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► To cite this version:

M. Balliel. Transient behaviour of the formation of primary dendrite arms. Journal de Physique IV Proceedings, 1993, 03 (C7), pp.C7-841-C7-846. 10.1051/jp4:19937132 . jpa-00251752

HAL Id: jpa-00251752

<https://hal.science/jpa-00251752>

Submitted on 1 Jan 1993

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Transient behaviour of the formation of primary dendrite arms

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Abstract

In this work a model describing the formation of new primary dendrite arms within a grain or single crystal during directional growth is presented. Therefore it is assumed that the initial distribution of primary dendrite arms has to be rearranged in order to provide locally sufficient space of undercooled interdendritic liquid. A two dimensional arrangement of three equidistant dendrites growing parallel to the imposed temperature gradient is investigated. The idea is to calculate the changes in the undercooled liquid and thereby in the free enthalpy in the free growth zone when the middle dendrite moves laterally in order to provide space for a new dendrite. An activation enthalpy for this mechanism is derived and the rate of formation is estimated. In analogy to the classical nucleation theory for equiaxed solidification the free enthalpy curve as a function of time or process advance exhibits the characteristic energy barrier (the maximum) to overcome and then a drop when a dendrite has formed as this is a step towards the stationary state. Therefore the process may be characterized as a nucleation mechanism.

1. Introduction

In two dimensional experiments Somboonsuk and Trivedi studied the transient behaviour of the directionally solidifying transparent model substance Succinonitrile-Acetone [1]. In one part of their experiments with a constant temperature gradient of about 6.7K/mm they varied the growth rate suddenly from 1.17mm/s to 5.8mm/s. They filmed the growing dendrites with a microscope camera and encountered that the new dendrite tip radius as well as the secondary dendrite arm spacing established very quickly while the primary spacing didn't change over several minutes. With a considerable delay of more than 10min after the acceleration a new primary arm formed out of a tertiary arm. Furthermore the authors provided a diagramme showing the lateral positions of the dendrite tips as a function of time. First, there is a lateral movement due to thermal and concentrational fluctuations and secondly the new primary arm formed in the region where the neighbouring dendrites had a maximum distance from each other. The authors state that the formation of a new dendrite only appears to occur when this distance was about twice the primary spacing. This is also reported in the thesis of H. Esaka [2] where the formation of new dendrites at diverging grain boundaries is studied in the same type of experiment.

The situation that will be investigated in this work is shown in figure 1a where a tertiary arm starts to grow in the middle between two primary dendrite arms. According to the above this is only possible when the distance between both dendrites is twice the primary dendrite arm spacing λ_d . In three dimensions this may occur earlier because of the additional degree of freedom. For a hexagonal arrangement of dendrites at $\sqrt{3}$

times the distance between two dendrites a new primary arm can form at the position of the cross in fig. 1b instead of the circle. In order to be accurate one should note that this is only true when the rotation about $\langle 001 \rangle$ the crystallographic direction of the primary arm permits a secondary arm $\langle 100 \rangle$ or $\langle 010 \rangle$ to grow to the position of the cross. As the assumption of a hexagonal arrangement is already an approximation for the partially random distribution, it is not worth to look at this rotational problem in detail.

Fig. 1a: Formation of a primary arm out of a tertiary arm at $a/2$

Fig. 1b: Hexagonal arrangement of dendrites (dots) and possible new dendrites (circle/cross)

The question is now: How can the favourable situation of such a distance between two dendrites occur?

One possibility is a diverging grain boundary as already treated elsewhere [2]. The second is to increase the temperature gradient G and or growth speed v in a way that the new stationary primary dendrite arm spacing is about half or in three dimensions $1/\sqrt{3}$ the spacing before the change. But this requires a great change as can be calculated by one of the stationary models, i.e. by Kurz-Fisher [3]:

$$\lambda_d = k \times G^{-1/2} \times v^{-1/4} \quad (\text{Eq. 1})$$

If $v = \text{const}$ then G must increase by a factor of 4 or 3 respectively, if $G = \text{const}$ then v must increase by a factor of 16 or 9 respectively. Both values are not achieved during usual directional solidification. The maximum increase in growth speed Somboonsuk and Trivedi treated in the above paper is from 1.17 to 10.7 mm/s which means by a factor of about 9 instead of the required 16. On the other hand they reported a decrease in time to reach the steady state with increasing speed difference.

The third possibility is the lateral movement of the primary dendrite arms due to random fluctuations in the thermal and concentrational field around the tip which will be treated in this work.

