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Distribution of extension rates of growth fronts along Rosiwal's line in the growing two-dimensional cell model

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A growing two-dimensional cell model is defined as follows. In an area there are Poisson-distributed nuclei. Arising from these nuclei, grains start to grow simultaneously. All grains grow circularly with the same constant radial growth rate $\dot{R}$. During the process of growth no new nuclei are formed. If two grains touch each other, growth is stopped there by formation of a straight grain boundary. We arbitrarily put a straight line, called Rosiwal's line, into the area. While grains are growing many straight grain boundaries and circular growth fronts cross Rosiwal's line. At a fixed fraction transformed, F(= crystallized area/total area), we consider the different extension rates of growth fronts (growing borders) along Rosiwal's line, $v(\dot{R} \leqslant v < \infty)$, in the left (or right) direction. The number of grains that have a growth front along Rosiwal's line into the left (or right) direction depends on F. Although the number changes with variation of F, we obtained theoretically the surprising result that the distribution density of reduced extension rates $V = v/\dot{R}$, $w(V)$, does not depend on F, and is always $V^{-2}(V^2 - 1)^{-1/2}$. In order to verify this result we found an experimental possibility to realize the growing two-dimensional cell model.

1. Introduction

At time-point t_0 nuclei are assumed Poisson-distributed in an area with mean nucleus density n, the number of all nuclei in the whole area divided by the area. If $t > t_0$ no new nuclei are formed. Arising from these nuclei, grains start to grow simultaneously. Each grain grows circularly with the same constant $\dot{R}$. Finally, when all grains are grown out, a grain is bounded by grain boundaries having the form of polygons (two-dimensional cell model). These conditions yield the definition of the growing two-dimensional cell model [1].

Fig. 1 shows the picture of an experiment in which the two-dimensional cell model at $F = 1/2$ was realized. Moreover, Fig. 1 shows Rosiwal's line, an arbitrarily placed line, crossed by grain boundaries and (in which we have a special interest) by growth fronts that have the form of circles or parts of a circle with the same grain radius R. The growth fronts extend along Rosiwal's line in the left (or right) direction with different extension rates dependent on the distance between the nucleus and Rosiwal's line (Y) and on R.

Fig. 2 shows the extension rate of a growth front along Rosiwal's line which belongs to a grain with radius R. The nucleus of this grain is a distance Y from Rosiwal's line. All growing grains have by definition the same radius R. Its relationship with F is given by the Avrami relationship $F = 1 - \exp(-n\pi R^2)$, simply deduced by Schulze [2].

In the first part of this paper we show theoretically that the extension rates of growth fronts along Rosiwal's line belong to a distribution density which does not depend on F. In the second part we verify this result experimentally in a very indirect way. We describe the experiment in which we made use of a thin foil of polypropylene to realize the two-dimensional cell model.

2. Theory [3]

2.1. Distribution of V

According to Fig. 2, we obtain

$$x = (R^2 - Y^2)^{1/2}$$

and the extension rate of the growth front along Rosiwal's line $v = \dot{x}$ is for a given Y

$$v = \dot{x} = R\dot{R}/(R^2 - Y^2)^{1/2} = \dot{R}/[1 - (Y/R)^2]^{1/2} \quad (1)$$

The solution for Y yields

$$Y = R(V^2 - 1)^{1/2}/V \quad (2)$$

where $V = v/\dot{R}$ is the reduced extension rate of growth fronts along Rosiwal's line. $\mathrm{d}Y/\mathrm{d}V$ yields, for a fixed R,

