ACTA STEREOL 1999; 18/1: 81-88
ORIGINAL SCIENTIFIC PAPER

USEFULNESS EVALUATION OF THE STEREOLOGICAL METHODS APPLIED FOR GRAIN SIZE ESTIMATION

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ABSTRACT

The computer simulated 3D models of one–phase polycrystalline materials were applied for assessment of precision and versatility of hitherto used methods for grain volume distribution. The precise quantitative description of model structures of sufficient grain size and shape inhomogeneity in 2D and 3D spaces enabled the unequivocal comparison of the results of grain size evaluation obtained with:

- classical method: Scheil–Schwartz–Saltykov's, Saltykov's, Spector's, Saltykov's method of inverse diameter,
- empirical equations: Hanson's, Nuñez-Domingo's, Okada–Katsulai–Oka's,
- modern stereology methods: point sampled intercepts and disector.

A basic criterion of the evaluation was a relative error of stereological parameters obtained with these methods. It was shown that method ensuring the required versatility and accuracy of grain size evaluation at acceptable labour consumption and cost had not been worked out yet.

Key words: grain size, 3D modelling, stereology.

INTRODUCTION

In polycrystalline materials grain size is a parameter of hitherto not agreed and universal definition. Well defined grain size should correlate with properties depending upon it. Parameter used for its determination should be obtainable through measurements of metallographics microsections.

Nowadays volume is considered as the most suitable and natural measure of size of grains as spatial objects (Kurzydłowski and Ralph, 1995). But grains' volume can not be determined on the basis of measurements performed on plane sections. Therefore it is necessary to employ indirect methods. In practice grain size is described through the relevant distribution's parameters: mean grain volume and coefficient of variation of grains volume. Mean grains volume in case of single-phase materials is usually replaced with a number of grains per unit volume $N_{1.}$.

METHODS OF GRAIN SIZE EVALUATION OF POLYCRYSTALLINE MATERIALS

The methods of $N_{1.}$ determination, most commonly applied in materials science, are collected in Table 1. It should be emphasized that the majority of the presented relations were

Table 1. Characterization of the stereological methods for particles and grains size estimation.

Method for grain size estimation
1. Scheil-Schwartz-Saltykov's method $(N_V)_j = \frac{1}{\Delta}[\alpha_i (N_A)_i - \alpha_{i+1}(N_A)_{i+1} - \alpha_{i+2}(N_A)_{i+2} - \ldots - \alpha_k (N_A)_k]$ $[mm^{-3}]$ $i=1..k$, $j=1..k$, $k_{max}=15$ k – number of class intervals of empirical distribution of plane sections and of spheres. For each „j" „i" varies from $i=j$ to k $\alpha_i, \alpha_{i+1}, \alpha_{i+2}\ldots$ – coefficients $\Delta=D_{max}/k$ – length of class interval
2. Saltykov's method $(N_V)_j = \frac{1}{D_j}[1.6461(N_A)_i - 0.4561(N_A)_{i-1} - 0.1162(N_A)_{i-2} - 0.0415(N_A)_{i-3} +$ $- 0.0178(N_A)_{i-4} - 0.0079(N_A)_{i-5} - 0.0038(N_A)_{i-6} - 0.0018(N_A)_{i-7} - 0.0010(N_A)_{i-8} +$ $- 0.0003(N_A)_{i-9} - 0.0002(N_A)_{i-10} - 0.0002(N_A)_{i-11}]$ $[mm^{-3}]$ j – number of sphere size (i=1 for the largest spheres) i – number of class interval of relative plane sections area's distribution A/A_{max} Subscripts of N_A vary from $i=j$ through i-1, i-2, ... up to 1.
3. Spector's method $(N_V)_j = \frac{4}{\pi\Delta^2}\left[\frac{(N_L)_i}{2i-1} - \frac{(N_L)_{i+1}}{2i+1}\right]$ $[mm^{-3}]$ $\Delta = l_{max}/k = D_{max}/k$ Δ – length of class interval k – number of class intervals i – number of class interval of chords $(N_L)_i$ – number of chords per unit length of test line in the „i-th" class interval $(N_V)_j$ – number of spheres (with mean diameter $j\Delta$) per unit volume, for the indicated j-size
4. Saltykov's method of inverse diameters $N_V = \frac{2}{\pi}\sum_i d_i^{-1}(N_A)_i$ $N_V = 0{,}6366\,\overline{d^{-1}}\,N_A$ $[mm^{-3}]$ $\overline{D} = 1{,}5788(\overline{d^{-1}})^{-1} = \frac{N_A}{N_V}$ $[mm]$ $\sigma(D) = \sqrt{\frac{4}{\pi}\overline{d}\,\overline{D} - (\overline{D})^2}$ N_V – number of spheres per unit volume $\overline{D}$ – mean diameter of spheres $\sigma(D)$ – standard deviation of sphere diameters