2. Model

For reasons of simplicity an arrangement of three equidistant dendrites growing parallel to the imposed temperature gradient G will be discussed (see fig. 2). The idea is now to calculate the changes in the undercooled liquid and thereby in the free enthalpy in the free growth zone region between tip and where the secondary arms first meet each other when the middle dendrite is moved laterally, i.e. to the right. In this model

only the envelope of the secondary arms is taken into account and furthermore it is approximated by straight lines. The tip angle α of these envelope lines is taken as 45° in order to simplify the following calculations, but may have another value which means an additional factor of $1/\tan\alpha$ for the undercooled area (eq. 2a-c).

Fig. 2: Schematic drawing of three equidistant dendrites (A) and after some lateral movement Δ of the middle dendrite (B) during directional solidification. The undercooled liquid in the free growth zone is marked by hatching.

The hatched triangles in figure 2 represent the undercooled liquid in the free growth zone, while the interdendritic liquid within the envelope lines is not taken into account as it is no more undercooled due to segregation and/or heating by liberation of the enthalpy of fusion (if it were undercooled, it would solidify immediately because it is in contact with the secondary dendrite arms).

Calculating the area of these triangles shows already an increase for the case of the lateral movement:

$$F = 1/4 \lambda_d^2 \quad (\text{Eq. 2a})$$

$$F_1 = 1/4 \lambda_{d1}^2 = 1/4 (\lambda_d^2 + 2\lambda_d \Delta + \Delta^2) \quad (\text{Eq. 2b})$$

$$F_2 = 1/4 \lambda_{d2}^2 = 1/4 (\lambda_d^2 - 2\lambda_d \Delta + \Delta^2) \quad (\text{Eq. 2c})$$

Additionally the temperature gradient G has to be taken into account (see fig. 2):

$$G = \frac{T_{Tip} - T_{FG}}{\lambda_d/2} \quad (\text{Eq. 3})$$

$$T(x) = T_{Tip} - G x \quad (\text{Eq. 4})$$

The undercooling ΔT is given by: $\Delta T = T_{liq} - T \quad (\text{Eq. 5})$

The integral average undercooling in the triangles is calculated as follows:

$$\begin{aligned}\Delta T_F &= T_{liq} - \frac{1}{F} \int_0^{\lambda_d/2} (T_{Tip} - Gx) \times 2 \times \left(\frac{\lambda_d}{2} - x\right) dx \\ &= T_{liq} - T_{Tip} + \frac{G}{6} \lambda_d\end{aligned}\quad (\text{Eq. 6})$$

$$\Delta T_{F1} = T_{liq} - T_{Tip} + \frac{G}{6} (\lambda_d + \Delta) \quad (\text{Eq. 7})$$

$$\Delta T_{F2} = T_{liq} - T_{Tip} + \frac{G}{6} (\lambda_d - \Delta) \quad (\text{Eq. 8})$$

The weighted middle of the undercoolings ΔT_{F1} and ΔT_{F2} is:

$$\begin{aligned}\Delta T_{F1,F2} &= \frac{\Delta T_{F1} \times F_1 + \Delta T_{F2} \times F_2}{F_1 + F_2} \\ &= T_{liq} - T_{Tip} + \frac{G\lambda_d}{6} + \frac{G\lambda_d \Delta^2}{3(\lambda_d^2 + \Delta^2)}\end{aligned}\quad (\text{Eq. 9})$$

The difference in undercooling before and after lateral movement:

$$\Delta T' = \Delta T_{F1,F2} - \Delta T_F = \frac{G\lambda_d \Delta^2}{3(\lambda_d^2 + \Delta^2)} \geq 0 \quad (\text{Eq. 10})$$

As $G > 0$ and $\lambda_d > 0$ there is an increase in undercooling associated with the movement. With the well-known equation 11 which is valid for small undercoolings the difference in free enthalpy ΔG associated with the undercooling can be estimated [4]:

$$\Delta G = \Delta S_f \times \Delta T = \frac{\Delta H_f}{T_f} \times \Delta T \quad (\text{Eq. 11})$$

Therefore the increase in free enthalpy $\Delta G'$ due to the movement becomes:

$$\Delta G' = \Delta S_f \times \Delta T' = \Delta S_f \frac{G\lambda_d \Delta^2}{3(\lambda_d^2 + \Delta^2)} \quad (\text{Eq. 12})$$

Differentiating this with respect to Δ gives the force against movement:

$$\frac{d\Delta G'}{d\Delta} = \Delta S_f \frac{2G\lambda_d^3 \Delta}{3(\lambda_d^2 + \Delta^2)^2} [= 0] \quad (\text{Eq. 13})$$

In order to find horizontal tangents to $d\Delta G'/d\Delta$ this is set to zero with the result $\Delta=0$ indicating the fact that the force is 0 when the primary arm is in the center between his neighbours.