$$\frac{1}{R}\mathrm{d}Y = \frac{\mathrm{d}V}{V^2(V^2 - 1)^{1/2}} \quad (3)$$

From Equation 3

$$w(V) = 1/V^2(V^2 - 1)^{1/2} \quad (4)$$

$w(V)$ being the distribution density of the reduced

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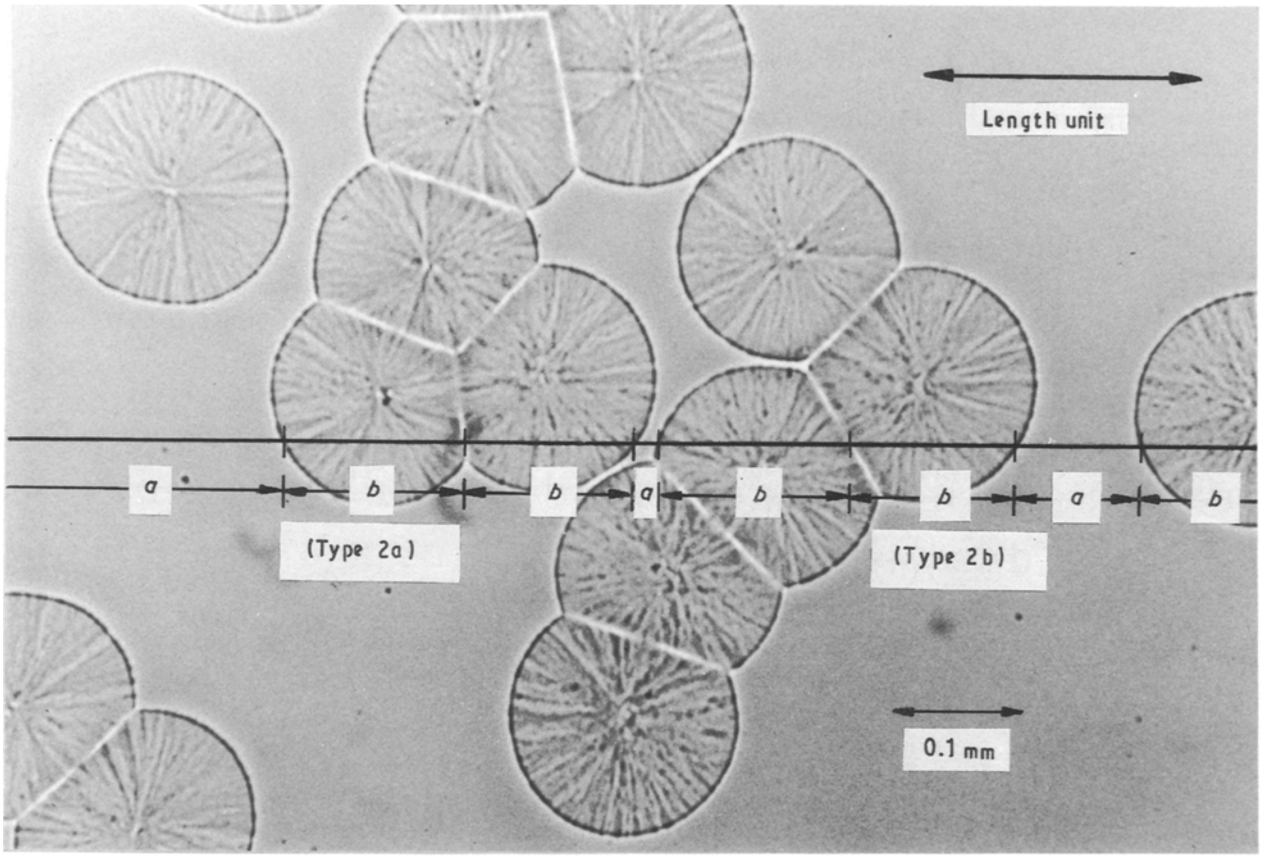

Figure 1. Micrograph of a growing two-dimensional cell model at $F = 1/2$. On Rosiwal's line there are random lengths through the undercooled melt (a) and through the grains (b). b is classified into three types: type 3 limits on both sides on grain boundaries, type 2 limits on one side on grain boundary and type 1 limits on no side on grain boundaries.