established after making certain initial assumption, most often concerning a spherical shape of grains. It is generally accepted that such an assumption is permissible in case of polycrystalline materials of equiaxial grains (Underwood, 1970).

Empirical equations derived from the results of the study of 3D computer simulated model structures by Hanson (1979) and Okada et al. (1990) as well as this established analyticaly by Nuñez and Domingo (1988) are often used for this purpose. In general, N_V is proportional to parameters that can be determined from plane section, such as N_L and N_A or combination of them. The mentioned above equations applied for N_V or $E_N(V)$ calculations are presented in Table 2.

The methods of modern stereology are succesfully applied in biomedical science (Gundersen et al. 1988a, 1988b). Sparse, so far, papers demonstrating the possibilities of using new stereological methods in materials science (e.g. Kurzydłowski and Ralph 1995, Karlsson and Cruz-Orive 1992) do not let estimate their suitability for a quantitative description of polycrystalline materials microstructure. Therefore, the point sampled intercepts method and disector were chosen for investigation as the most promissing methods.

Table 2. Characterization of equations for N_V and $E_N(V)$ estimation.

Equations for grain size estimation
1. Hanson's empirical equation $N_V = \left(\frac{2{,}4150}{\sqrt{N_A}} - \frac{1{,}4552}{N_L} \right)^{-3} \quad [mm^{-3}]$ N_V – number of grains per unit volume N_A – number of grain sections per unit area N_L – number of intercepts per unit length of test line
2. Nuñez-Domingo's equation $N_V = \frac{6}{\pi}\left(\frac{4}{K_S}\right)^3 N_L^3 = 122{,}3\left(\frac{N_L}{K_S}\right)^3 \quad [mm^{-3}]$ N_L – number of intercepts per unit length of test line K_S – dimensionless shape factor $K_S = (S/V)\,d_e$ S, V – surface area and volume of grain or particle d_e – equivalent diameter of grain $d_e = (6V/\pi)^{1/3}$
3. Okada–Katsulai–Oka equations $E_N(V) = K_3\left(8\sqrt{6}\,\frac{E(l^3)}{\pi^2}\right) \quad [mm^3]$ $E(D) = K_2\left(\frac{2\sqrt{6}}{\pi}\,E(l)\right) \quad [mm]$ $E_N(V)$– mean volume of grains $E(D)$ – mean equivalent diameter of grains $E(l)$ – mean length of grain plane section chords

The point sampled intercepts method allows for an unbiased estimation of volume-weighted mean grain volume according to the equation (Jensen and Gundersen, 1985):

$$E_V(V) = \frac{1}{n}\frac{\pi}{3}\left[l_0^3 + 2\sum_{i=1}^{k} \left| l_{i+}^3 - l_{i-}^3 \right| \right] \tag{1}$$

where: n - number of random points,
l_0 - intercept length containing the sampling point,
l_{i+}, l_{i-} - distances from sampling point to the ends of sections of n-th intercept segment.
Graphic interpretation of symbols is presented in Fig. 1. This estimator is true for particles of arbitrary shape. For convex particles the equation (1) can be reduced to:

$$E_{V'}(V) = \frac{\pi}{3}\overline{l_0^3} \tag{2}$$

where: l_0 - intercept length crossing the sampling point.

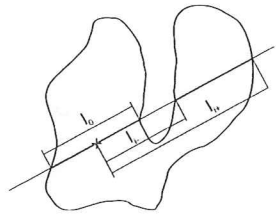


Fig. 1. Method of intercepts length measurement.

