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Grain size statistics

N. A. HAROUN
College of Engineering, King Abdul Aziz University, Jeddah, Saudi Arabia

A method for the computation of spatial grain size distributions from intercept data based on a tetrakaidecahedron grain model is developed. The necessary inverse matrix is presented. The method is applied to a range of metallic and ceramic specimens. The derived distributions are analysed to show that they are not necessarily log–normal. Statistical techniques are applied to determine the minimum sample sizes and these are shown to increase as the distributions become more dispersed. The constant relating the average grain size to the average intercept length is also shown to be sensitive to the grain size distribution.

1. Introduction

In this article, a number of problems related to the evaluation of grain size and distribution will be tackled. These problems include finding a satisfactory method for the measurement of grain size distributions in a variety of polycrystalline materials, investigation of the nature of these distributions, determination of optimum sample sizes and the relation of mean grain size to the mean intercept length. Apparently, little interest has so far been shown for employing statistical techniques in this field.

2. Matrix for grain size distribution determination

Methods for determining the three-dimensional grain size distributions from planar sections employ either area distributions [1–3] or intercept distributions [4–8]. The former are more tedious. Most of these methods ignore the actual grain shapes and assume them to be spherical [1, 2, 4, 7] or ellipsoidal [8], e.g. Cahn and Fullman's method [7] based on sphere chord distribution data was employed to determine the grain size distributions in MgO [9] and Al_2O_3 [10]. As the spherical grain model is unrealistic [11, 12] successive subtraction techniques based on sectional area distributions [3] and, later, on intercept distributions in a tetrakaidecahedral grain model were developed [11]. However, successive subtraction techniques suffer from the drawback of accumulating errors. The calculation should start with the largest intercept groups. These are usually the few grains in which the relative error is large. Hence, a matrix approach will be developed hereforth.

Intercept length distribution for a tetrakaidecahedral grain was previously shown [11], see Table I. Assume a distribution: $N_1, N_2 \ldots N_8$ of three-dimensional grains of sizes $D_1, D_2 \ldots D_8$. On sectioning, the largest size grains D_1 will contribute $0.365N_1$ intercepts of size l_1, $0.295N_1$ intercepts of size $l_2 \ldots 0.013N_1$ intercepts of size l_8. Similarly, grains of size D_2 will contribute $0.365N_2$ intercepts of size l_2, $0.295N_2$ intercepts of size $l_3 \ldots 0.005N_2$ intercepts of size l_8. Going down the distribution, grains of size D_8 will contribute only $0.365N_8$ intercepts of size l_8. It can thus be shown that the resulting two-dimensional distribution $n_1, n_2 \ldots n_8$ is a product of the two matrices shown in Equation 1:

TABLE I

Relative size range*	0–0.13	0.13–0.18	0.18–0.24	0.24–0.30	0.30–0.42	0.42–0.56	0.56–0.75	0.75–1.00
%	1.3	0.5	2.6	4.7	8.3	17.6	29.5	36.5

*The class means were chosen to decrease in a geometric progression.

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$$\begin{vmatrix} n_1 \\ n_2 \\ n_3 \\ n_4 \\ n_5 \\ n_6 \\ n_7 \\ n_8 \end{vmatrix} = \begin{vmatrix} 0.365 & 0.295 & 0.176 & 0.083 & 0.047 & 0.026 & 0.005 & 0.013 \\ 0 & 0.365 & 0.295 & 0.176 & 0.083 & 0.047 & 0.026 & 0.005 \\ 0 & 0 & 0.365 & 0.295 & 0.176 & 0.083 & 0.047 & 0.026 \\ 0 & 0 & 0 & 0.365 & 0.295 & 0.176 & 0.083 & 0.047 \\ 0 & 0 & 0 & 0 & 0.365 & 0.295 & 0.176 & 0.083 \\ 0 & 0 & 0 & 0 & 0 & 0.365 & 0.295 & 0.176 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0.365 & 0.295 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0.365 \end{vmatrix} \times \begin{vmatrix} N_1 \\ N_2 \\ N_3 \\ N_4 \\ N_5 \\ N_6 \\ N_7 \\ N_8 \end{vmatrix} \quad (1)$$

i.e.

$$A_2 = BA_3, \quad (2)$$

Hence

$$A_3 = B^{-1}A_2, \quad (3)$$

where B^{-1} is the inverse matrix to B, shown in Table II. Hence, three-dimensional distributions can be obtained from two-dimensional distributions by post multiplication of the latter by the inverse matrix given. For computational reasons, the two-dimensional intercepts should be classed in groups 0.75 to 1, 0.56 to 0.75 . . . 0 to 0.13 of the maximum measured intercept length.