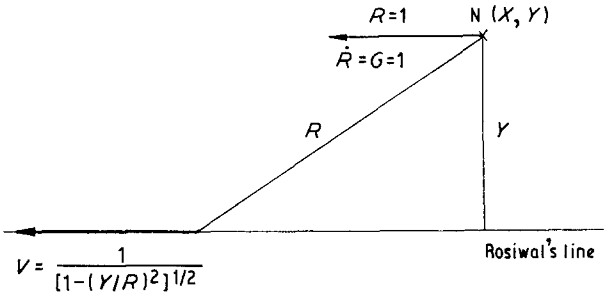

Figure 2. The reduced velocity (V) of a grain of grain radius R and nucleus distance Y from Rosiwal's line. The constant radial growth rate $\dot{R} = G$ is assumed to be unity.

extension rates of growth fronts along Rosiwal's line (V, with $1 \leqslant V \leqslant \infty$).

Fig. 3 shows $w(V)$, and is

$$\int_{V=1}^{\infty} w(V)\,\mathrm{d}V = 1$$

Fig. 3 as a step-diagram with $\Delta V = 0.1$ is given by the dotted line in Fig. 8 (below).

2.2. Nucleus density, q

In order to reduce the following theoretical deductions we introduce a "length unit". We choose one length unit of such a length that n is one nucleus per (length unit)^2.

We will prove that the nucleus density of those nuclei whose grains can grow at a fixed R to the left (or right) direction on Rosiwal's line is constant and is $\exp(-\pi R^2)$. Of course, this constancy of the nucleus density is given only within a band along Rosiwal's line of breadth $2R(-R \leqslant Y \leqslant R)$ and length ∞, otherwise, it is zero.

We call a nucleus type 1 if its grain grows in both directions on Rosiwal's line. A nucleus is called type 2a if its grain grows to the left on Rosiwal's line but to the right it is grown together with another grain. The grains of types 1 and 2a with grain radius R are all of the grains that grow in the left direction on Rosiwal's line.

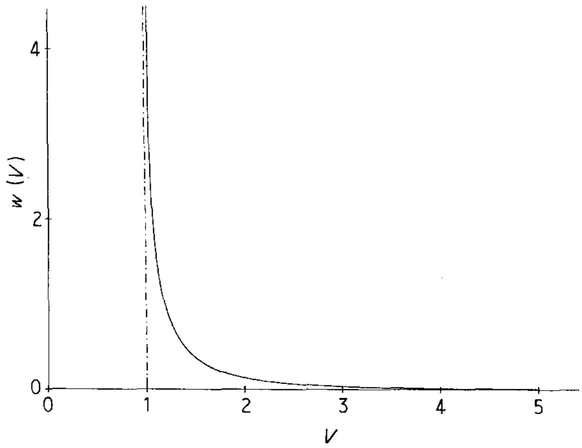

Figure 3. Distribution density $w(V)$ of the reduced rate. The area between V_1 and V_2 (the integral $\int_{V_1}^{V_2} w(V)\,\mathrm{d}V$) gives the fraction of all nuclei that lie between V_1 and V_2.

Fig. 4 shows a grain of type 1 with chord intercept b. The chord intercept can grow if no nucleus lies within the area $S = S_L + S_R = 2S_L$ of Fig. 5. Since by definition the nuclei are Poisson-distributed with density one nucleus per (length unit)^2, the probability of finding no nucleus within area S [2] is

$$q_1(Y; R) = \exp[-S(Y, R)] \quad (5)$$

Therefore, $q_1(Y; R)$ also gives the nucleus density to find a grain of type 1 at $(Y; R)$.

The relationships for a grain of type 2a are shown in Fig. 6. Since the left-hand side of b grows, the area of the circle around x_L must not contain any nucleus. This probability is $\exp(-S_{circle})$. Since the right-hand side of b is grown out, area S_{rest} must contain at least one nucleus. This probability is $1 - \exp(-S_{rest})$. Consequently, the probability of the area

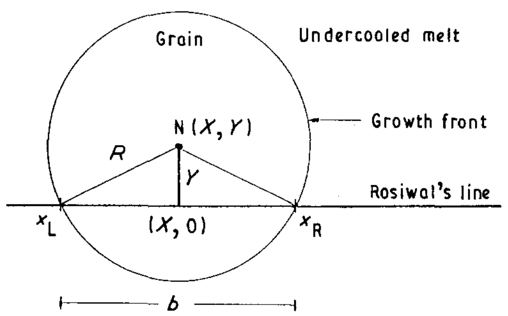

Figure 4. Nucleus $N(X, Y)$ is of type 1. x_L and x_R are the borders between the grain and the undercooled melt. Y is the distance between the nucleus and Rosiwal's line. R is the radius of all growing grains.

