on the same parameters as A (given previously) but also on the assumed geometry of the phases.

It is proposed that a competitive growth model could then be used with Eqs. [10] and [11] to give the line between regions 3 and 4 and with Eqs. [8] and [11] to give the line between 4 and 5*c*.

It is suggested that stability analyses together with the competitive growth condition provide a basis for at least a qualitative understanding of the different regions in a three component eutectic system. The discussion assumed that each phase grew in a nonfaceted fashion[3] and implicitly that each phase was present in the two phase and three phase structures as an appreciable volume fraction.

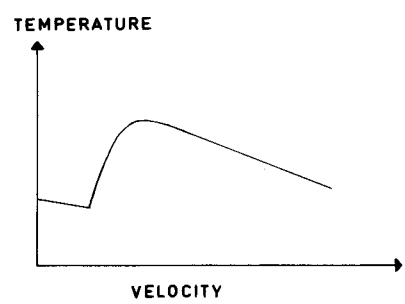

Fig. 4—Schematic variation of the growth temperature with velocity at a constant gradient for a two phase structure. (Eqs. [6] and [10]).

The effect of one phase being faceted in a two component eutectic tends to blur the dividing line between primary faceted phase and the eutectic.[14] The volume fraction and the size of the faceted phase often seems to gradually increase from the eutectic composition. The most probable reason for this is that there is no equivalent well defined primary freezing temperature for the faceted plate or needle. The needles or plates do not usually grow as regular arrays of similar size plates; the primary freezing temperature thus depends on the position and size of the particular growing primary phase. It seems probable tha the regions on Fig. 2 will become less well defined when faceted phases are present in a three component system.

Similar difficulties arise when one phase is present only as a very small volume fraction even though both are nonfaceted. There appears as before to be no sudden transition between well defined eutectic and the presence of primary. Again the reason is probably associated with the difficulty in forming a dendrite array of a very small volume fraction phase.

SUMMARY

1) Five main structural regions are expected during the directional growth of a three component eutectic system (Fig. 2).
2) The limits of the various regions may be predicted or at least qualitatively understood in terms of stability analysis and the competitive growth condition.
3) Models have been proposed for the growth of a two phase eutectic and for single phase dendrites or cells in three component systems.

APPENDIX A
PLANAR TWO PHASE EUTECTIC GROWTH IN A THREE COMPONENT SYSTEM

During steady state planar interface growth of a two phase eutectic in a three component system, the alloy $C_{\infty B}$, $C_{\infty C}$ will grow with solid compositions very near $C^E_{\alpha B}$, $C^E_{\alpha C}$ and $C^E_{\beta B}$, $C^E_{\beta C}$ from a liquid composition near C^E_B, C^E_C where the compositions are the corners of the relevant tie triangle (see Fig. 5). As the steady state growth rate approaches zero the interface temperature, T_I, will approach the liquidus groove temperature T_0, for composition C^E_B, C^E_C. Measuring the undercooling from this temperature gives

$$\Delta T_\alpha = m_{\alpha B}(C^E_B - C_B) + m_{\alpha C}(C^E_C - C_C) + a_{\alpha/r} \quad \text{[A1a]}$$

$$\Delta T_\beta = m_{\beta B}(C^E_B - C_B) + m_{\beta C}(C^E_C - C_C) + a_{\beta/r} \quad \text{[A1b]}$$

where the subscripts α, β refer to phases and B, C to components, $m_{\alpha B} = (\partial T/\partial C_B)_{C\,C}$ similarly $m_{\beta B}$, $m_{\alpha C}$ and $m_{\beta C}$, r is the radius of curvature and a the Gibbs Thomson coefficient for that phase.