The ability of this technique to reproduce excellently the three-dimensional distributions is demonstrated in Fig. 1, where a hypothetical intercept distribution based on a single-size Kelvin grain model was used to reproduce the three-dimensional (mono-size) distribution. It is envisaged that the matrix in Table II can be fed to a microprocessor interphased with an automatic counting microscope to measure directly the spatial grain size distribution.

2. The nature of grain size distributions

Grain size distributions control the kinetics of grain growth [13], the relation between the average grain size and intercept length measurements [14] as well as the mechanical behaviour of materials. It was shown that rather than depending on $1/D$, the rate of grain growth should depend on the rate of disappearance of grains [13], which is a function of both the average grain size and the grain size distribution. Microcrack formation during cooling brittle materials is possible only for grains larger than a critical size [15] and hence should depend on the grain size distributions; a consideration which is important in fracture studies.

In the literature, only a few determinations of the grain size distributions have been carried out so far, some of which were merely two-dimensional [16–17]. Examples are Feltham's [16] log-normal and Hillert's [17] skewed log-normal distributions. Spatial distributions in MgO were shown to be log-normal during normal grain growth but not otherwise. Similarly, Al_2O_3 showed log-normal spatial distributions only in the presence of uniformly dispersed MgO additive [10]. Theoretically, it was proposed that the type of grain size distribution should not remain constant, but would follow cycles of one-peak and two-peak distributions [56]. Measurements of the grain size distributions of some hot-pressed MgO specimens did not conform to simple distribution functions [4]. To sum up, it can be suggested that strictly log-normal distributions could be the exception rather than the rule.

A variety of typical polycrystalline specimens shown in Table III were selected for the study.

Linear intercepts measured on photomicrographs, were grouped in classes 0.75 to 1, 0.56 to 0.75 . . . etc. of the maximum intercept length. Three-dimensional distributions were computed therefrom as explained above. A typical example is shown in Fig. 2 for the microstructure in Fig. 3.

TABLE II

2.740	− 2.214	0.469	0.066	− 0.129	0.055	0.062	− 0.157
0	2.740	− 2.214	0.469	0.066	− 0.129	0.055	0.062
0	0	2.740	− 2.214	0.469	0.066	− 0.129	0.055
0	0	0	2.740	− 2.214	0.469	0.066	− 0.129
0	0	0	0	2.740	− 2.214	0.469	0.066
0	0	0	0	0	2.740	− 2.214	0.469
0	0	0	0	0	0	2.740	− 2.214
0	0	0	0	0	0	0	2.740

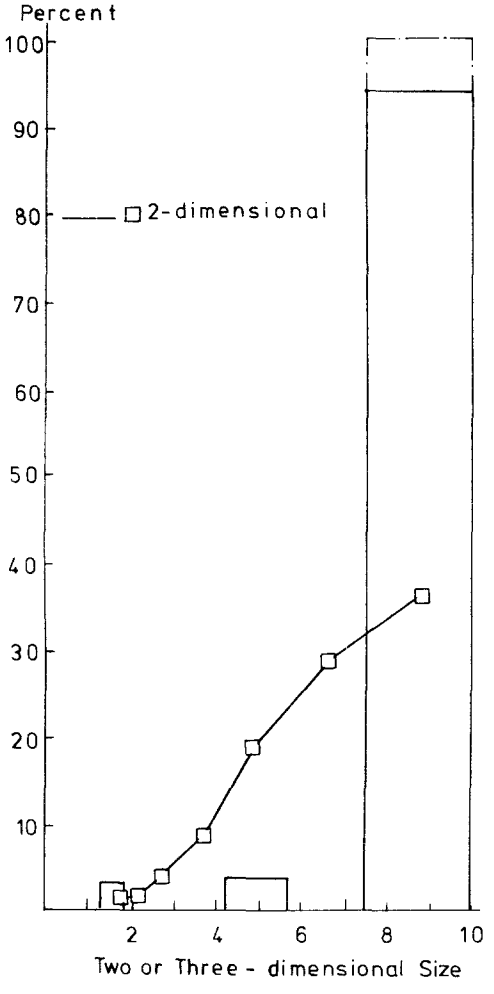

Figure 1 Two- and three-dimensional distributions in a hypothetical mono-sized specimen.