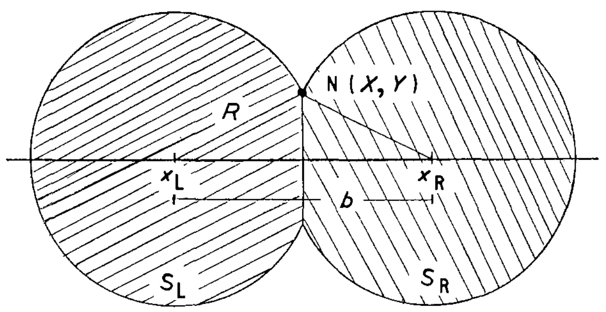

Figure 5. The area $S_L + S_R$ must not contain any nucleus in order for b (type 1) to grow.

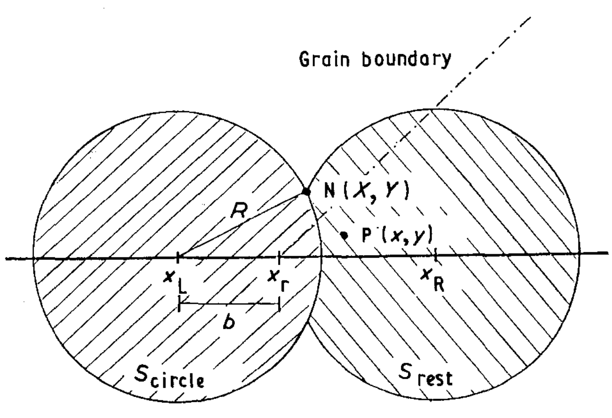

Figure 6. Type 2a yields: area S_{circle} must not contain any nucleus, and area S_{rest} must contain at least one nucleus.

$S = S_{circle} + S_{rest}$ is

$$\begin{aligned} q_{2a}(Y; R) &= \exp(-S_{circle})[1 - \exp(-S_{rest})] \\ &= \exp(-S_{circle}) \\ &\quad - \exp[-(S_{circle} + S_{rest})] \\ &= \exp(-S_{circle}) - q_1(Y; R) \end{aligned} \quad (6)$$

If we consider only the grains possessing a growth front into the left-hand direction on Rosiwal's line, we can calculate the probability of their nucleus density $q_{1+2a}(R)$:

$$\begin{aligned} q_{1+2a}(R) &= q_1(Y; R) + q_{2a}(Y; R) \\ &= \exp(-S_{circle}) \end{aligned}$$

$$q_{1+2a}(R) = \exp(-\pi R^2) \quad (7)$$

Accordingly, the nucleus density of grains that grow in the left direction on Rosiwal's line, q_{1+2a}, depends only on R and not on Y. Therefore, for each Y within $-R < Y < R$, q_{1+2a} is constant, and elsewhere it is zero. The constant nucleus density is equal to $\exp(-\pi R^2)$.

2.3. Number of grains, N

The number (N) of grains that grow to the left-hand side at one length unit on Rosiwal's line in dependence on R, is

$$N(R) = 2\int_{Y=0}^{R}\int_{x=x_0}^{x_0+1} q_{1+2a}(R)\,dx\,dY$$

$$N(R) = 2\exp(-\pi R^2)\int_{Y=0}^{R} dY$$

$$N(R) = 2R\exp(-\pi R^2) \quad (8)$$

Fig. 7 shows $N(R)$. The maximum of $N(R)$ is reached at $R = 0.4 \approx F$.