Following Jackson and Hunt[9] the compositions in the liquid are given by

$$C_B = C_{\infty B} + B_{oB}\exp(-vz/D) + \sum_{n=1}^{\infty} B_{nB}\cos(n\pi x/S_\alpha + S_\beta)\exp(-n\pi z/S_\alpha + S_\beta) \quad \text{[A2a]}$$

$$C_C = C_{\infty C} + B_{oC}\exp(-vz/D) + \sum_{n=1}^{\infty} B_{nC}\cos(n\pi x/S_\alpha + S_\beta)\exp(-n\pi z/S_\alpha + S_\beta) \quad \text{[A2b]}$$

(Note that Jackson and Hunt defined $C_{\infty B}$ in the two component system as $C_E + C_\infty$). The B_o and B_n terms must now be evaluated. As for the two component system[9]

$$B_{nB} = \frac{2}{n\pi^2}(S_\alpha + S_\beta)\frac{V}{D}C_{oB}\sin(n\pi S_\alpha/S_\alpha + S_\beta) \quad \text{[A3a]}$$

$$B_{nC} = \frac{2}{n\pi^2}(S_\alpha + S_\beta)\frac{V}{D}C_{oC}\sin(n\pi S_\alpha/S_\alpha + S_\beta) \quad \text{[A3b]}$$

where $C_{oB} = C^E_{\beta B} - C^E_{\alpha B}$ and $C_{oC} = C^E_{\beta C} - C^E_{\alpha C}$. The evaluation of the B_o terms is however much more complex. In principal B_{oB} and B_{oC} could be evaluated using the method proposed by Series *et al*[15] for a two component system. From the lever rule

$$\frac{S_\beta}{S_\alpha} = \frac{-(C_{\infty B} - \overline{C}^S_{\alpha B})}{(C_{\infty B} - \overline{C}^S_{\beta B})} = \frac{-(C_{\infty C} - \overline{C}^S_{\alpha C})}{(C_{\infty C} - \overline{C}^S_{\beta C})} \quad \text{[A4]}$$

where $\overline{C}^S_{\alpha B}$ is the average solid α composition of component B similarly $\overline{C}^S_{\beta B}$ and so forth. The average solid compositions may be obtained from the four

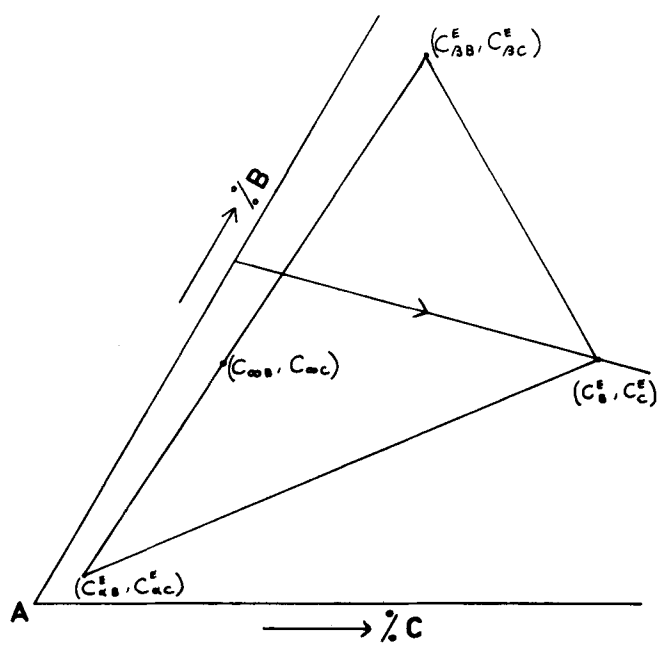

Fig. 5—Three phase tie triangle at a temperature T_0.

expressions of the type

$$\overline{C}^S_{\alpha B} = C^E_{\alpha B} + \frac{m_{\alpha\beta}}{n_{\alpha\beta}}\left(\overline{C}^L_{\alpha\beta} - C^E_B\right) + \frac{m_{\alpha C}}{n'_{\alpha\beta}}\left(\overline{C}^L_{\alpha C} - C^E_C\right) \quad [A5a]$$

where superscript L refers to liquid, $n_{\alpha B} = [(\partial T)/(\partial C^S_B)]_{C^L_\xi}$ and $n'_{\alpha\beta} = [(\partial T)/(\partial C^S_B)]_{C^L_\beta}$. The last terms on the right-hand-side arises because the B content of the solid depends both on the change in the C content of the liquid as well as the change in B. The average liquid compositions may be obtained from Eqs. [A2a] and [A2b] giving four equations