The activation enthalpy for the formation of a new primary dendrite ΔG^* can be derived from the condition that the expanded distance λ_{d1} is twice the primary spacing. Therefore the critical movement Δ^* after changing G and or v is given by:

$$\Delta^* = 2 \lambda_{d,n} - \lambda_{d,p} \quad (\text{Eq. 14})$$

Here $\lambda_{d,n}$ is the new primary spacing and $\lambda_{d,p}$ the spacing before the change as calculated by a steady state model (see eq. 1). Hence the activation enthalpy is the value of ΔG^* at $\Delta = \Delta^*$ (see eq. 12):

$$\begin{aligned} \Delta G^* &= \Delta S_f \frac{G \lambda_d}{3} \frac{(\Delta^*)^2}{\lambda_d^2 + (\Delta^*)^2} \\ &= \Delta S_f \frac{G \lambda_d}{3} \frac{(2 \lambda_{d,n} - \lambda_d)^2}{\lambda_d^2 + (2 \lambda_{d,n} - \lambda_d)^2} \end{aligned} \quad (\text{Eq. 15})$$

Figure 3a shows ΔG^* as a function of Δ , while figure 3b shows ΔG^* as a function of $\lambda_{d,n}$. It can be seen that the activation enthalpy is zero for $\lambda_{d,n} = 1/2 \lambda_d$ as $\Delta = 0$. At this value immediate growth of tertiary arms to form new primary arms should occur. Further, this should occur not just at one location, but between any two primary arms resulting in a sharp decrease of the spacing by a factor of about two.

In figure 3 it is also indicated that the lateral movement Δ should be limited to $\lambda_d/3$, respectively $\lambda_{d,n} < 2/3 \lambda_d$ because then the distance between the moved and the right dendrite equals half the expanded distance. Any further movement requires the movement of one or more neighbouring dendrites which causes additional effort.

Fig. 3: (a) Increase in free enthalpy ΔG^* as function of lateral movement Δ
(b) Increase in activation enthalpy ΔG^* as function of new primary spacing $\lambda_{d,n}$

After the formation of the new dendrite the adjacent dendrites will move sideways to equalize the distances to their next neighbours and thereby minimize the free enthalpy as calculated above. This may spread until the lateral movements are in the order of average random fluctuations. This process of equalizing the primary spacing can also be seen in the above mentioned experiments of Somboonsuk and Trivedi [1].

The assumption of thermal activation leads to an Arrhenius equation for the number of new dendrites N^* :

$$N^* = N_0 \times \exp\left(\frac{-\Delta G^*}{RT}\right) \quad (\text{Eq. 16})$$

R is the gas constant and N_0 is the number of potential sites for formation of a new dendrite which is in two dimensions the number of spaces between dendrites or the number of dendrites. In three dimensions using the hexagonal array irrespective of the rotation N_0 equals the number of dendrites, too. It should be noted that this equation is also valid when the stationary state is established. Consequently there is a stationary distribution of gaps between dendrites and eventually new dendrites may form. As the number of dendrites must not increase monotonically in a stationary state, an elimination mechanism is required. This can be achieved for example when a dendrite tip eventually grows slower than the adjacent tips and then is prevented from further growth by the secondary arms of the neighbours.

The rate of formation I can now be calculated with the assumption that the number of new dendrites is small compared to the number of critical gaps (steady-state rate):

$$I = k_1 \times N^* = k_1 \times N_0 \times \exp\left(\frac{-\Delta G^*}{RT}\right) \quad (\text{Eq. 17})$$

This assumption is not very accurate, but it is used here to keep things simple. For a detailed discussion see reference [5].

3. Discussion

A model describing the formation of new primary dendrite arms within a grain or single crystal during directional growth has been presented. This model is based on the changes in free enthalpy due to lateral movement of dendrites. An activation enthalpy for this mechanism is derived and the rate of formation is estimated. In analogy to the classical nucleation theory for equiaxed solidification the free enthalpy curve as a function of time or process advance exhibits the characteristic energy barrier the maximum to overcome and then a drop when a dendrite has formed as this is a step towards the stationary state. Additionally a stationary distribution of critical sites and a rate of formation can be calculated. The secondary arms which have tertiary arms which finally become a new primary arm can be thought of as clusters, then nuclei and finally new dendrites. But during the relatively long process there are always different 'candidate' secondary arms which develop one after the other farther and the final one becomes a new dendrite. However, in the classical nucleation theory for solidification the largest clusters which grow to nuclei and then to grains are also only candidates as they may shrink. Summarizing this, the process can be characterized as a nucleation mechanism.

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