3. Experiment

We used a foil of polypropylene (from Hoechst AG) of thickness 4 μm and area A of about 15 mm × 35 mm. Its molecular weight was 300 000 and its isotacticy

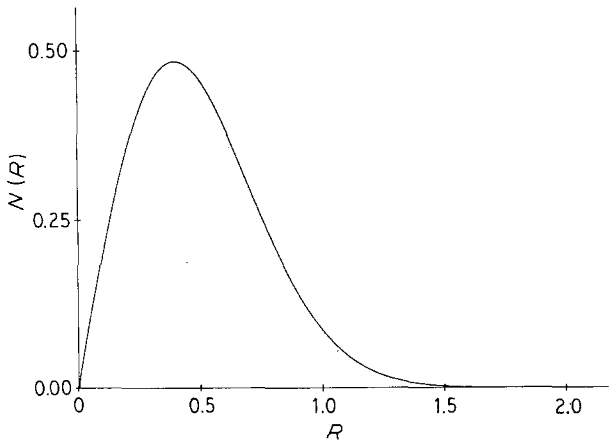

Figure 7. The number of grains $2R\exp(-\pi R^2)$ that grow to the left-hand side on Rosiwal's line referred to one length unit in dependence on R.

96%. No stabilizers existed in the product. In the heat treatment the temperature was decreased from 200 to 138 °C in an undercooled state, because the melting point was 168 °C. Then the temperature was decreased for 1 min to 125 °C, at which temperature nuclei were formed. We then increased the temperature to 138 °C, at which the grains grow but no new nuclei are formed. Fig. 1 shows the growth of spherulites (grains) at $F = 1/2$. Finally, when all grains were grown out, we decreased the temperature to 20 °C. Then we counted the number of all nuclei (M) in area A. Hence, we obtained the nucleus density M/A (number mm^{-2}). In order to normalize to a nucleus density of one nucleus per (length unit)^2, all measured lengths (mm) were transformed by multiplication by the factor $(M/A)^{1/2}$. In the experiment we obtained a mean nucleus density of 7.6 nuclei mm^{-2}. Hence, we calculated 1 length unit $= 7.6^{-1/2}$ mm.

We used an MM 10 microscope in combination with a Nikon micrometer stage to measure to an accuracy of 1 μm the co-ordinates (x, Y) of all nuclei the grains of which reached Rosiwal's line. We then transformed all measured nuclei co-ordinates. A fixed grain radius of $R = 0.4$ length units was chosen because at this R the number of grains that grow in the left direction on Rosiwal's line, $N(R)$, has a maximum. Furthermore, we could distinguish the co-ordinates of nuclei that belong to type 1 and type 2a grains at $R = 0.4$ length units. These co-ordinates (x, Y) are listed in the quantity $|Y|$ with $0 \leqslant |Y| \leqslant 0.4$.

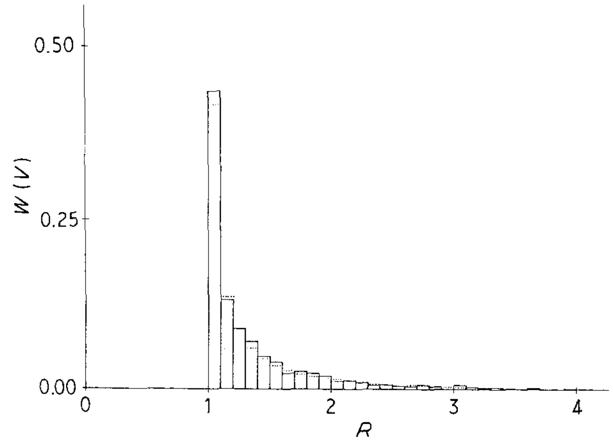

Figure 8. Fig. 3 as a histogram: (· · · ·) theoretical values and (——) experimental values.