$$\overline{C}^L_{\alpha\beta} = C_{\infty B} + B_{oB} + \phi C_{oB}/S_\alpha D_B \quad [A6a]$$

$$\overline{C}^L_{\alpha C} = C_{\infty C} + B_{oC} + \phi C_{oC}/S_\alpha D_C \quad [A6b]$$

$$\overline{C}^L_{\beta B} = C_{\infty B} + B_{oB} - \phi C_{oB}/S_\beta P_B \quad [A6c]$$

$$\overline{C}^L_{\beta C} = C_{\infty C} + B_{oC} - \phi C_{oC}/S_\beta P_C \quad [A6d]$$

where $\phi = 2(S_\alpha + S_\beta)^2 VP$ and $P = \Sigma_{n=1}^{\infty} \sin^2(n\pi S_\alpha/S_\alpha + S_\beta)/(n\pi)^3$.

The four Eqs. [A6] should now be substituted in Eqs. [A5] and then into [A4] finally giving two equations containing B_{oB} and B_{oC}. These could in principal be solved in terms of B_{oB} and B_{oC} as functions of the ratio $\xi = S_\beta/S_\alpha$. The values of B_{oB} and B_{oC} should then be substituted into the average undercooling equations.

$$\Delta\overline{T}_\alpha = m_{\alpha\beta}(-\Delta C_B - \phi C_{oB}/S_\alpha D_B) + m_{\alpha C}(-\Delta C_C - \phi C_{oC}/S_\alpha D_C) + a^L_\alpha/S_\alpha \quad [A7a]$$

$$\Delta\overline{T}_\beta = m_{\beta B}(-\Delta C_B + \phi C_{oB}/S_\beta D_B) + m_{\beta C}(-\Delta C_C + \phi C_{oC}/S_\beta D_C) + a^L_\beta/S_\beta. \quad [A7b]$$

This procedure is necessary to ensure that the particular value of S_β/S_α is used which allows $\Delta\overline{T}_\alpha = \Delta\overline{T}_\beta$ thus also satisfying heat flow.[15] $\Delta C_B = B_{oB} + C_{\infty B} - C^E_B$ and $\Delta C_C = B_{oC} + C_{\infty C} - C^E_C$ are the differences between the average liquid compositions and the relevant groove composition.

Even in a two component system this procedure is not tractable, Jackson and Hunt[9] proceeded by noting that only $\Delta\overline{T}$ and B_o were sensitive functions of S_β/S_α so that B_o could be eliminated from the two undercooling equations and S_β/S_α then assumed to have its equilibrium value.

A similar procedure may be carried out in a three component system. Writing

$$M_\alpha = m_{\alpha\beta} + m_{\alpha C}\Delta C_C/\Delta C_B \quad [A8a]$$

$$M_\beta = m_{\beta B} + m_{\beta C}\Delta C_C/\Delta C_B \quad [A8b]$$

as the effective three component liquidus slopes on the liquidus surface. Provided $\Delta C_C/\Delta C_B$ is small, (it will be shown later that $\Delta C_C/\Delta C_B \sim C_{oC}/C_{oB}$) M_α and M_β will be relatively insensitive to the precise value of the ratio $\Delta C_C/\Delta C_B$.

Substituting into Eqs. [A7a] and [A7b] gives

$$\Delta\overline{T}_\alpha = -M_\alpha\Delta C_B - (m_{\alpha\beta}C_{oB}/D_B + m_{\alpha C}C_{oC}/D_C)\phi/S_\alpha + a^L_\alpha/S_\alpha \quad [A9a]$$

$$\Delta\overline{T}_\beta = -M_\beta\Delta C_B + (m_{\beta B}C_{oB}/D_B + m_{\beta C}C_{oC}/D_C)\phi/S_\beta + a^L_\beta/S_\beta \quad [A9b]$$

Eliminating ΔC_B gives

$$\Delta T\left(\frac{1}{M_\alpha} - \frac{1}{M_\beta}\right) = -(m_{\alpha\beta}C_{oB}/D_B + m_{\alpha C}C_{oC}/D_C)\phi/S_\alpha M_\alpha - (m_{\beta B}C_{oB}/D_B + m_{\beta C}C_{oC}/D_C)\phi/S_\beta M_B + a^L_\alpha/S_\alpha M_\alpha - a^L_\beta/S_\beta M_B. \quad [A10]$$