The only requirement of the method presented is that the intercept length should be measured on a secant of random orientation in relation to the particle. To put it more precisely, the orientation of the straight line must meet the requirement of isotropic uniform random (IUR) orientation towards the particle, so each orientation must be equally probable (Gundersen, 1986).

The point sampled intercepts method can also be used to estimate the volume-weighted coefficient of variation of grain volume. A merit of the point sampled intercepts method is, from the point of view of its application in materials science, the possibility of determining parameters of volume-weighted distribution of grains volume.

The main sampling method in case of polycrystalline materials is cutting the material with one random or oriented plane and making a metallographic specimen. The probability of cutting grains with the cutting plane is proportional to the size of grains, and to be more specific – it is proportional to the height of grains in a direction perpendicular to the plane. Therefore larger grains are cut more often than smaller ones. This problem is solved by disector which is simultaneously a way of sampling and determining $N_{V'}$. This method belongs to the group of methods in which two plane sections of tested material are used (Sterio, 1984).

In disector method $N_{V'}$ is calculated from the formula:

$$N_{V'} = \frac{\sum_{i=1}^{n} Q_i^-}{\sum_{i=1}^{n} a_i \cdot h} \tag{3}$$

where: a_i - measuring frame area, h - distance between reference plane and look–up plane.

Disector is a method, the rule of which is relatively simple. This method has a widespread application in biology and medical science. However, disector application in materials science is connected with very laborious and time–consuming studies. Besides, the algorithms of automatic recognition of parallel sections coming from the same grain have not been defined so far.

EXPERIMENTAL PROCEDURE

The investigations were based on three types of computer simulated in 3D space models of one–phase granullar material microstructures: Williams, cell and Johnson-Mehl model structures. Algorithms of generation and characteristic of these structures is presented elsewhere (Chrapoński 1997, Chrapoński and Maliński 1997b, Maliński and Chrapoński 1997). Plane section area of grains as well as intercepts length of grain sections were determined on random plane sections of model structures N_A and N_L values were calculated. Shape factor K_S for Nuñez–Domingo relation was computed through accurate date obtained numerically in simulation procedures.

In order to estimate the volume–weighted mean grains volume by means of point sampled intercepts method an add–on module for image analysis software VISILOG 4.1 system has been worked out. This module performs the procedure of measurement of random oriented intercepts crossing the points which are randomly located on the microstructure image.

Grains counting in disector method was carried out on parallel sections of model structures distant from each other at 20 to 80% 3D equivalent diameter to the smallest grain. Since there have been presented neither algorithms nor programmes of automatic image analysis by means of disector method so far, it was necessary to eliminate subjectivism of studies resulting from a visual assessment of parallel sections. For this purpose programmes facilitating grain counting for model grain structures have been worked out according to disector method. This counting is error–free since each grain is unambiguously identified in 3D space and each grain plane section is ascribed to a specific grain.

Usefulness of stereological methods applied to description of grain size was evaluated through comparison of results obtained from measurements on plane section of model structures with corresponding values of stereological parameters calculated numerically with simulation programs (these value will be designated as "true value").

A basic criterion of the evaluation was a relative error RE calculate as follows:

$$RE = \frac{|V_T - V_M|}{V_T} \tag{4}$$

where: V_T – true value of a parameter,

V_M – parameter's value obtained with an evaluated method.

A relative error calculated according to formula (4) includes all factors, occurring at various stages of investigations, affecting a final results of each method. Homogeneity of methodics of measurements performed on models structures' plane sections allows to accept the relative error as a parameter suitable for comparison of results obtained using different methods.

The results of comparison of true and reproduced distributions was additional criterion evaluating the usefulness of methods used to determine distributions of diameters of equivalent sphere's system. 3D and 2D parameters were partitioned into classes in compliance with recommendations of the evaluated methods. True distributions of diameters of equivalent spheres' system were formed with similar classes' partition as that of the reconstructed distributions. Kolmogorov–Smirnov test of goodness of fit was used to compare both kinds of distributions. For all distributions compared with this statistical test, a critical value of Kolmogorov–Smirnov distribution at significance level $1-\alpha=0.95$ amounts $\lambda=1.358$.