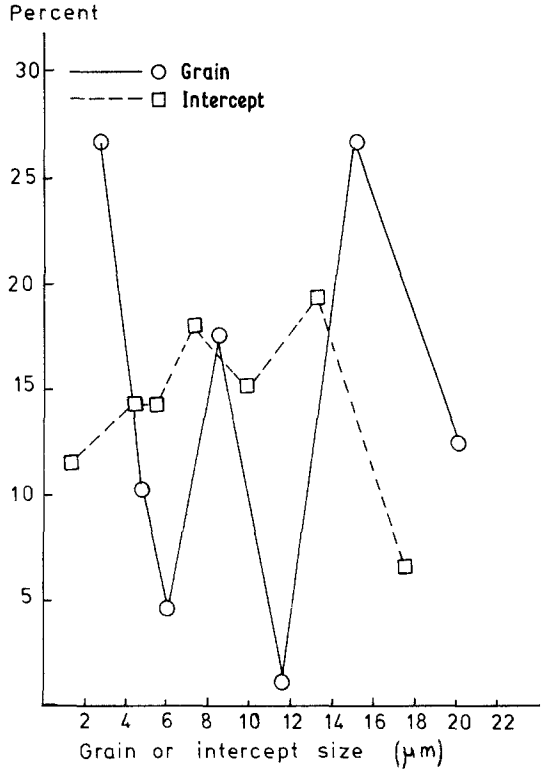

Figure 2 Microstructure of MgO.

The spatial distributions derived by the matrix approach are somewhat different from those obtained by successive subtraction.

The distributions were plotted on log-probability paper, and least-square fit straight lines were obtained, e.g. Fig. 4. If the distributions were log-normal, they should satisfy the criteria of: (i) quality of fit to straight lines, measured by a correlation coefficient $r = (\Sigma XY)/(\Sigma X^2 + \Sigma Y^2)$

TABLE III

Specimen number	Composition–treatment	Source or reference
1	Steel, 0.1% C, full anneal	"Buehler", USA
2	Pure iron	[13]
3	Tough pitch copper, 0.04% O	"Met. Serv", England
4	Brass, 59% Cu–41% Zn	"Struers", Denmark
5	Al_2O_3–0.25 w/o NiO, 1675° C for 40 h	Prepared [18]
6	Duralumin	[11]
7	MgO, vacuum, hot pressed, annealed 1300 and 1400° C	Prepared [19]

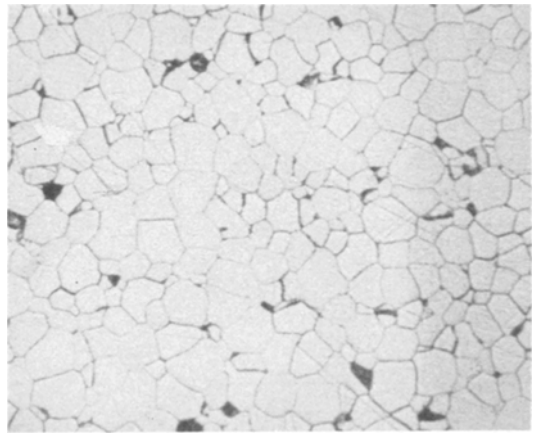

Figure 3 Intercept and derived grain size distribution of MgO.

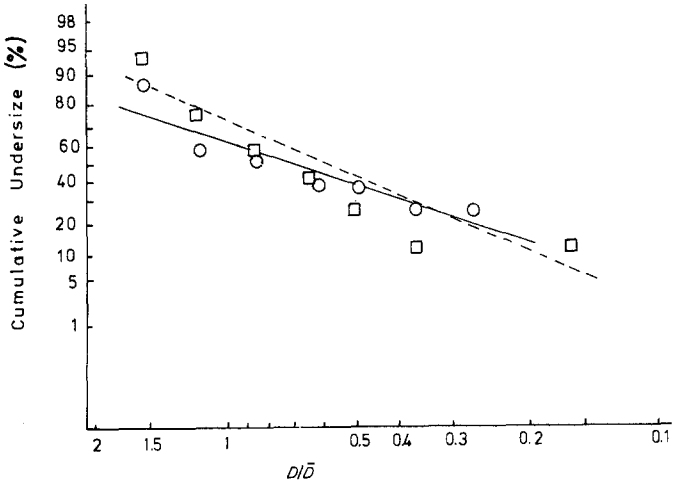

Figure 4 Cumulative probability plots of distributions.

[20]. The results shown in Table IV suggest that although some intercept distributions conform to log-normality, none of the derived three-dimensional distributions can be described as log-normal. While the log-normality of some intercept distributions is a natural consequence of the randomness of intercepts, the evolution of three-dimensional distributions should be more complex and depends on the detailed history of grain growth. It can be concluded that grain size distributions are not necessarily log-normal.

4. Sample sizes for grain size measurements

In most of the literature on grain growth [9, 10, 19, 21–45] there seems to be little concern to assess whether the sample sizes employed were statistically adequate. In ASTM 112-47 [46], it was recommended to increase the sample size if the 95% confidence limits of a measurement seems too large.