The value of the ratio $\Delta C_C/\Delta C_B$ may be estimated using Eqs. [A4] and [A5] writing

$$(C_{\infty B} - C^E_{\alpha B}) = S_\beta C_{oB}, (C_{\infty B} - C^E_{\beta B}) = -S_\alpha C_{oB}$$
$$(C_{\infty C} - C^E_{\alpha C}) = S_\beta C_{oC}, (C_{\infty C} - C^E_{\beta C}) = -S_\alpha C_{oC} \quad [A11]$$

and using Eq. [A4] for the small solid composition changes corresponding to liquid composition changes ΔC_B and ΔC_C

$$S_\beta C_{oB}\Delta C^S_{\beta C} - S_\alpha C_{oC}\Delta C^S_{\alpha\beta} = S_\beta C_{oC}\Delta C^S_{\beta B} - S_\alpha C_{oB}\Delta C^S_{\alpha C} \quad [A12]$$

where $\Delta C^S_{\alpha\beta} = \overline{C}^S_{\alpha\beta} - C^E_{\alpha\beta}$ and similarly ΔC^S_{ij} assuming that the liquid compositions averaged over each phase separately is approximately the same as that averaged over the whole interface Eqs. [A5] give

$$\Delta C^S_{\alpha\beta} = K_{\alpha\beta}\Delta C_B + K'_{\alpha\beta}\Delta C_C$$
$$\Delta C^S_{\beta B} = K_{\beta B}\Delta C_B + K'_{\beta B}\Delta C_C$$
$$\Delta C^S_{\alpha C} = K_{\alpha C}\Delta C_C + K'_{\alpha C}\Delta C_B$$
$$\Delta C^S_{\beta C} = K_{\beta C}\Delta C_C + K'_{\beta C}\Delta C_B \quad [A13]$$

where $K_{ij} = m_{ij}/n_{ij}$ and $K'_{ij} = m_{ij}/n'_{ij}$. This gives

$$\frac{\Delta C_C}{\Delta C_B} = \frac{(S_\beta C_{oC}K_{\beta B} + S_\alpha C_{oC}K_{\alpha\beta} - S_\alpha C_{oB}K'_{\alpha C} - S_\beta C_{oB}K'_{\beta C})}{(S_\beta C_{oB}K_{\beta C} + S_\alpha C_{oB}K_{\alpha C} - S_\alpha C_{oC}K'_\alpha - S_\beta C_{oC}K'_{\beta B})}. \quad [A14]$$

Extensive experimental data must be available to evaluate this expression accurately. In general however $K_{ij} \gg K'_{ij} \to 0$ for most solidus and liquidus surfaces so that

$$\frac{\Delta C_C}{\Delta C_B} \sim \frac{(S_\beta K_{\beta B} + S_\alpha K_{\alpha B})C_{oC}}{(S_\beta K_{\beta C} + S_\alpha K_{\alpha C})C_{oB}} = \frac{K_B C_{oC}}{K_C C_{oB}} \quad [A15]$$

where K_B is the weighted eutectic K value for the B component and K_C that for the C component. Since $K_{\beta B}$, $K_{\alpha B}$, $K_{\beta C}$ and $K_{\alpha C}$ will usually be of similar size as a first approximation

$$\frac{\Delta C_C}{\Delta C_B} \sim \frac{C_{oC}}{C_{oB}}. \quad [A16]$$

Thus for alloys where $K_B \sim K_C$ and $C_{\infty C}$ is small and

thus C_{oC}/C_{oB} is small Eq. [A10] becomes assuming $\Delta C_C/\Delta C_B = C_{oC}/C_{oB}$

$$\Delta T/M = \left(\frac{C_{oB}}{D}\right)(S_\alpha + S_\beta)\phi/S_\alpha S_B - a_\alpha^L/S_\alpha M_\alpha + a_\beta^L/S_\beta M_\beta \quad [A17]$$