From Equation 2 every Y/R is associated with V. The V-intervals 1.0–1.1, 1.1–1.2, . . . , 3.9–4.0 belong to the Y-intervals presented in Table I. We then counted the nuclei that lay within the Y-intervals. We divided this by all nuclei at $R = 0.4$ in order to normalize the counted nuclei, finally obtaining the histogram shown in Fig. 8.

The χ^2-test showed a significance of $\alpha = 10\%$ [4].

TABLE I

V_1	V_2	Y_1	Y_2	ΔY	Measured no. of nuclei, N	$\Delta Y/R$, $R = 0.4$	$N/1680$
1.0	1.1	0	0.1680	0.1680	731	0.4200	0.4351
1.1	1.2	0.1680	0.2229	0.0549	220	0.1262	0.1310
1.2	1.3	0.2229	0.2577	0.0348	150	0.0870	0.0893
1.3	1.4	0.2577	0.2822	0.0246	119	0.0615	0.0708
1.4	1.5	0.2822	0.3006	0.0183	82	0.0457	0.0488
1.5	1.6	0.3006	0.3148	0.0142	68	0.0355	0.0488
1.6	1.7	0.3148	0.3261	0.0113	40	0.0283	0.0238
1.7	1.8	0.3261	0.3353	0.0092	45	0.0230	0.0268
1.8	1.9	0.3353	0.3429	0.0076	40	0.0190	0.0238
1.9	2.0	0.3429	0.3492	0.0063	33	0.0158	0.0196
2.0	2.1	0.3492	0.3546	0.0054	20	0.0135	0.0119
2.1	2.2	0.3546	0.3592	0.0046	22	0.0115	0.0131
2.2	2.3	0.3592	0.3631	0.0039	17	0.0098	0.0101
2.3	2.4	0.3631	0.3666	0.0035	10	0.0087	0.0059
2.4	2.5	0.3666	0.3696	0.0030	12	0.0075	0.0071
2.5	2.6	0.3696	0.3722	0.0026	9	0.0065	0.0054
2.6	2.7	0.3722	0.3746	0.0024	7	0.0060	0.0042
2.7	2.8	0.3746	0.3766	0.0020	12	0.0050	0.0071
2.8	2.9	0.3766	0.3785	0.0019	7	0.0048	0.0042
2.9	3.0	0.3785	0.3802	0.0017	1	0.0043	0.0006
3.0	3.1	0.3802	0.3817	0.0015	11	0.0038	0.0065
3.1	3.2	0.3817	0.3830	0.0013	6	0.0033	0.0036
3.2	3.3	0.3830	0.3843	0.0013	4	0.0033	0.0024
3.3	3.4	0.3843	0.3854	0.0011	5	0.0028	0.0030
3.4	3.5	0.3854	0.3864	0.0010	2	0.0026	0.0012
3.5	3.6	0.3864	0.3874	0.0010	0	0.0026	0.0000
3.6	3.7	0.3874	0.3882	0.0008	3	0.0020	0.0018
3.7	3.8	0.3882	0.3890	0.0008	0	0.0020	0.0000
3.8	3.9	0.3890	0.3898	0.0008	1	0.0020	0.0006
3.9	4.0	0.3898	0.3904	0.0006	3	0.0015	0.0018
					$\Sigma N = 1680$		

4. Summary

The nucleus density of grains that grow on Rosiwal's line in the left-hand direction is constant at a fixed R within $-R \leqslant Y \leqslant R$. For this reason the distribution density $w(V)$ of Equation 4 is also valid in the case of $R = 0.4$ length units. This case was verified experimentally.

For the V-intervals 1.0–1.1, . . . , 3.9–4.0 we obtained Y-intervals in which we counted the measured nuclei. The counted nuclei were divided by the sum of all nuclei the grains of which reached Rosiwal's line in order to normalize them. Fig. 8 shows the theoretical (dotted) and the experimental (drawn) histogram, which are in a good agreement. The χ^2-test showed a significance of $\alpha = 10\%$.

In addition, this has proved that the nuclei of the 15 mm × 35 mm area are Poisson-distributed [5]. This was the supposition for our derivation of $q_{1+2a}(R)$.

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