As the representative sample sizes could be sensitive to the type of grain size distribution, statistical techniques were applied to evaluate the critical minimum sample sizes for the specimens in Table III. For a small sample of 100 intercepts, a preliminary estimate of the sample size n_i [47] at 95% confidence level, was taken as

$$n_i = \frac{1.96\sigma_i}{\delta}^2 \quad (2)$$

with σ_i being the preliminary sample standard deviation, and δ the maximum deviation tolerated and taken as 10% of the sample mean. Two independent samples whose size is the estimated n_i were taken. The significance of the difference between their means was evaluated by a Z-test [20] at the 95% significance level, and their standard deviations were compared by an F-test at the 95% significance level [47]. The sample size was subsequently either increased or decreased (in multiples of ten-steps) and the procedure was repeated until a critical sample size was reached. Finally, two grain size distributions (of the critical size) were compared by a χ^2-test [20] at a 95% significance level. The final sample sizes arrived at are shown in Table V, as well as the dispersions

TABLE IV

Specimen number	Three-dimensional		Two-dimensional	
	r	Cumulative probability at $\bar{D}$	r	Cumulative probability at $\bar{l}$
1	0.79	72%	0.81	62%
2	0.76	50%	0.82	58%
3	0.81	70%	0.82	57%
4	0.78	72%	0.83	54%
5	0.78	83%	0.77	60%
6	0.71	72%	0.77	53%
7	0.82	62%	0.76	73%

TABLE V

Specimen	Minimum sample size	Dispersion (two-dimensions)
Single size hypothetical	30	0.32
1	220	0.79
2	60	0.34
3	200	0.85
4	120	0.69
5	65	0.40
6	100	0.54
7	50	0.52

of the distributions. The dispersion is taken as the coefficient of variation = (standard deviation/mean intercept size) [20], which is a more representative expression than the standard deviation. The single-size samples in Table V were obtained by operating random numbers on a list of serial numbers (1–999) assigned to different groups of the hypothetical intercept distribution. It can be concluded that the minimum sample sizes should range between 30 and 220 measurements, and generally increase with the dispersion of the distribution.

5. The average grain size $\bar{D}$ against the average intercept length $\bar{l}$

The linear intercept model has been accepted as the most reliable and straightforward method for determining the average grain size, as well as the ASTM grain size number [46]. A relation of the type $\bar{D} = K\bar{l}$ is usually employed. The constant K depends on:

(i) the assumptions made for the grain shape and size distribution.

(ii) the definition of a grain size or "diameter".

For uniformly spherical grains, $K = 1.5$ [48], or in a slightly different derivation [49] $K = 1.62$. Spheres are not space-filling bodies and since $S_v = 2/\bar{l}$ [50], it was shown that for uniform tetrakaidecahedral grains $K = 1.68$ [51] or $K = 1.776$ [10]. For a slightly modified grain model [52], it was shown that $K = 1.74$ [53] or 1.78 [54]. However, the presence of a distribution of grain sizes should be considered. Mendelson [14] has shown for a log-normal distribution that: $K = 1.775e - 2.5 \ln^2$, where the grain size was defined as the caliper diameter. Applied to the log-normal [16] and the skewed log-normal distributions [17] this led to $K = 1.558$ and 1.570, respectively. Experimental evaluation of K as the ratio of the mean volume surface grain size to $\bar{l}$, has shown these to be lower than those calculated according to the Mendelson [14] equation. The difference was attributed to errors in the Cahn and Fullman [7] method which overemphasizes the smaller sizes. Abnormal distributions led to K values higher than calculated.

In this investigation, K was determined experimentally as the ratio of the average derived grain size to the average intercept length. The grain size was taken as the caliper dimension [14] and equals 2.45 times the edge length of a tetrakaidecahedral grain model which is 0.82 times the maximum intercept on that model [55]. As shown in Table VI, K varied between 0.95 and 1.26 and, as expected [9, 14, 49], seems to vary with the grain size distribution. On the other hand, if the grain size is taken as the maximum caliper dimension (circumdiameter), K ranges between 1.16 and 1.54. These, as well as previous results, suggest that a good deal of disagreement exists on what values of K should be taken; a situation accentuated by the wide variations in grain size distributions between different materials differently heat treated and by the different definitions of the term "grain size". It seems that reasonable accurate grain size data can be only obtained through grain size distribution determinations on lines similar to those described in this article.

TABLE VI

Specimen	K ($= \bar{D}/\bar{l}$), based on	
	Average caliper dimension	Circumdiameter
Single size hypothetical	1.21	1.48
1	1.02	1.24
2	1.17	1.42
3	0.97	1.19
4	1.11	1.36
5	1.23	1.50
6	1.26	1.54
7	0.95	1.16

6. Conclusion

The spatial average grain size cannot be inferred directly from the average intercept size, as they are related with a "constant" that depends on the nature of the grain size distributions. These were shown to be variable and not necessarily log-normal. Besides, the minimum representative sample size also varies. Hence, the procedure recommended is to measure about 500 intercepts, and compare two of these (250 each) using at least a Z-test as described in the text. If no significant difference is found (otherwise increase the sample size), the spatial grain size distribution is computed employing the inverse matrix and the technique given.

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