where $1/M = 1/M_B - 1/M_\alpha$. Substituting $\xi = S_\beta/S_\alpha$ assuming $D_B = D_C$ and minimizing with respect to lamellar spacing $\lambda = 2(S_\alpha + S_\beta)$ gives a similar expression to that obtained by Jackson and Hunt.[9]

$$V\lambda^2 = a^L/Q_L \quad [A18]$$

$$\Delta T = 2M(a_L Q_L V)^{1/2} = A'V^{1/2}$$

where $a^L = 2(1 + \xi)(a_\beta^L/\xi M_\beta - a_\alpha^L/M_\alpha)$

$$Q_L = P(1 + \xi)^2 C_{oB}/\xi D \quad [A19]$$

The changed sign in a^L arises because Jackson and Hunt[9] defined M_α positive. The approximation, Eq. [A15], produces a more complex expression of similar form.

If the undercooling is measured from the pure binary eutectic temperature assuming that the slope of the liquidus groove is constant

$$\Delta T_E = A'V^{1/2} + M_C C_C^E \quad [A20]$$

where $M_C = dT/dC_C$ is the slope of the eutectic groove and $C_C^E = C_{\infty C}/k_C$ is the relevant liquid groove composition. $k_C = k_{\alpha C}S_\alpha/S_\alpha + S_\beta + k_{\beta C}S_\beta/S_\alpha + S_\beta$ is the weighted eutectic distribution coefficient (note K_{ij} is of similar form but not equal to k_{ij}).

APPENDIX B
SINGLE PHASE DENDRITE GROWTH IN A THREE COMPONENT SYSTEM

Measuring the dendrite undercooling from the liquidus temperature for the bulk alloy $C_{\infty B}$, $C_{\infty C}$, the tip undercooling is given by

$$\Delta T_D = m_{\alpha\beta}(C_{\infty B} - C_{TB}) + m_{\alpha C}(C_{\infty C} - C_{TC}) + \frac{2a}{R} \quad [B1]$$

where C_{Ti} is the tip concentration of either B or C, a is the Gibbs-Thomson coefficient.

Following the order of magnitude model proposed by Burden and Hunt[11] for a two component system, the composition in the liquid is divided into two parts

$$C_B = \overline{C}_B + \delta C_B \quad [B2a]$$

$$C_C = \overline{C}_C + \delta C_C \quad [B2b]$$

where $\overline{C}_i$ is the average composition across the interface and δC_i is the additional part ($i = B$ or C). Eq. [B1] becomes

$$\Delta T_D = m_{\alpha\beta}(C_{\infty B} - \overline{C}_{TB}) + m_{\alpha C}(C_{\infty C} - \overline{C}_{TC}) - (m_{\alpha\beta}\delta C_B + m_{\alpha C}\delta C_C) + 2a/R. \quad [B3]$$

The average compositions $\overline{C}_i$ can be obtained by solving

$$\frac{d^2\overline{C}_i}{dx^2} + \frac{V}{D}\frac{d\overline{C}_i}{dx} = 0$$

giving

$$\left(\frac{d\overline{C}_i}{dx}\right)_{x=0} = \frac{V}{D_i}(C_{\infty i} - \overline{C}_{Ti}). \quad [B4]$$

Between the dendrites away from the tip there is sufficient time for the liquid to become almost homogeneous. Thus the composition gradients along the interface

$$\frac{dC_i}{dx} = \frac{d\overline{C}_i}{dx} \quad [B5]$$

In this region the composition gradients will be almost constant and related to the temperature gradient by

$$G = \frac{dT}{dx} = m_{\alpha B}\frac{dC_\beta}{dx} + m_{\alpha C}\frac{dC_C}{dx} \quad [B6]$$