RESULTS AND CONCLUSION

The analysis of relative errors of stereological parameters estimation by evaluated methods collected in Table 3 as well as the results of comparison of true and reproduced distributions

Table 3. Relative error of stereological parameters estimation by evaluated methods.

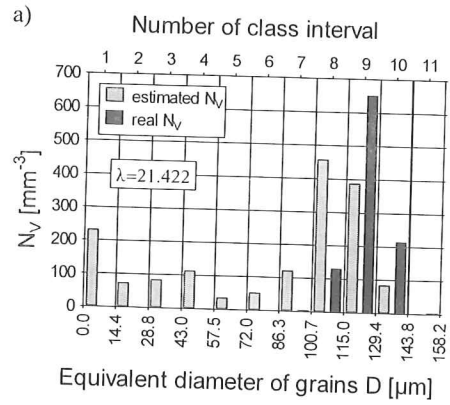
Method	Model structures										
	W1	W2	W3	W4	W5	C1	C2	C3	JM1	JM2	JM3
	Relative error [%]										
	N_V										
1	49,7	40,1	58,8	52	51,8	67,1	67,1	73,3	65,2	45,7	28,7
2	84,2	80,8	91	68,2	81,7	102,9	103,7	110,2	124	779,7	76,4
3	292,2	152,3	327,7	370	358,8	486,9	466,2	388,1	206,4	170,4	89,9
4	48	32,9	59,2	48	51,4	67,8	82,1	82	70,7	49	54,6
	N_V										
5	29,8	27,2	20,6	14,6	24,2	11,3	15,7	22,5	27	21,4	59
6	38,8	62,8	48	27,8	42,1	3,1	3,5	4,5	76,9	63,8	87,5
	$E_N(V)$										
7	47,3	141,7	61,1	18,5	38	13,3	10,1	11,5	11,8	42	121
	$E_V(V)$										
8	8,7	5,8	5,8	5,3	3,2	4,9	5,4	10,1	3,1	13,3	32
	N_V										
9	1,5	5,6	2	1,5	11,2	1,2	2,9	0,4	6,4	9,2	20,3

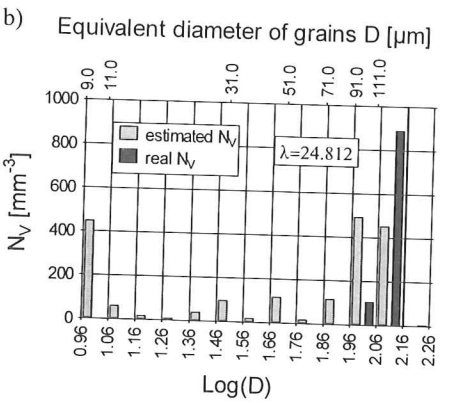
W1÷W5 - Williams' structures, C1÷C2 - cell structures, JM1÷JM3 - Johnson-Mehl structures;
Classical stereological methods: 1 – Scheil–Schwartz–Saltykov's, 2 – Saltykov's, 3 – Spectors', 4 – Saltykov's method of diameters' inverses; Empirical equations: 5 – Hanson's, 6 – Nuñez–Domingo, 7 – Okada–Katsulai–Oka; 8 – Point sampled intercept; 9 – Disector.

of equivalent diameters of grains (Chrapoński, 1997) on Fig. 2 leads to the following conclusions:

- classical stereological methods: Scheil–Schwartz–Saltykov's, Saltykov's, Spectors' and Saltykov's method of diameters' inverses work out for quantitative assessment of polydispersive system of spheres and recommended in some basis stereological literature for grain size evaluation – that is while widely applied in the study of polycrystalline materials – do not assure correct estimation of grain volume distribution as well as of the parameters of this distribution.
- empirical equations being used for calculation of number of grains per unit volume N_V are not versatile. They do not ensure satysfying accuracy of grain size evaluation. Nuñez–Domingo formula in case of cell type structures of small grains volume variation enables N_V estimation with relative error less than 5% but for the other models this error is significantly larger,
- the point sampled intercepts method makes it possible to determine correctly the volume–weighted mean grain volume $E_1(V)$, so it can be recommended for grain size estimation in polycrystalline materials. It is necessary, however, to apply the general equation (1) and improve procedures of automatic intercepts length measurement to make the method less labour and time consuming,
- the disector provides a correct estimation of the number of grains per unit volume of material N_V (for one phase materials – the mean grain volume $E_1(V)$). The suitability of disector method in materials science investigations depends, however, on a solution of the following methodological problems:
  - to gain unambiguous information whether the requirements of this method concerning: plane sections parallelism, precision of measurement of the distance between them, and first of all the choice of this distance, the requirements ensuring counting of all grains, can be met in opaque polycrystalline materials investigations,
  - to automate identification and grain plane sections counting on the look–up and reference plane.

  Thus, at present the dicector method has a limited application in polycrystalline materials investigations. It particularly refers to fine grain materials.





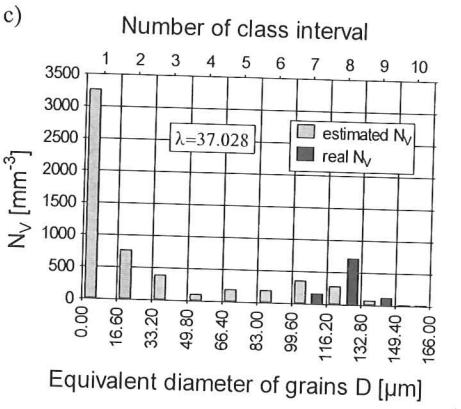
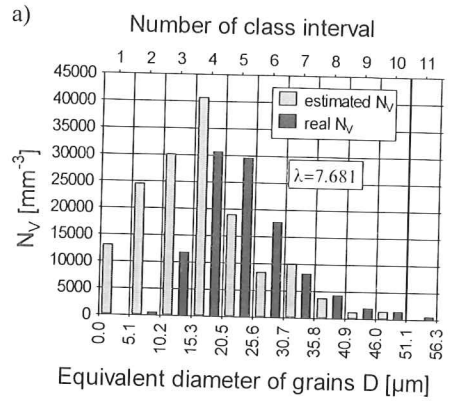
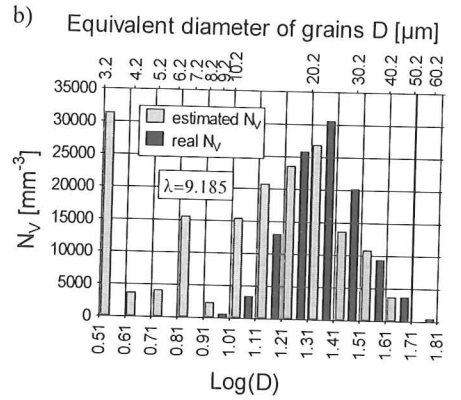


Fig. 2.Comparision of true and reproduced distributions of equivalent diameters of grains for C2 model structure: a) Scheil-Schwartz-Saltykov method, b) Saltykov method, c) Spector method.





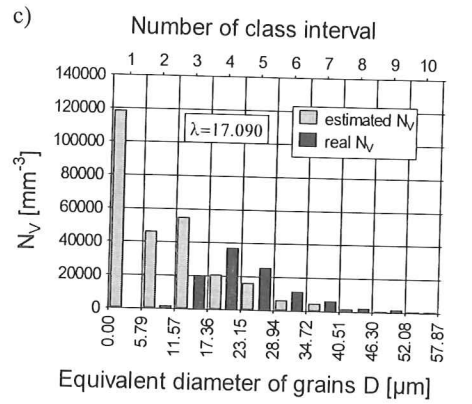


Fig. 3.Comparision of true and reproduced distributions of equivalent diameters of grains for JM2 model structure: a) Scheil-Schwartz-Saltykov method, b) Saltykov method, c) Spector method.

ACKNOWLEDGEMENTS

The investigations were sponsored by the Committee Scientific Research within the grant No. 7 T08A 029 10.

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Presented at the 7th European Congress for Stereology, Amsterdam, April 20th to 23rd, 1998.