Making a similar assumption to that made by Burden and Hunt for the two component system, that the average composition gradients behind the interface are similar to the average at the dendrite tips gives assuming $D_B = D_C$ substituting Eq. [B4] into Eq. [B3] using Eqs. [B5] and [B6]

$$\Delta T_D = \frac{DG}{V} - (m_{\alpha\beta}\delta C_B + m_{\alpha C}\delta C_C) + 2a/R \quad [B7]$$

the δC_i terms may now be estimated by considering mass balance at the tip. From Eqs. [B2a] and [B2b]

$$\left(\frac{\delta C_i}{\partial x}\right)_{x=0} = \left(\frac{d\overline{C}_i}{dx}\right)_{x=0} + \left(\frac{\partial(\delta C_i)}{\partial x}\right)_{x=0} \quad [B8]$$

and from mass balance

$$\left(\frac{\partial C_i}{\partial x}\right)_{x=0} = -\frac{V}{D}(1 - k_{\alpha i})(\overline{C}_{Ti} + \delta C_{Ti}) \quad [B9]$$

In the original work it was assumed that

$$\left(\frac{\partial(\delta C)}{\partial x}\right)_{x=0} \simeq \frac{-\delta C_T}{R}$$

which is the classical Zener approximation. It is slightly better to assume that

$$\left(\frac{\partial(\delta C_{Ti})}{\partial x}\right)_{x=0} \simeq \delta C_{Ti}\left(\frac{1}{R} + \frac{V}{D}\right). \quad [B10]$$

This expression is valid for a wide range of Pecklet numbers.

Substituting Eqs. [B9] and [B10] into Eq. [B8] using Eq. [B4] gives

$$\delta C_{Ti}\left(\frac{1}{k} + \frac{k_{\alpha i}V}{D}\right) = -\frac{V}{D}(1 - k_{\alpha i})C_{\infty i} + \frac{Vk_{\alpha i}}{D}(\overline{C}_{Ti} - C_{\infty i})$$

Provided

$$\frac{1}{R} \gg \frac{k_i V}{D}$$

$$\delta C_{Ti}/R = \frac{V}{D}(1 - k_{\alpha i})C_{\infty i} + k_{\alpha i}\left(\frac{d\overline{C}_i}{dx}\right)_{x=0} \quad [B11]$$

Substituting Eq. [B11] into Eq. [B7] gives

$$\Delta T_D = \frac{DG}{V} - \frac{RV}{D}\left[\left(m_{\alpha\beta}(1 - k_{\alpha\beta})C_{\infty B} + \frac{k_{\alpha\beta}G_B D}{V}\right) + \left(m_{\alpha C}(1 - k_{\alpha C})C_{\infty C} + \frac{k_{\alpha C}G_C D}{V}\right)\right] + 2\frac{a}{R} \quad [B12]$$

where

$$G_B = m_{\alpha B}\frac{d\overline{C}_B}{dx} \quad G_C = m_{\alpha C}\frac{d\overline{C}_C}{dx}$$

It is now necessary to apply a minimum undercooling condition

$$\Delta T_D = \frac{DG}{V} + 2\sqrt{2}\left\{-\frac{aV}{D}\left[\left(m_{\alpha B}(1 - k_{\alpha B})C_{\infty B} + k_{\alpha B}\frac{G_B D}{V}\right) + \left(m_{\alpha C}(1 - k_{\alpha C})C_{\infty C} + k_{\alpha C}\frac{G_C D}{V}\right)\right]\right\}^{1/2} \quad [B13]$$

As in the two component dendrite the kGD/V term is only important compared with $m(1 - k)C_\infty$ when the planar interface has just broken down to cells. In this region G_B and G_C will be given to a very good approximation by the values G'_B and G'_C present at the critical breakdown condition. The critical breakdown velocity $V_{\text{crit}} < V$ of alloy $(C_{\infty B}, C_{\infty C})$ in a gradient G is given from Eq. [5] of the main text by

$$V_{\text{crit}} = -GD\Big/\left(m_{\alpha B}\left(\frac{1}{k_{\alpha B}} - 1\right)C_{\infty B} + m_{\alpha C}\left(\frac{1}{k_{\alpha C}} - 1\right)C_{\infty C}\right) \quad [B14]$$

which may be used to give

$$G_B \simeq G'_B = \frac{-m_{\alpha B}(1 - k_{\alpha B})C_{\infty B}}{Dk_{\alpha B}}V_{\text{crit}}$$

$$G_C \simeq G'_C = \frac{-m_{\alpha C}(1 - k_{\alpha C})C_{\infty C}}{Dk_{\alpha C}}V_{\text{crit}}.$$

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